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The Uppermost Corrosive Composites in Petroleum Oils and Metallic Corrosion

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ABSTRACT

Petroleum oils are the essential compounds in the purposes of industrial mechanisms and a few of other applications since obtaining some adverse impacts from such crude oils on the commonly applicable materials because of the corrosive composites of such crude oils. The investigations of the corrosion rates of seven different types of ferrous metal that applicable in the industry of crude oils refining due to the effects of salts, organic acids, elemental sulfur and Mercaptans of two different types of crude oils were the objectives of the existing research. The chemical compositions of selected ferrous metals, elemental sulfur contents, salt contents, Mercaptans contents and organic acid contents of two different types of crude oils were tested by in order of XRF detector, XRF analyzer, salt analyzer, and titration methods. A bath of prepared seven different types of metal coupons was dipped in both crude oils samples separately and homogeneously. In order of after 15, 30, and 45 days from the immersion the corrosion rates of such metal coupons were determined by the weight loss method with the aid of the qualitative microscopic analysis of the corrosion. According to the results that there were obtained the lower corrosion rates from stainless steels especially with the chemical composition at least 12% of chromium with sufficient amount of nickel because of the self corrosion protection film of such metals, relatively higher impact from salts on the metallic corrosion, formations of FeS, Fe_2O_3, corrosion cracks and irregular pitting on the metal surfaces.

Keywords: *Petroleum oils, Corrosive properties, Ferrous metals, Weight loss, Decay, Corrosion*

1. INTRODUCTION

Metallic destruction is a natural phenomenon that caused through either electrochemical or chemical reaction during the interaction between the metal and the corrosive aided environment. Usually, the metal needs to react either with some sort of strong oxidizing agent that stronger than Fe^{2+} or any other environment which is composed of water and oxygen (Khana et al., 2009). When considering the special case of regarding the corrosion of ferrous metals normally it is a chemical oxidizing process with the formation of metal sulfides, oxides or hydroxides on the metallic surface (Calister et al., 2003). The petroleum oils are the mixture of hydrocarbons in large amounts and trace amounts of organic acids, salts, and sulfur which is more prone in the augmentation of the rates of above metal destruction processes (Alsahhafet al., 2010). According to the metal destruction process, types of compounds and the conditions of environment the metal destruction can be subdivided into various categories such as the galvanic corrosion, general corrosion or rust, pitting corrosion and stress corrosion (Bolton et al., 1994). Based on the investigations of the impact of corrosive compounds in petroleum oils, identification of the types of corrosion and the testing of the stability of the metals against the corrosion there were implemented some important research works, literature reviews and case studies under different categories and discovered valuable information related with the study field of chemical and petroleum engineering (Okpokwasili and Oparaodu, 2014).

In this research there were desiderated to speculate the impact of the elemental sulfur, active sulfur, organic acids and salts which are composed in two different types of petroleum oils separately on the destruction of seven different types of ferrous metals which are frequently used in the petroleum refining industry due to the corrosion and to investigate the important changes of the metallic surfaces due to the corrosion with important observations for the further analysis.

2. MATERIALS AND METHODS

2.1 Materials

As the petroleum oils samples to test the corrosion rates of metals two different types of petroleum oils were selected. Those are Murban and Das Blend and quietly different in their chemical compositions and also Das Blend is known as a "sour" petroleum oil because of its higher sulfur content which is known as a corrosive compound as discussed under the introduction although Murban is accustomed to petroleum oil which is used in most of the petroleum refineries in the world (Davis and Davis, 2003).

There were selected seven different types of ferrous metals including three types of carbon steels, three types of stainless steels and Monel metal which are having a vast range of applications in the industry of petroleum refining including some major tasks under both volatile and non-volatile environments as given in Table 1.

Table 1. Application of the Selected Metals in the Petroleum Refining Industry

Metal	Applications
Carbon Steel (High)	Transportation tubes
Carbon Steel (Medium)	Storage Tanks
Carbon Steel (Mild Steel)	Storage Tanks
410-MN:1.8 420-MN: 2.8 (Stainless Steel)	Heat exchangers
410-MN:1.7 420-MN: 1.7 (Stainless Steel)	Crude distillation columns
321-MN:1.4 304-MN:1.9 (Stainless Steel)	Crude distillation columns
Monel 400	Preheaters

2.2 Methodology

The chemical compositions of the selected seven types of metals were tested by the XRF detector. This is an instrument that works based on the principles of X-ray diffraction and according to them, there can be detected the elemental compositions of some materials including all of the metals and most of the nonmetals excluding carbon as a percentage.

