



Experimental Approach to Monitoring the Degradation Status of Pipelines Transporting Hydrocarbons

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Abstract

Improve the reliability of the gas and oil transportation process is a primary objective of the pipeline designers because it interests the safety of the goods and the people, the availability and the performance of pipelines as well as the economy of the hydrocarbon transport. Corrosion is a present phenomenon that occurs inside and outside of buried pipes, causing the pipeline to be pierced, leading to gas and oil leaks and causes consequences of the major economic losses. In this context, our study focused on the corrosion monitoring of metals used in the transport of hydrocarbons by two approaches based on electrochemical techniques. Monitoring the evolution of the corrosion potential using an elaborated reference instead of a commercial reference electrode, and by electrochemical impedance spectroscopy (EIS) coupled with the gravimetric method. The obtained results showed the efficiency of our approach for the realization of a corrosion sensor intended for the monitoring of corrosion in pipelines.

Keywords: Corrosion; Electrochemistry; Monitoring; Pipeline; Sensor

1. Introduction

Pipelines are the most practical, economical, and safest way of transporting hydrocarbons around the world. A study done by Green and Jackson (2015), comparing the safety of transporting hydrocarbons by pipelines versus rail, revealed that both ways are safe, but pipelines are considered the safest mean of hydrocarbon transportation. Pipelines can suffer from different types of defects, such as corrosion, stress corrosion cracking (SCC), fatigue cracks, dent, etc (Hussain et al., 2020; Nanninga et al., 2010; Cosham & Hopkins, 2003). These defects, if not properly managed, can result in pipeline failures, including ruptures and leaks, which could lead to extensive hazards to people and damage to assets and the environment. There are many pipeline accidents every year around the world, and three of the North American pipelines accidents in 2016 resulted in over 2,000 metric tons of oil and gas leak and spill (Xie & Tia, 2018). Recently, corrosion has become a major concern for the oil and gas industry. Moving pipelines to safer operating conditions is the primary goal of all pipeline owners.



The corrosion controls are various preventive strategies considered during the design and construction of pipelines which include, material selection, corrosion inhibitors, and protective coating.

Material selection (Craig, 2008) is fundamental when considering engineering design for corrosion mitigation. Before selecting the pipeline material, it is necessary to know as much information as possible about the operating environment and the material behavior in it. The overwhelming majority of pipelines worldwide are constructed from carbon steel due to its availability, low cost, good mechanical properties (Moosavi, 2017). Duplex stainless steel is also widely used in pipes, mainly because it exhibits a good combination of mechanical properties and high corrosion resistance (Nilsson, 1992).

Corrosion inhibitors have been used in the oil and gas industry to modify the process environment in order to limit and control corrosion processes. The corrosion inhibitors are injected into pipelines and are absorbed on the inner wall to form a passive film that acts as a protective layer to prevent direct contact with the fluid (Kartsonakis et al., 2020).

Protective coatings have been used to prevent external surface areas from being in direct contact with the corrosive environment and therefore hinder the flow of ions through the electrolyte, therefore limiting corrosion (Ammar et al., 2018). The most common types of coatings used in the oil and gas industry are: paint (Tezdogan & Demirel, 2014), Fusion-bonded epoxy, Three-Layer Polyolefin (Romano & Adhesives, 2005), etc.

Despite the massive investments in pipeline protection technologies, corrosion is never completely prevented. For example, it can be pointed out that although the protective coating has excellent corrosion resistance, it is susceptible to failure in harsh operating environments. Therefore, corrosion monitoring represents the main solution for this problem. Corrosion monitoring aims to monitor the condition of pipelines, identify the environmental variables that can influence the corrosion process, and also determine if the corrosion protection methods in place are effective or not. Corrosion monitoring includes non-electrochemical and electrochemical techniques.

Corrosion coupon is a non-electrochemical monitoring technique that involves the use of test coupons of the same pipe materials. The coupons were exposed to the corrosive environment for a given period of time and then retrieved for visual inspection, microscopy, and weight loss analyses in order to obtain corrosion information (Ameh et al., 2017).

