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Crevice corrosion behavior of stainless steels and nickel-based alloy in the natural seawater – effect of crevice geometry, temperature and seawater world location

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ABSTRACT

Crevice corrosion is one of the main corrosion problems for metallic alloys used in reverse osmosis (RO) desalination plants. This type of corrosion depends on many factors, that is, alloy composition and/or metallurgy, seawater location, biofilm, temperature, service conditions and crevice geometry. Corrosivity of gulf seawater was compared with the heated Brest seawater (France) for different stainless steels and nickel-based alloy. Maintaining the same experimental conditions, similarity of crevice corrosion performance in both sites relied on the tested alloys. Both crevice corrosion initiation and propagation were evaluated and compared with previous studies. Duplex S32205 and nickel-based N06625 suffered from crevice corrosion contrary to the superaustenitic S31266. However, corrosion results of superduplex S32750 and superaustenitic S31254 were reported very randomly, confirming the "borderline" behavior of these grades in some seawater applications. The impact of the results on the RO plants materials selection is outlined.

Keywords: RO desalination; Crevice corrosion; Gulf seawater; Stainless steel; Duplex; Nickel alloy; Biofilm

1. Introduction

Desalination is a vital and strategic sector because of the shortage of potable water around the world and especially in Gulf Cooperation Council (GCC) countries. Middle East and North African (MENA) countries are expected to spend an estimated $300 billion on water and desalination projects by 2022 [1]. The worldwide trend of desalination processes could be observed in Fig. 1 where membrane based, that is, reverse osmosis (RO) process seems to be the main choice. This could be explained by the fact that thermal processes (such as Multiple Stage Flash-MSF-) reached a maturity and pose problems of energy consumption and environment [2].

Materials selection for RO process handling corrosive seawater requires a very high strength and highly Corrosion Resistant Alloys (CRAs) especially for the high pressure section. Many authors have discussed the materials selection for RO desalination plants [3–5]. Indeed, due to the high level of chlorides and oxygen, RO plants require the use of CRAs such as high alloy Stainless steels and nickel-based alloys. The development of these materials with different composition, metallurgy and variable cost had a significant impact on the materials selection in RO plants [6]. For example, UNS S31603 (316 L) is no longer used due to repeated failures in the form of crevice corrosion [7–9].

One of the major challenges for these CRAs in RO plants is the crevice corrosion resistance which is known to be critical in high pressure parts (pumps, pipes and valves)

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due to the presence of severe crevice geometries. Crevice corrosion mechanisms and factors are well studied but still reported between authors [10–13]. It is well established that crevice corrosion depends on alloy composition, seawater (chloride level, biofouling), temperature, crevice geometry (gasket nature and gasket pressure), and so on [14]. In gulf seawater, some studies of crevice corrosion have been carried out [15–18]. However, experimental test parameters are so variable that it is difficult to compare results. For example, the applied tightening torque, the crevice former material, the exposure duration, and so on, which are all impacting on corrosion results need to be fixed for proper comparison between the tested alloys. All of these factors and their impact on both initiation and propagation of crevice corrosion have been studied and modeled by many authors [19–22]. Another important factor is the seawater location around the world. This parameter is related to the chloride level, temperature profile and biological activity. It is well accepted that biofilm development affects both initiation and propagation of crevice corrosion on CRAs [23]. There has been much speculation from many authors about the effect of potential (E_{corr}) ennoblement on the initiation and propagation of pitting and crevice corrosion. Therefore, the previous results in this regard depend on the CRAs crevice corrosion resistance [23]. On the other hand, temperature and chlorination are directly related to the crevice initiation and biofilm development on the metal surfaces [24].

Except Salvago et al. [25], all references agree on the increase of cathodic reaction efficiency in the presence of biofilm. Several hypotheses have been suggested in the literature to explain this ennoblement, that is, localized changes in the interfacial pH [26], formation of "a thin organic film" on the surface [27], or production of organometallic complexes [28], enzymes [29] or extracellular substances [30].

The fundamental mechanism of crevice corrosion in presence of biofilm does not contain the scope of the present work but the purpose of this research work was to explore the impact of crevice geometry in two different seawaters. The first one is located in the Gulf at Jubail (Saudi Arabia) and the second one is heated seawater in Atlantic Ocean (Brest, France). One of the objectives was to check whether the heating of the seawater from temperate area can simulate crevice corrosion performance in tropical seawaters such as in Gulf Sea.