The elemental sulfur contents, Mercaptans contents, organic acid contents and salt contents of both Murban and Das Blend petroleum oils by the standard methods and instruments as summarized in Table 2.

Table 2. Methodologies for the analysis of the corrosive compounds in crude oils

Property	Method	Readings
Sulfur content	Directly used the crude oil samples to the XRF analyzer.	Direct reading
Acidity	Each sample was dissolved in a mixture of toluene and isopropyl and titrated with potassium hydroxide.	Endpoint
Mercaptans content	Each sample was dissolved in sodium acetate and titrated with silver nitrate.	Endpoint
Salt content	Each sample was dissolved in an organic solvent and exposed to the cell of the analyzer.	Direct reading

Batches of similar sized metal coupons were prepared as six from each type of metals altogether forty-two metals coupons from seven different types of selected metals as all were in same dimensions namely same as in the width, length and thickness according to the necessity of further calculations. The initial weights and the dimensions of well cleaned each metal coupon were measured by in order of the electronic balance, ruler and a micrometer. The prepared metal coupons have appeared as shown in Figure 1.

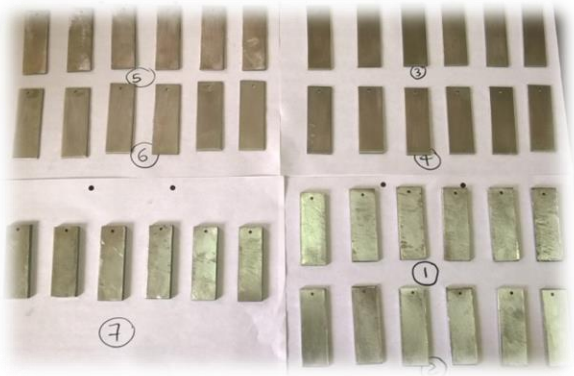

Fig.1. Prepared metal coupons

The prepared metal coupons were entirely immersed in both Murban and Das Blend petroleum oils separately according to the metal type as three metal coupons for each crude oil container as shown in Figure 2 and 3.

Fig.2. Apparatus of the experiment

Fig.3. Experimental setup

After 15 days from the immersion, a metal coupon was taken out with respect to each petroleum oil container as represented both petroleum oil and type of metal altogether fourteen metal coupons as the first batch. The corrosion rate of each metal coupon was determined by the relative weight loss of such metal coupons due to the corrosion. The mathematical expression and the terms of the weight loss method are given in below (Okpokwasili and Oparaodu, 2014).

$$\mathbf{CR} = \mathbf{W} * \mathbf{k}/(\mathbf{D} * \mathbf{A} * \mathbf{t}) \qquad (1)$$

Where;
W = weight loss due to the corrosion in grams
k = constant (22,300)
D = metal density in g/cm^3
A = area of metal piece (inch^2)
t = time (days)
CR= Corrosion rate of metal piece

The corroded surfaces of metals were cleaned by the sandpapers and isooctane while observing through the 400X lens of a laboratory optical microscope and the final weight of each metal coupon was measured by the electronic balance according to the necessity for the determinations of the corrosion rates of such metal coupons. The same procedure was repeated for another remaining two same batches of metal coupons in order to after 30 and 45 days from the immersion to determined the corrosion rates of each type of metals with respect to both Murban and Das Blend petroleum oils same as the previous batch of metal coupons and finally the average corrosion rate of each type of metal with respect each petroleum oil was calculated and interpreted.

Based on the objectives of the qualitative analysis of the corrosion compounds that formed on the surfaces of metal coupons as results of the interaction between such metals and petroleum oil the microscopic analysis component was performed with the aid of laboratory optical microscope. The corroded metal surfaces were observed through the 400X lens of that optical microscope to identify and clarify corrosion compound through the visible features of such corrosion compounds foremost the color and surface changes such as cracks.