An interesting alternative to the traditional coupon technique is the electrical resistance (ER) technique described in ASTM Standard (ASTM G96-90, 2001) and International Organization for Standardization (ISO 11844-2, 2005). Several sensors have been developed according to the principle of the electrical resistance method, and applied in different fields, inclining in the hydrometallurgical industry (Hren et al., 2021), the automotive sector (Kosec et al., 2019), in nuclear waste management (Marja-aho et al., 2018), and in other general applications (Gartner et al., 2016; Kourli et al., 2014; Huang et al., 2016). In the oil and gas industry, Xu et al. (2016) proposed a sensor to monitor corrosion of pipes in real-time via an electrical resistance. He found that the electrical resistance increases as the wall becomes thinner due to corrosion.

The electrochemical techniques, like potential measurements, Electrochemical Impedance Spectroscopy (EIS), and Linear Polarization Resistance (LPR) are also used for corrosion monitoring. These techniques have been adapted for in situ application by using probes, and can



therefore provide important information about the state of metallic objects. Several probes have been made in the last few decades. Among these, we cite:

LPR-based corrosion detection (Connolly et al., 2015) is the most commercialized method among the electrochemical probes due to its relatively simple operation and data interpretation. For most of the commercial LPR probes, the electrodes are often made of the same material instead of following a conventional electrochemical three-electrode system (Wright et al., 2019).

EIS in-situ corrosion sensor has been developed by Davis et al. (1998) to monitor corrosion of boiler tubes, pipes, and painted structures. The sensor was successful in all applications, especially in the painted structure where corrosion was detected starting from the initial stages of corrosive species ingress through the paint film and an incipient attack of the underlying metal.

Potential probes (referred to as reference electrode) are generally implemented for the purpose of monitoring the potentials of buried or immersed metallic structures (Brenna et al., 2017).

The aim of this paper is to present the results of the corrosion monitoring of different metals and to determine their suitability for use in a saline environment. Corrosion monitoring was performed by two electrochemical techniques: EIS coupled with gravimetric test and potential measurements test. During the potential measurements test, a reference electrode was made and the protectiveness of the coatings was evaluated. The results obtained help to identify the best metal in order to make a sensor for corrosion monitoring in subsea pipelines. This sensor will be made from the same metal used in the pipes.

2. Material and Methods

2.1 Chemical Composition

Three metals (API 5L X42 Carbon Steel, Cast Iron GGG40 and Lean Duplex Stainless Steel LDX 2101) were used in this study. The chemical compositions of the metals were evaluated by an optical emission spectrometer type “FOUNDRY-MASTER Pro”. The results are shown below:

Table 1: Chemical composition of API 5L X42 carbon steel.

Elements	C	Mn	P	S	Nb	Ti	V
% (by mass)	0.154	0.694	0.0236	0.0098	< 0.004	< 0.001	0.0015

Table 2: Chemical composition of cast iron GGG40

Elements	Fe	Mo	Al	Cu	Mn	V	P	Cr	Ni
% (by mass)	82.8	0.0517	< 0.0050	0.383	0.107	0.005	0.0437	0.0196	0.0517
Elements	Ti	Co	W	C	S	Pb	Nb	Si	
% (by mass)	0.0123	< 0.005	0.325	3.325	0.004	0.108	< 0.005	1.57	

Table 3: Chemical compositions of lean duplex stainless steel 2101

Elements	C	Cr	Ni	Mn	Si	Mo	P	S	Cu
% (by mass)	0.0212	16.6	10.3	1.94	0.255	2.15	0.0283	0.015	0.322

2.2 Preparation of the samples and tests solution

Two different types of samples were prepared for each metal. First, the metals were cut into four small specimens. One specimen was used in the electrochemical impedance spectroscopy



measurements and the other three were used in the potential measurements. The samples used for the EIS test were mechanically polished with different grades of emery paper, rinsed with double distilled water, cleaned with acetone, and finally dried with warm air, while the samples used for the potential measurements test were connected with copper wire, embedded in epoxy resin, leaving only the top cross-section exposed as working area, then polished and cleaned in the same way as before.

The test solution 3 wt.% NaCl was prepared from analytical grade reagents and distilled water.