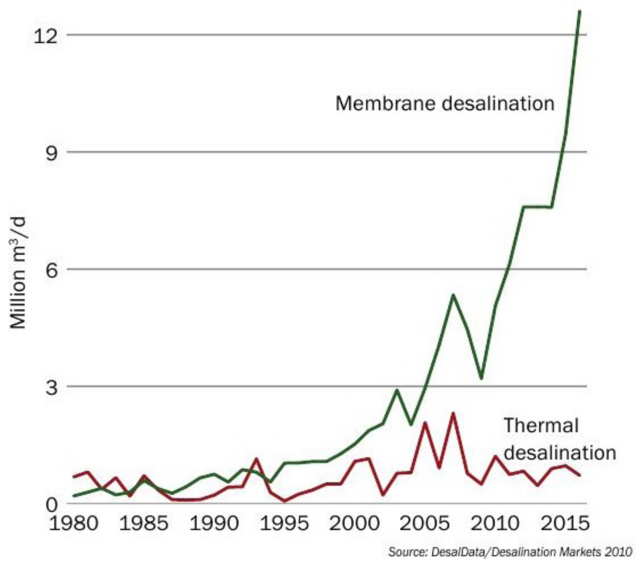

Fig. 1. Worldwide desalination processes trend. [2]

2. Experimental

2.1. Materials

The crevice corrosion resistance in seawater was determined for different stainless steels and nickel-based alloy. The chemical compositions and the Pitting Resistance Equivalent Number (PREN) of these alloys are given in Table 1.

2.2. Tested samples

To be statistically significant, five plate samples were used for each tested alloy in as-received surface finish, with average surface roughness Ra = 2–3 μm. The crevice assembly used for this study is based on the CREVCORR crevice assembly developed within a European funded project (CREVCORR) which aimed at developing and qualifying of a crevice corrosion test for stainless steels to be used in marine environments [31]. The assembly has the following characteristics: crevice formers were made of

Table 1
Composition (wt%) and PREN of the tested materials per type of crevice enhancing specimens (PREN = wt.% Cr + 3.3 wt.% Mo + 16 wt. % N)

Grade	Fe	Cr	Ni	Mo	N	W	Cu	Mn	PREN
Stainless steel									
S32205	Bal.	22.16	4.91	2.55	0.16	0.01	0.12	1.81	33.2
S32750	Bal.	24.83	6.58	3.74	0.26	1.99	0.08	0.85	41.4
S31254	Bal.	19.94	18.08	6.15	0.20	0.05	0.68	0.71	43.4
S31266	Bal.	24.12	22.61	5.59	0.46	0.01	1.61	2.97	50.0
Nickel based									
N06625	Bal.	22.50	60.20	8.10	0.02	0.01	0.05	0.08	49.4[a]

[a]PREN formula not adapted to nickel-based alloy ($PREN_{WNb}$ = wt%Cr + 1.5 (wt%Mo + wt%W + wt%Nb) + 30 wt%N).

Polyvinylidene fluoride (PVDF); all fasteners were made of titanium grade 2 and electrically isolated from the tested specimen. According to the standard "CREVCORR" testing method, the crevice former should be tightened to the test specimens with a force of about 900 N (i.e., pressure of about 3 N/mm²), which corresponds to a torque of 3 N·m with the crevice assembly that was used. For the present investigation, the CREVCORR crevice assembly was adapted to allow gasket pressures up to 20 N/mm² in order to get more severe crevice configuration which may be encountered in actual applications (e.g., at flange gaskets). Photographs of the crevice assembly are shown in Fig. 2. All the PVDF gaskets were polished with 600 grit paper to ensure a similar surface finish on crevice formers. The anode (surface under crevice formers) to cathode (surface in contact with bulk environment) ratio was 1:30 (Plate size: 100 mm × 150 mm).

2.3. Electrochemical tests

For Corrosion potential (E_{corr}) measurement, samples were connected to high impedance autonomous data logger through titanium wire mounted in a small hole located in a corner of each sample. The used reference electrode was calibrated silver-silver chlorides (Ag/AgCl). For comparison purpose with previous studies, E_{corr} values are all given versus Saturated Calomel reference Electrode (SCE).