According to the necessity of the explanation of the observed invisible weight losses of some metal coupons while determining the corrosion rate after interacting with the petroleum oils the decayed metal concentrations of each petroleum oil sample were measured by the atomic absorption spectroscopy (AAS). There were measured the decayed ferrous concentrations into petroleum oil samples which were interacted with carbon steel coupons and stainless steel coupons also the decayed copper concentrations into petroleum oil samples which were exposed to Monel metal coupons. As the methodology, 1 ml of each petroleum oil sample was diluted with 9 ml of 2-propanol and filtered well before sending the petroleum oil samples to the suction of the instrument of atomic absorption spectroscopy (AAS).

As the investigations and confirmation stage of the formation of the corrosion, the deduction of the initial hardness of each metal coupon due to the formation of the corrosion was measured by the Vicker's hardness tester. According to the working principles of such instrument, it gives the value for the hardness at some certain point on the metallic surface. Therefore, it was measured the hardness of at least three points on the metal coupons at once and the averages values were calculated as the hardness of such metal coupon. In the case of the experiment, the initial hardness and the hardness after corrosion on each metal coupon were measured. The working principles of the Vicker's hardness tester are given in the below.

Fig. 4. Indenter of the Vicker's hardness tester

$$\mathrm{HV} = 1.854 * \mathrm{P}^2/\mathrm{L}^2 \qquad (2)$$

Where;
P= Applied Load on the surface of metal
L= Diagonal length of square
HV= Hardness

3. RESULTS AND DISCUSSION

According to the observed results of the XRF detector test, the essential elements of the chemical compositions of selected metals are given in Table 3.

Table 3. Chemical compositions of selected metals

No	Metal	Fe (%)	Ni (%)	Cr (%)	Cu (%)
1	Carbon Steel (High)	98.60	0.17	0.14	0.37
2	Carbon Steel (Medium)	99.36	-	-	-
3	Carbon Steel (Mild Steel)	99.46	-	<0.07	-
4	410-MN: 1.8; 420-MN: 2.8 (Stainless Steel)	88.25	0.18	10.92	0.10
5	410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	87.44	-	11.99	-
6	321-MN:1.4; 304-MN:1.9 (Stainless Steel)	72.47	8.65	17.14	-
7	Monel 400	1.40	64.36	<0.04	33.29

The basic observations regarding the chemical compositions of selected ferrous metals that there were identified the relatively higher ferrous concentrations in carbons steels, intermediate ferrous concentrations in stainless steels and trace ferrous concentration of ferrous in Monel metal. Apart from that, the nickel and chromium concentrations were found in stainless steels and higher concentrations of nickel and copper were found in Monel metal. The stainless steel is a modification stage of the ferrous metal by doping some trace elements such as nickel and chromium in a certain amount with the seeking of the enhancements of some important mechanical and chemical properties such as the hardness, strength and the resistance of the corrosion (Singh et al., 2006).

The obtained results for the fundamental corrosive properties of both Murban and Das Blend petroleum oils are given in Table 4.

Table 4. Corrosive properties of both crude oils

Property	Murban	Das Blend
Sulfur content (Wt. %)	0.758	1.135
Salt content (ptb)	4.4	3.6
Acidity (mg KOH/g)	0.01	0.02
Mercaptans content (ppm)	25	56

Above results showed approximately some higher corrosive tendency regarding the Das Blend petroleum oil than the corrosive tendency of Murban petroleum oil because of the relatively higher amounts of elemental sulfur, Mercaptans, and the organic acids although weaker regarding the salt contents than Murban Petroleum oil.

Organic acids are some kind of foremost corrosive compounds present in petroleum oils since the occurrences of such petroleum oil also called as "naphthenic acids" which are having the general formula of "RCOOH" (Afaf et al., 2007). Acidity is a term that indicates the total amount of such acids present in some certain petroleum oil (Ahmed, Elnour, and Ibrahim, 2014). The general chemical resections of the corrosion of metals due to the organic acids are given in equation 3-5.

$$\mathrm{Fe + 2RCOOH \rightarrow Fe(RCOO)2 + H2} \quad (3)$$
$$\mathrm{FeS + 2\,RCOOH \rightarrow Fe(COOR)2 + H2S} \quad (4)$$
$$\mathrm{Fe(COOR)2 + H2S \rightarrow FeS + 2\,RCOOH} \quad (5)$$