2.3 Electrochemical Impedance Spectroscopy measurements

The study of this process was carried out by EIS technique combined with gravimetric method. Gravimetric tests were performed according to the ASTM Standard (ASTM G31-72, 2004). The samples were exposed to 3 wt.% NaCl solution for a period of 10 days. The samples were weighed with an electronic weighing balance before and after exposure to obtain the weight loss. During the exposure period, EIS was measured daily. EIS measurements were performed in a conventional three-electrode cell, including a working electrode (API 5L X42, GGG40, LDX 2101), an Ag/AgCl in 3.5 M KCl as a reference electrode, and a platinum plate as a counter electrode. The three electrodes were connected to a Potentiostat/Galvanostat model VersaStat 3 controlled by a computer using VersaStudio software. EIS scans were made from 100 KHz to 10 mHz with 10 points per decade and an AC amplitude of 10 mV.

2.4 Potential measurements

The corrosion monitoring of metals can be performed by obtaining the potential of the metal with the use of Ag/AgCl 3.5 M KCl reference electrode or any other suitable reference electrode. In our case the reference is not a commercial electrode but it was made from the same metal as those used as a working electrode.

2.4.1 Manufacturing of the reference electrodes

Two different reference electrodes have been manufactured. The first reference (RE₁) was made using the surface of the embedded sample which has been divided into two identical parts: one part of the metal is exposed to the corrosive environment and corrodes, while the other is protected against corrosion by a paint coating, and serves as a reference. This kind of reference has been used in a previous study by Li et al. (2019). The second reference electrode (RE₂) is the embedded sample whose work area has been completely covered with an appropriate coating in order to protect the metal surface from the corrosive environment. Different coatings have been applied to the metal, including an epoxy resin, a transparent barrier, and paint. It should be noted that the work of Huang et al. (2016) motivated us to develop this kind of reference electrode.

2.4.2 Measurements

The potential measurements were carried out in a typical two-electrode cell, taking the uncovered metals as a working electrode and the covered metals as a reference electrode. The two electrodes were connected to a Potentiostat/Galvanostat in order to perform the test. The tests were carried out in a 3 wt.% NaCl solution at room temperature.