2.4. Exposure conditions

The samples were placed in tanks where non-chlorinated seawater was continuously renewed at a rate of 60 L/h, leading to several complete seawater exchange per day (Fig. 3).

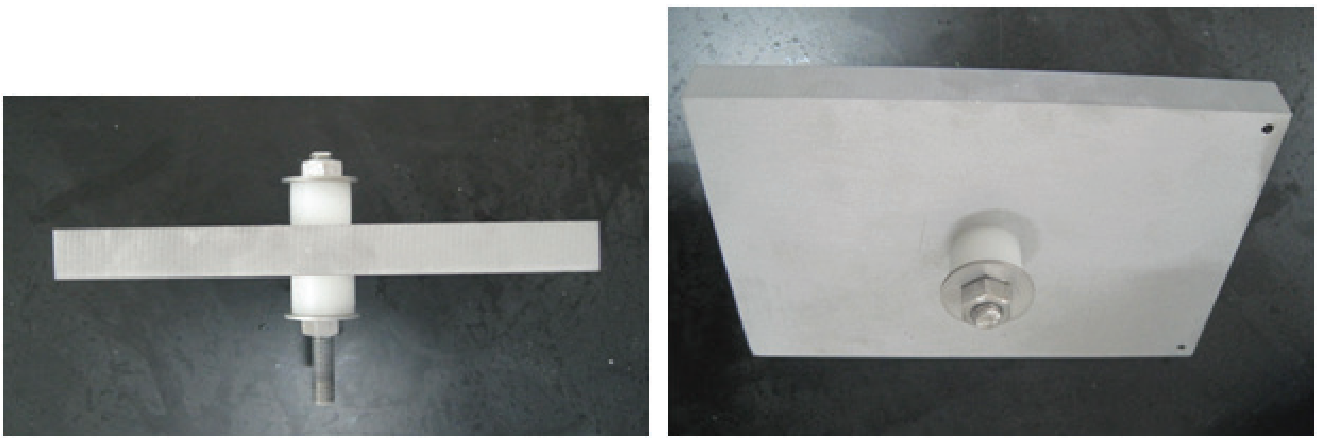

Fig. 2. Assembled crevice corrosion testing plate.

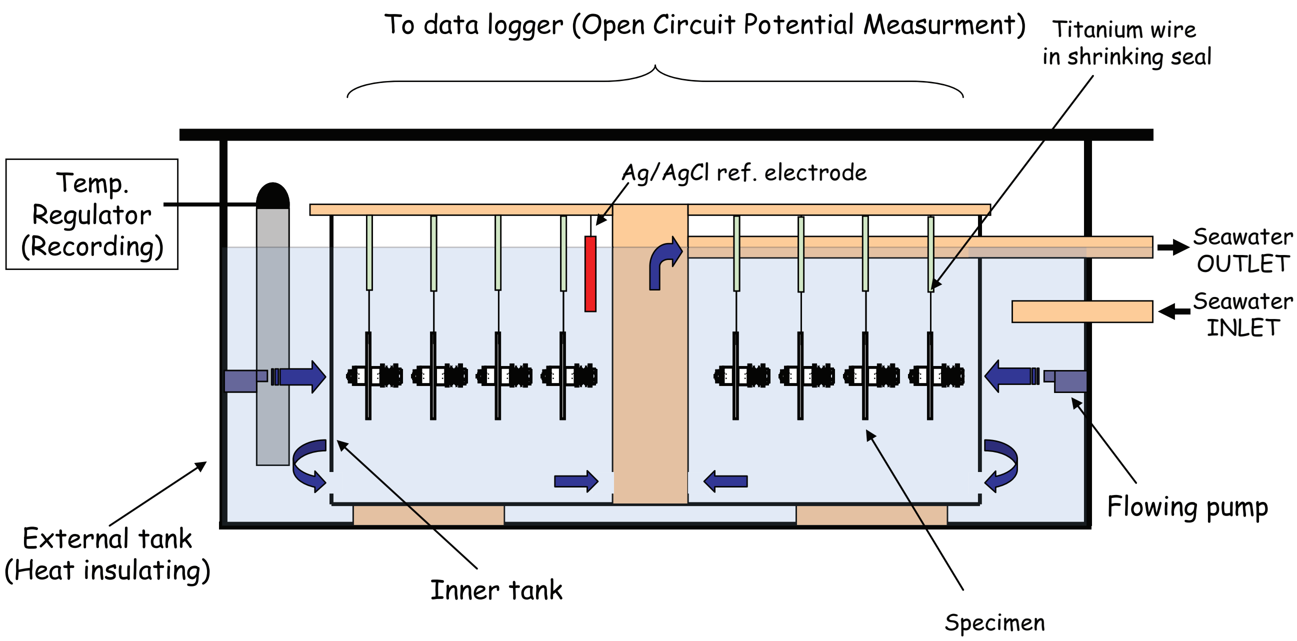

Fig. 3. Schematic description of seawater tank and test samples with heating and circulation systems.

The characteristics of natural seawater in both Brest (France) and Jubail (Saudi Arabia) are indicated in Table 2. Samples were exposed to seawaters for long duration up to 6 months covering two seasons for Jubail (Saudi Arabia) location, that is, summer and beginning of winter.

The gulf seawater temperature profile in Jubail location for 2 years (2012 and 2013) is shown in Fig. 4 with maximum temperature reaching 38°C. To simulate Jubail seawater in terms of temperature, Brest seawater was heated up to 30°C ± 1°C using regulated Titanium Heater. Submersible pumps were used to ensure homogeneous temperature in tanks without overheating which could affect the surface (biofilm) on exposed samples. For all exposure conditions, the dissolved oxygen level was at saturation level (typically 6 ppm at 30°C).

3. Results

3.1. Crevice corrosion initiation

The results of crevice corrosion testing up to 6 months in the two tested seawaters and at two gasket pressures (3 and 20 N.mm^2) are summarized in Table 3.

It appears clearly that duplex UNS32205 (2205) and nickel-based N06625 (625) were affected by crevice geometry factor in the two seawaters with much higher risk of crevice corrosion initiation at pressure of 20 N/mm^2, compared with 3 N/mm². Contrary to these alloys, superduplex (SD) S32750 and superaustenitic S31266 (B66) resisted crevice corrosion resistance in the two seawaters at both gasket pressures. For superaustenitic (SA) S31254, crevice corrosion occurred on one sample out of five, in the heated Brest seawater at the highest gasket pressure of 20 N/mm². However, no corrosion occurred on S31254 in the gulf seawater using similar configuration. It is important to consider the stochastic nature of crevice corrosion where in some cases crevice corrosion occurred randomly on specimens with sometimes only one or two occurrence out of five replicates. Therefore, it is difficult to conclude safely on the impact of crevice geometry and type of seawater for this borderline alloy showing random corrosion results between replicates. The aspect of crevice corrosion attack on duplex S32205 and nickel-based N06625 plates in crevice area is depicted in Fig. 5.

3.2. Crevice corrosion propagation

The averaged maximum crevice corrosion depths were measured on creviced plates (specimens that were free of corrosion were not considered). The obtained rates of crevice corrosion propagation on the corroded alloys at pressure gasket of 20 N/mm^2 are plotted in Fig. 6. It appears that duplex S32205 is showing the severe corrosion attack in both seawaters. Nickel-based alloy N06625 shows insignificant propagation compared with corroded stainless steels. Comparison between the two seawaters results indicates rather close values with slightly more propagation in the heated Brest seawater.

The results of metallographic examination of samples are summarized in Table 4. Non-selective corrosion was observed for the tested superaustenitic and nickel-based alloys.

For duplex S32205, the result was affected by the severity of the crevice geometry as shown in Fig. 7 (more severe propagation at the highest torque), but the mechanism of corrosion was similar in for both sites, Gulf and Brest heated

Table 2
Seawater characteristics

Parameter Location	TDS (ppm)	pH	Chlorides (ppm)	Dissolved oxygen (ppm)	Conductivity (µS/cm)
Brest	35,000	8.1 ± 0.1	19,373	6	54,000
Jubail	43,800	8.1 ± 0.1	24,090	7	62,800

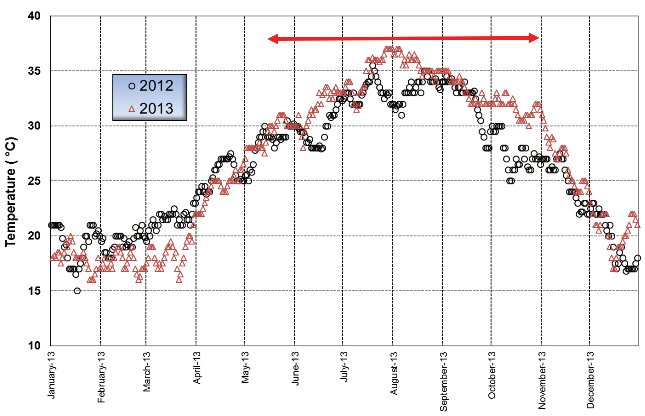

Fig. 4. Gulf seawater temperature profile (in °C) during 2012 and 2013 years.