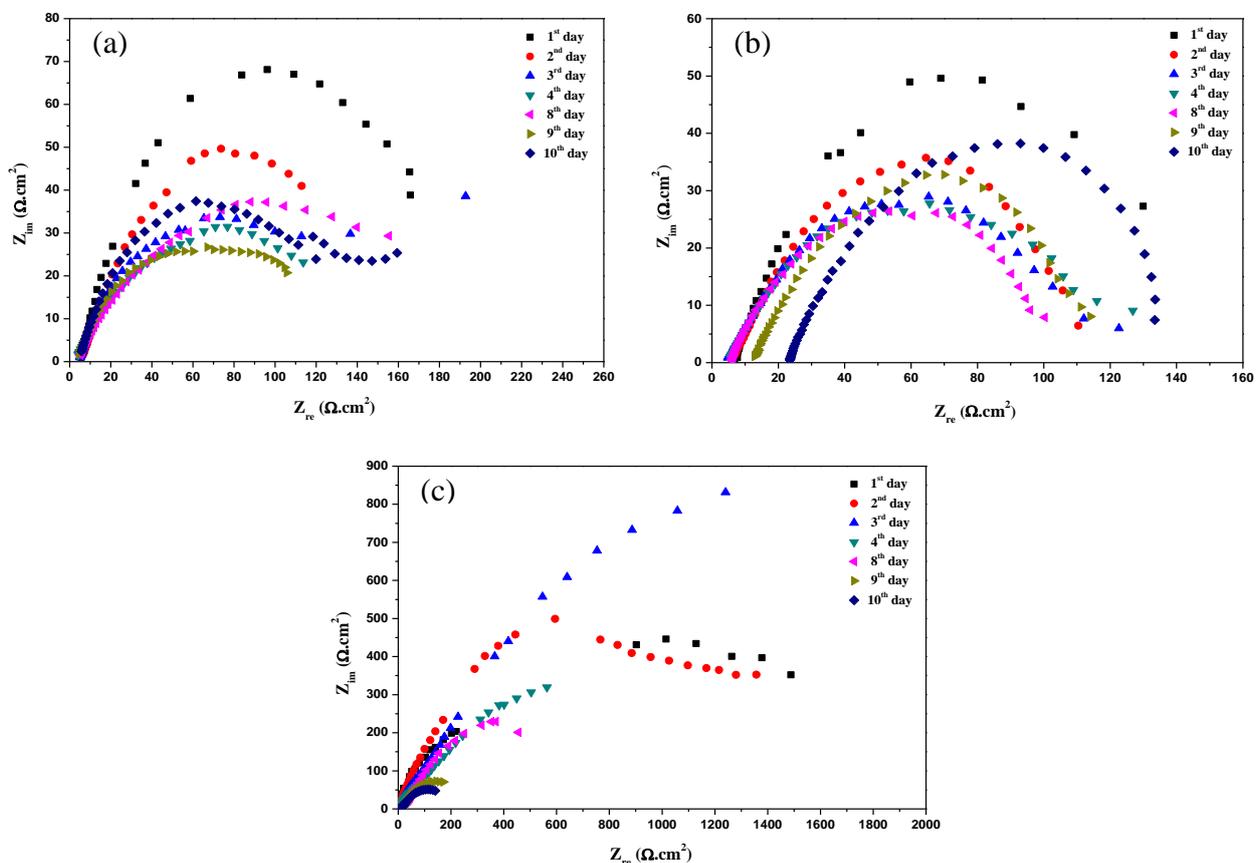


3. Results and Discussion

3.1 Electrochemical Impedance Spectroscopy and weight loss measurement

The main objective of our study is to manufacture a sensor intended to monitor the corrosion of pipes in a real marine environment. The sensor will be made from the same metal as those used as structural metal in the pipes. In this context, a study of the electrochemical impedance spectroscopy coupled with the gravimetric test was carried out on three different metals coming from the pipes. The impedance diagrams are given in the Nyquist representation. Fig. 1(a), 1(b) and, 1(c) show the electrochemical impedance spectra of carbon steel, cast iron, and duplex stainless steel respectively in 3 wt.% NaCl solution at room temperature as a function of immersion time. According to the shape of the electrochemical impedance spectra obtained for the three metals, a single semicircle was observed, indicating the presence of the charge transfer phenomenon. The diameter of the semicircle represented the charge-transfer resistance. This charge-transfer resistance has been calculated from the difference in impedance at high and low frequencies on the real axis. Electrochemical impedance spectroscopy has often been used to gain insight into the electric characteristics of the surface oxide layer, such as resistive and capacitive behavior (Pan et al., 1998). In fig. 1(a), it can be noted that the capacitive effect depends on the immersion time since the imaginary part of the curves decreased with the increase of immersion time. As well, the charge-transfer resistance decreased over time. The reason is that the aggressive species (such as Cl⁻) has been penetrated to the metal surface and induced corrosion. As shown in fig. 1(b), that after four

Figure 1: Impedance diagram in Nyquist representation of (a) carbon steel, (b) cast iron and (c) duplex stainless steel as a function of the immersion time



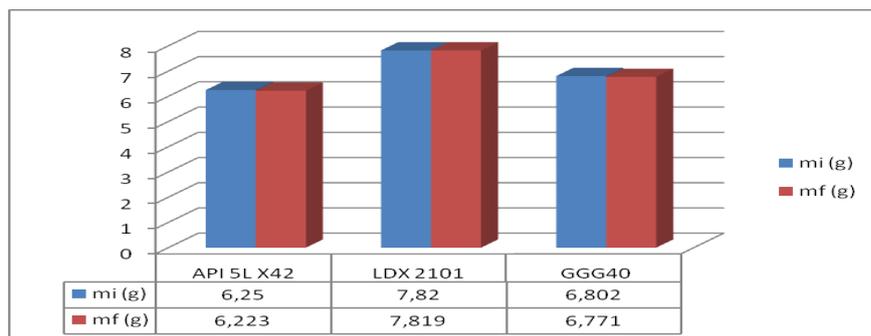


days of exposure, the solution resistance increases. This increase can be explained by the fact that the solution has been changed. For example, the solution has become less conductive. It should be noticed that the solution resistance was obtained by crossing at high frequencies of the semicircle with the real axis. As shown in fig. 1(c), that in the first stage of immersion, a huge charge-transfer resistance can be seen in the impedance plane, which explains the difficulty of seeing a complete semicircle. By the third day, it has been found that the charge-transfer resistance decreased with the increase of immersion time, due to chloride ions that were gradually penetrated to the metal surface and interacted with the oxide film, leading to the dissolution of oxide film and the corrosion of duplex stainless steel.