Table 3
Crevice corrosion tests results in two different seawaters

	S32205		S32750		S31254		S31266		N06625	
Gulf seawater (28°C–36°C)	1/5	4/5	0/5	0/5	0/5	0/5	0/5	0/5	0/5	1/5
Brest heated seawater (30°C)	0/5	4/4	0/5	0/5	0/5	1/5	0/5	0/5	0/5	3/5
Gasket pressure (N/mm^2)	3	20	3	20	3	20	3	20	3	20

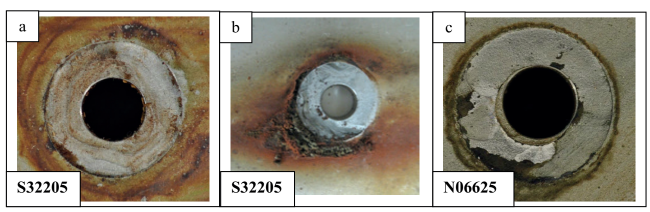

Fig. 5. Aspect of corroded S32205 and N06625 plates exposed to (a,c): heated Brest seawater up to 30°C, and (b) gulf seawater (gasket pressure 20 N/mm^2).

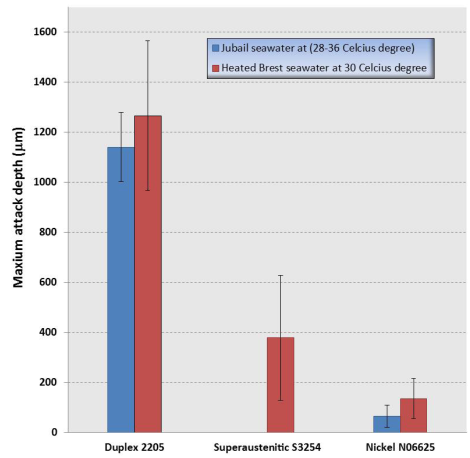

Fig. 6. Crevice corrosion propagation rates for corroded samples exposed to gulf seawater and heated Brest seawater up to 30°C (gasket pressure 20 N/mm^2).

Table 4
Metallographic inspection results of corroded crevice areas

Alloys	Microstructure	Corrosion propagation path
Stainless steel		
S32205 and S32750	Austenitic + ferritic	For slight crevice (3 N/mm^2): propagation in ferritic phase
		For severe crevice (20 N/mm^2): propagation in austenitic phase
S31254 and S31266	Austenitic	Non selective corrosion
Nickel based		
N06625	Austenitic	Non selective corrosion

seawater. An initial/superficial corrosion is observed in α-ferritic phase (Fig. 7(a)) followed by deep corrosion where the propagation was on the γ-austenitic phase (Fig. 7(b)). The Preferential attack of either α-phase or -γ-phase is due to partitioning of the alloy elements between the two phases [32].

3.3. Electrochemical measurements

The crevice corrosion behavior of the studied alloys was examined through free corrosion potential (E_{corr}) monitoring for 6 months as shown in Figs. 8 and 9. E_{corr} curves examination allowed to monitor the phenomena occurring at the surface of the studied alloys (biofilm development and crevice corrosion initiation and propagation). It is well accepted that the positive shift of E_{corr} for the studied alloys indicates the marine biofilm development on the surface of passive alloys, corresponding to the so-called biofilm ennoblement. The biofilm development on the tested alloys surface was carried out by SEM but it is considered for another scope of study. Based on the results, biofilm development occurred in both seawaters while the maximum temperature was slightly different. The biofilm ennoblement was clearly observed with averaged stabilized potentials in the range of +300 ± 20 mV/SCE. A first potential increase step was globally observed after few days of exposure to reach about +250 mV/SCE, before increasing again (second step) to reach stabilized values above +300 mV/SCE.

It should be mentioned that the quantification and identification of the developed biofilm does not comprise the scope of this work. However, what could be observed is the kinetic of biofilm formation which is slightly faster in gulf seawater (within 1–3 d) compared with the heated Brest seawater (within 2–4 d; Fig. 10).

On the other hand, the crevice corrosion initiation induces a modification of the E_{corr} which is a mixed potential between cathodic and anodic processes. This is clearly

(a) (b)

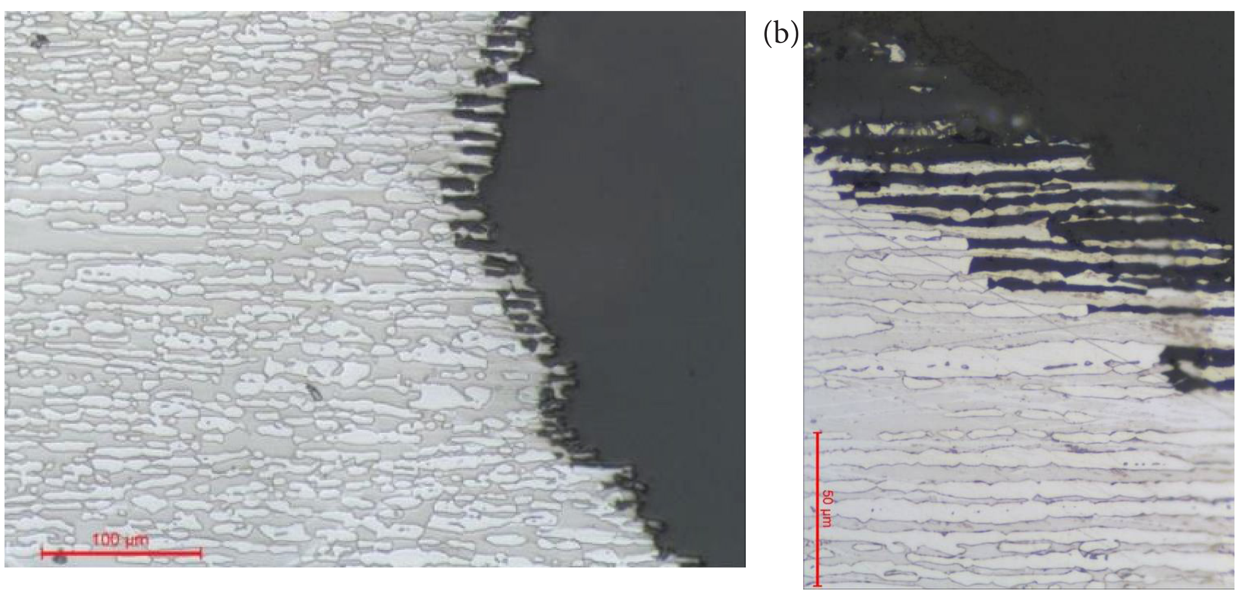

Fig. 7. Metallography of attacked S32205 samples in (a) heated Brest seawater up to 30°C, and (b) gulf seawater (gasket pressure 20 N/mm^2).

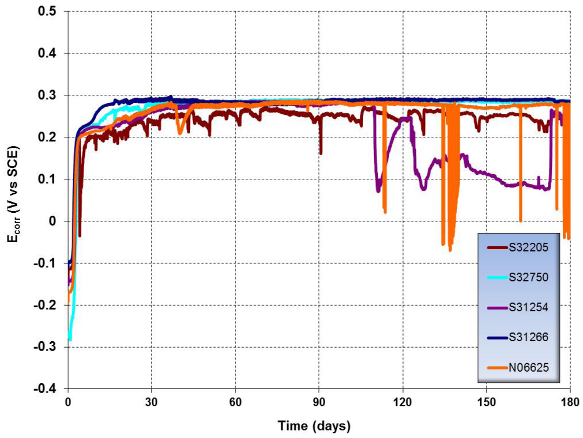

Fig. 8. Corrosion potential (E_{corr}) of different stainless steel and nickel based alloys in the heated Brest seawater up to 30°C (gasket pressure 20 N/mm²).