Comparing the charge-transfer resistance of the three metals seems to indicate that the charge-transfer resistance of duplex stainless steel is higher than those of carbon steel and cast iron, revealing that the surface film of duplex stainless steel presents higher resistance to the charge transfer process.

The mass loss obtained after immersing carbon steel, duplex stainless steel, and cast iron in 3 wt.% NaCl solution over a period of 10 days is shown in fig. 2. As can be observed, the cast iron sample showed a weight loss around 31 mg followed by carbon steel sample 27 mg, while the lowest mass loss around 1 mg corresponded to the duplex stainless steel sample.

Figure 2: Weight loss of carbon steel, duplex stainless steel, and cast iron after immersion in 3 wt.% NaCl for a period of 10 days



So the results of weight loss measurements after 10 days of exposure are thus in good agreement line with that obtained with the EIS measurements.

3.2 Potential measurements

In this part, different reference electrodes were manufactured and used in a potential measurements test in order to select the most suitable reference electrode and to evaluate the level of corrosion protection provided by the coating.

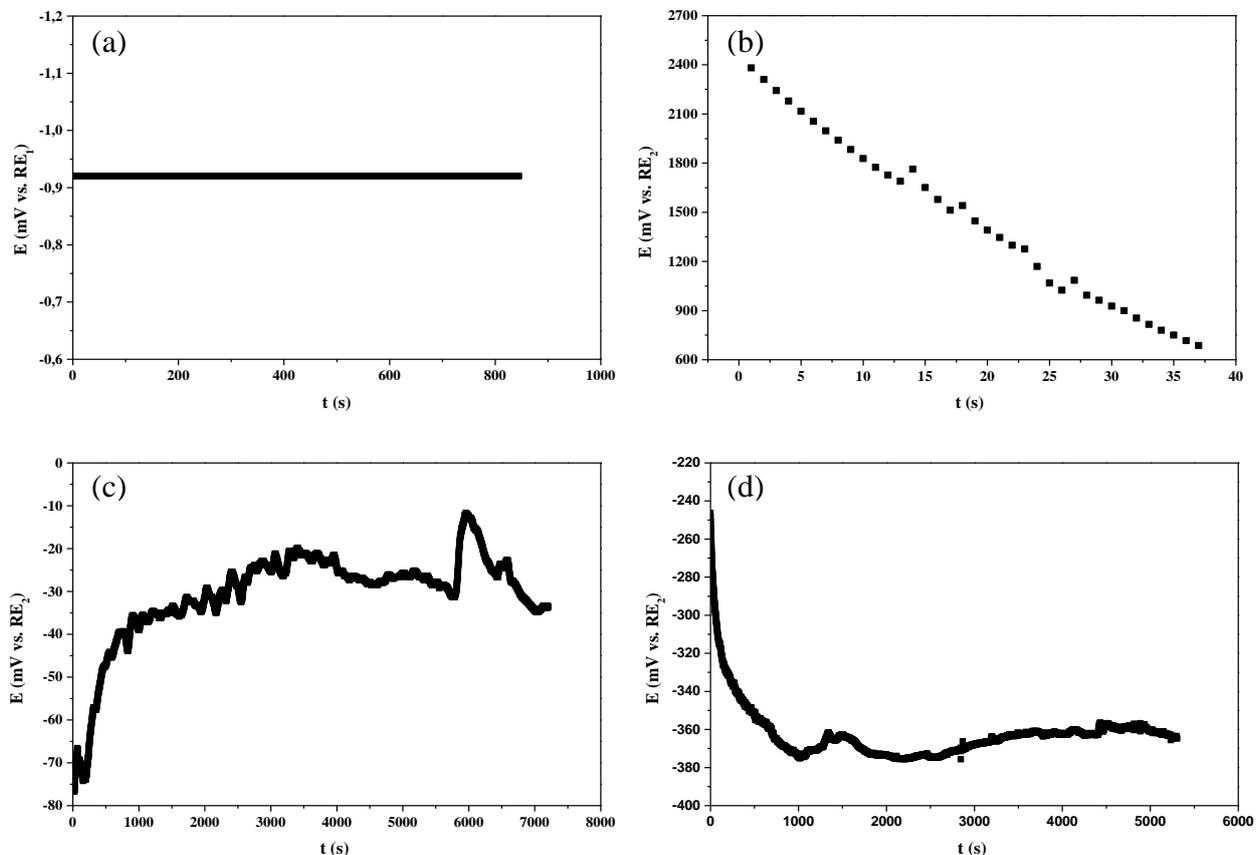
The potential measurements test can be considered as the most effective method to monitor corrosion activities in pipelines. Oki et al. (2015) conducted research indicating that corrosion monitoring of external pipelines using potential measurements is really necessary and should be performed regularly at least once in a year in order to avoid catastrophic consequences. Performing a potential measurement test requires a reference electrode (RE).