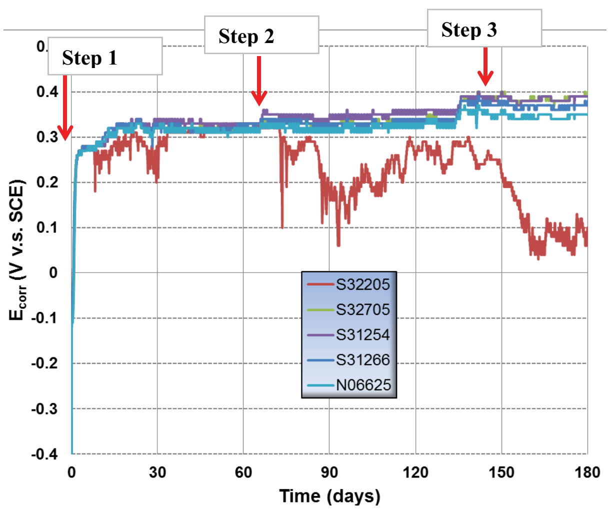

Fig. 9. Corrosion potential of different stainless steel and nickel based alloys in the gulf seawater (gasket pressure 20 N/mm²).

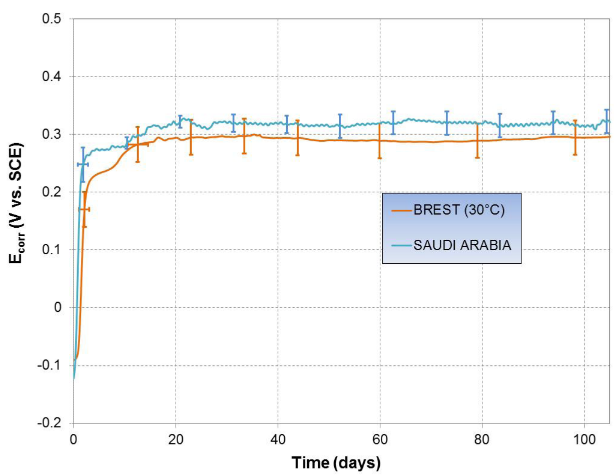

Fig. 10. E_{corr} comparison between heated Brest seawater up to 30°C and ambient gulf seawater for different stainless steel and nickel based alloys (gasket pressure 20 N/mm²).

observed in the case of corroded duplex S32205 in the gulf seawater. The crevice corrosion initiation could be observed when E_{corr} starts to drop in the negative direction in the first 20 d of immersion, before repassivation and re-depassivation around 60 d. For the nickel-based alloy N06625, potential drops indicating corrosion initiation/repassivation are also clearly observed.

4. Discussion

The obtained results can be discussed in terms of crevice corrosion risk for each tested alloy. The duplex S32205 was confirmed to be highly susceptible to localized corrosion with severe crevice configurations (i.e., high gasket pressure). Combination of biofouling, temperature and crevice geometry factors increases the likelihood of crevice corrosion for this alloy. Many authors obtained the same behavior either in less or high severe crevice geometry in natural gulf seawater for long-term exposures [33,34]. A practical case was recently recorded in pump handling gulf seawater where crevice corrosion occurred between bolt and nut made of duplex S31803 [35]. The authors observed a preferential attack on ferrite phase on the nut. Larché et al. [24] also showed the poor crevice corrosion resistance of this material in natural seawater (Brest, France) at two different temperatures, that is, 20°C and 30°C.

Corrosion resistance of superduplex S32750 could be considered to be very close to that of superaustenitic S31254 due to their close PREN number. However, in the present work, this similarity was observed only in the gulf seawater. In recent work, Larché et al. [24], observed this similarity where both alloys failed in crevice corrosion tests carried out at 30°C and for gasket pressure similar to us (20 N.mm²). These controversial results outline two issues:

- The random trend of crevice corrosion which applies to borderline alloys (in terms of corrosion resistance) and merits more investigation. Sharland et al. [36] in his review on the crevice corrosion modeling, reported the strong degree of randomness and irreproducibility of results. This author suggests that this could be due to certain instabilities in the dynamics of the system. At the same time, statistical models have been used to model localized corrosion initiation processes successfully, but the stochasticity of the system is still unproved. For the propagation stage, the same author claims that there is no single model that includes all phenomena to predict quantitatively and qualitatively.
- The use of a standard torque as the dominant parameter, when assembling the crevice, is not sufficient to ensure crevice similarity as concluded by Watson et al. [37].

In addition, one of the consequences of this stochasticity (which is dependent on the studied alloy) is the relationship between the crevice corrosion occurrence and PREN. Crevice corrosion occurrence does not necessarily decreases with PREN increase as observed in the results of Yakuwa et al. [33]. This can be attributed to the fact that PREN only considers the composition and does not consider very important parameters such as the metallurgy, the product form, intermetallic precipitation, and so on. Malik et al. [34]

obtained better crevice corrosion resistance performance in the natural gulf seawater for S32750 compared with S31254 when they adopted multiple crevice former. However, when these authors used flat washer as crevice former the performance was inversed.

On the other hand, a previous study carried out in gulf seawater for more than one year of immersion, less severe crevice geometry (gasket pressure of 2 N/mm^2) and surface finishing using SiC abrasive grit 600, superaustenitic S31254 failed contrary to the superduplex S32750 [33]. This finding emphasizes the importance of crevice gap geometry affected by surface finishing on the crevice corrosion results, as studied by several authors [15,38]. This factor plays an important role on the crevice gap geometry and then on the solution chemistry occluded inside responsible for depassivation.

The crevice corrosion attack obtained for nickel-based alloy N06625 in the two selected seawaters for this study is in good line with the feedback and field experience. Previous results indicate that nickel-based alloy N06625 has been found susceptible to crevice corrosion in natural and chlorinated seawater [39]. Larché et al. [24] obtained the same result with very superficial corrosion at 20°C and more significant rate of crevice corrosion at 30°C. This result is in agreement with previous studies on the impact of temperature on the crevice corrosion [40].

The excellent performance of the most recently developed superaustenitic S31266 was confirmed in the two seawaters. Larché et al. [24] obtained the same result at two seawater temperatures, that is, 20°C and 30°C. The better crevice corrosion resistance of S31266 compared with nickel-based N06625 was obtained previously by Grolleau et al. [41]. It has to be mentioned that this alloy was designed to combine beneficial influence of chromium, tungsten, molybdenum and nitrogen on the materials properties. In addition, its mechanical characteristics are quite high, equivalent to those of alloy N06625, and only inferior by 100 MPa than that of superduplex S32750.

The similarity between the results in the two seawaters means that heated seawater at 30°C from temperate area can simulate the crevice corrosion behavior of materials in tropical seawater. The role of biofouling on crevice corrosion is present in both sites (ennoblement of E_{corr}) but its quantification and impact on the propagation stage was not explored in the present study.

The chloride concentration was higher in gulf seawater compared with the heated Brest seawater, but the pitting potential dependence on the chloride concentration is more pronounced in the case of low grades austenitic stainless steels, that is, 316 L, 317 L. However, for the high alloy grades this factor is not affecting, as previously showed by Malik et al. [15].

For the temperature factor, it is well known that an increase of temperature will decrease the critical crevice corrosion potential (E_{crev}) which will initiate the crevice attack. However, the temperature range in the two seawaters was close and no significant difference was observed for the attacked alloys.

The involvement of metabolic processes activation within the developed biofilm outside the crevice (cathodic area) by heating in Brest seawater was outlined. This is supported by deeper attack observed in Brest seawater conditions which confirm that biofouling increases the rate of crevice corrosion propagation [42].

The direct practical consequence of these results is related to the materials selection in RO plants. For instance, it is confirmed that duplex S32205 should be avoided due to its very low crevice corrosion resistance in natural seawater. Superduplex and superaustenitic stainless steels such as S32750 and S31254 may be adequate alternative alloys in applications where crevice geometries are not considered as critical, for the use of these alloys in controlled service conditions. With severe crevice configuration, the Nickel-based alloy N066625 was found to be susceptible to crevice corrosion in seawater at about 30°C, and superaustenitic S31266 (in its hot rolled product form) appeared to be the most crevice CRA.

5. Conclusions

Crevice corrosion resistance was studied for hot rolled high grade stainless steels and nickel alloy in two different seawaters keeping the same experimental procedure. The following conclusions could be drawn:

- The results of the study confirmed the very low crevice corrosion resistance for duplex S32205 and the best crevice corrosion resistance for superaustenitic S31266.
- Hot rolled superaustenitic S31266 could be suggested for RO plants due to its high corrosion resistance contrary to duplex S32205.
- For crevice corrosion testing, tropical seawaters can be simulated by heating temperate seawater to tropical temperatures (i.e., 30°C).
- The crevice geometry should always be considered while carrying out crevice corrosion studies.

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