The reference electrodes used in this study were carbon steel protected by coatings against corrosion. The potential measurements were carried out in 3 wt.% NaCl solution at room temperature. The results are presented in fig. 3. Fig. 3(a) presents the evolution of the corrosion potential of the uncovered part of carbon steel with respect to the covered part



(referred to as RE₁) over time. It can be seen that the potential remained stable at approximately -0.92 mV vs. RE₁ during the test. The reason is that the reference part does not have the ability to follow the change in potential of the exposed part and, therefore, it cannot be used as a reference electrode. Fig. 3(b), 3(c), and 3(d) present the evolution of the corrosion potential of carbon steel with respect to carbon steel entirely covered with epoxy resin, transparent barrier, and paint respectively (referred to as RE₂) as a function of time. In fig. 3(b), it can be observed that the potential decreases throughout the exposure period. The corrosion potential values of the carbon steel estimated by the manufactured reference were unrealistic, due to the thick coating presented into the metal surface. The thickening of the coating does not allow the exchange of charge between the electrode-electrolyte interface and therefore, this reference cannot be used. From fig. 3(c), it can be seen that at the beginning of the exposure, the potential gradually increased, and after approximately 1040 s a value of nearly -35 mV vs. RE₂ was reached. The potential then remained almost constant and after 1800 s, there was a destabilization. This is due to the coating failure areas, where the carbon steel is in direct contact with the solution, and this will lead to corrosion of the reference electrode. In this case, the reference electrode will give an erroneous estimate of the corrosion potential value. Therefore, it can be deduced that the transparent coating was a bad choice to produce the reference electrode, since the coating applied on the surface was not able to provide protection from the corrosive environment for a long period of time. From fig. 3(d) it can be observed, that the initial corrosion potential of the carbon steel was

Figure 3: Corrosion potential evolution of the carbon steel electrode with respect to the carbon steel electrode covered (a) partially with paint, entirely with (b) epoxy resin, (c) transparent barrier, and (d) paint over time

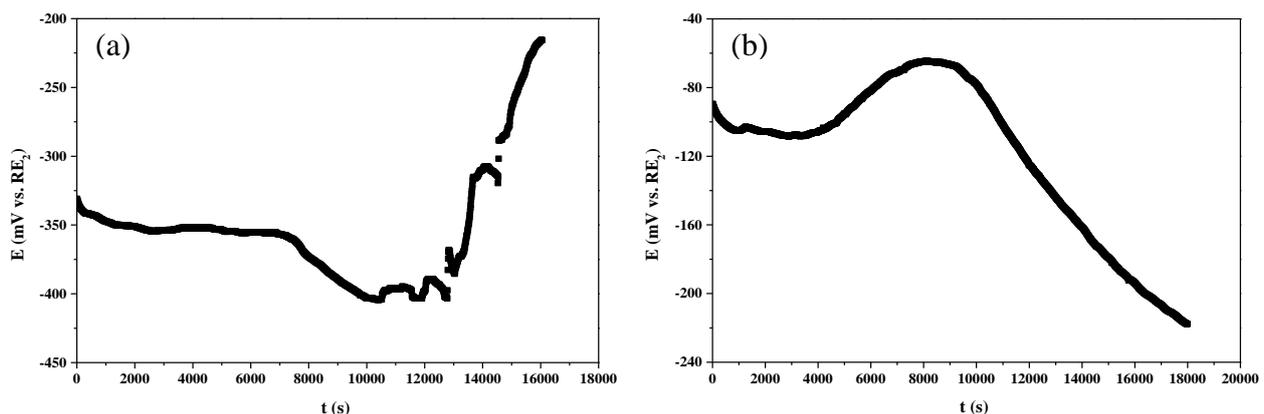




approximately -246 mV vs. RE₂, and then it gradually decreased to reach over 1000 s a steady value at nearly -374 mV vs. RE₂. The potential remained stable until the end of the test. This result might be explained by the fact that the coating was intact and there was no misalignment, or damage indicating that the paint coating applied over the carbon steel provides sufficient protection even under aggressive exposure conditions (Lyon et al., 2017). In this case, it can be concluded that the corrosion data collected by the manufactured reference are reliable and effective.

Based on the last result, it has been decided to apply paint coating to cast iron and duplex stainless steel samples, and to test them in a potential measurements test to assess their effectiveness. The potential measurements were carried out in a 3 wt.% NaCl solution at room temperature. The results are shown in fig. 4. Fig. 4(a) shows the evolution of the corrosion potential of cast iron with respect to painted cast iron over time. It can be noted that at the beginning of the exposure, the potential took a stable state, due to the presence of a protective film that isolates the metal from the aggressive solution. After 7500 s, the potential decreased. This decrease was explained by the penetration of corrosive species such as oxygen and chloride through the protective film and then corrosion of cast iron. Thereafter, the potential gradually increased towards more positive values. This was explained by the formation of a layer of corrosion products covering the whole surface of cast iron. Fig. 4(b) shows the evolution of the corrosion potential of duplex stainless steel with respect to painted duplex stainless steel over time. According to the shape, it can be seen that in the first moments following the immersion of the sample, the potential of the duplex stainless steel took a value close to -90 mV vs. RE₂. The potential remained stable during a period; this is due to the formation of a Cr rich, passive oxide film into the duplex stainless steel surface which is highly resistant to the corrosive environment. After 5000 s, the potential increased slightly due to passivation, and then decreased sharply. This decrease indicates the rupture of the passive film, which made it possible for corrosion to occur.

Figure 4: Evolution of potential difference between the uncoated and coated (a) cast iron, and (b) duplex stainless steel electrode with paint over time



As can be observed from fig. 4(a) and 4(b), the potential has not reached a stable state despite long exposure. The stability of the potential is very important and it is one of the main requirements for a satisfactory reference electrode. It has been reported by Szabo and Bakos (2010), that a good reference electrode should be easy to prepare, reach the equilibrium state fast, and keep it firm, it should be non-polarizable and exhibit environmentally friendly properties. According to these proprieties, the carbon steel electrode has been chosen as the most suitable reference electrode.



4. Conclusion

The conclusions drawn from the results of the experiments are as follows:

- The results obtained from electrochemical impedance spectroscopy, gravimetric, and potential tests allowed us to select the most suitable metal for the realization of a sensor intended for corrosion monitoring in subsea pipelines. Despite the excellent corrosion resistance of duplex stainless steel, they have not given feasible results in the potential test. So the best choice was going for carbon steel.
- The main parameters adopted from the potential difference test were the influence of the metal surface, the choice of the type of coating used, and its thickness. The coating should be very thin, expected to provide corrosion protection and ensure the transfer of charges at the interfaces generated by the contact between the electrode metal and the electrolyte. The paint coating has provided the best protection against the saline environment.
- Potential measurements test can be considered a reliable tool for corrosion monitoring in pipelines. It should be performed regularly in order to avoid catastrophic consequences.

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