According to the natural occurrences of petroleum oils, they may consist of $CaCl_2$, $MgCl_2$, and NaCl (Davis and Davis, 2003). The summation of such halides in some petroleum oil is known as the salt content in such petroleum oil (Luther and Rickard, 2007). During some high-temperature process such as the heating, such salts tend to be dissociated into HCl molecules even though behave as non-corrosive compounds as explained the chemical reaction in equation 6.

$$\mathrm{CaCl2 + H2O \rightarrow CaO + 2HCl} \quad (6)$$

When reducing the temperature of the system such HCl molecules reacted with the moisture and the water presence in such system and tend to form hydrochloric acids and hydrogen sulfide gas and both are considered as corrosive compounds although hydrogen sulfide has less retention time in the system because of its gassy phase (Speight et al., 1999). Therefore, the hydrochloric acid plays a dominant role in the formation of metallic corrosion as explained in equation 7-9.

$$\mathrm{HCl + Fe \rightarrow FeCl2 + H2} \quad (7)$$
$$\mathrm{H2 + S \rightarrow H2S} \quad (8)$$
$$\mathrm{FeCl2 + H2S \rightarrow FeS + 2HCl} \quad (9)$$

Sulfur is a distinguished compound that presence in petroleum oils and also causes the metallic corrosions. In the analysis of the compositions of petroleum oils, the sulfur can be identified in several forms such as elemental sulfur, Mercaptans, thiophenes, and hydrogen sulfide and most of them are identified as the corrosive compounds (Fang, Nesic and Young, 2008). The elemental sulfur tends to react with the metal directly at about 80^0C in the proper way and the process is known as the "localized corrosion". The other sulfur compounds tend to react with the metals according to the reactivity of the fractions and functional groups of them such as the Mercaptans which is having a chemical formula of "RSH" at about 230^0C also the process is known as the "sulfidation" (Davis and Davis, 2003). The general chemical reactions of both processes are given in equation 10-11.

$$\mathrm{S8(s) + 8\,H2O\,(l) \rightarrow 6\,H2S\,(aq) + 2\,H2SO4\,(aq)} \quad (10)$$
$$\mathrm{8\,Fe + S8 \rightarrow 8\,FeS} \quad (11)$$

Based on some assumptions of the proper progress of above chemical processes there can be concluded Das Blend is having some higher corrosive tendency than the corrosive tendency of Murban in the experiments. But in the actual analysis, the impact of some corrosive compound on the metallic corrosion must be discussed with the concentration of such corrosive compound and the required environmental conditions for the relevant chemical process such as mostly considered temperature.

Regarding the important analysis of the determination of the corrosion rates of metals with respect to two different types of crude oils after certain immersion periods, the obtained results have been listed in Table 5 and 6.

Table 5. Corrosion rates of metals in Murban

Metal	Corrosion Rate after 15 Days (cm^3 inch^{-1}day^{-1})	Corrosion Rate after 30 Days (cm^3 inch^{-1} day^{-1})	Corrosion Rate after 45 Days (cm^3 inch^{-1} day^{-1})	Average Corrosion Rate (cm^3 inch^{-1} day^{-1})
(1)Carbon Steel (High)	0.811971	0.466425	0.068794	0.4490632
(2)Carbon Steel (Medium)	0.817791	0.180339	0.073358	0.3571623
(3) Carbon Steel (Mild Steel)	0.10973	0.048244	0.038592	0.0655217
(4) 410-MN: 1.8 420-MN: 2.8 (Stainless Steel)	0.041784	0.016075	0.011801	0.02322
(5) 410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	0.11626	0.011968	0.007574	0.0452676
(6) 321-N:1.4 304-MN:1.9 (Stainless Steel)	0.016612	0.007453	0.005599	0.009888
(7)Monel 400	0.356263	0.034877	0.026729	0.13929

Table 6. Corrosion rates of metals in Das Blend

Metal	Corrosion Rate after 15 Days (cm^3 inch^{-1} day^{-1})	Corrosion Rate after 30 Days (cm^3 inch^{-1} day^{-1})	Corrosion Rate after 45 Days (cm^3 inch^{-1} day^{-1})	Average Corrosion Rate (cm^3 inch^{-1} day^{-1})
(1)Carbon Steel (High)	0.350249	0.224901	0.024738	0.1999627
(2)Carbon Steel (Medium)	0.481055	0.140654	0.05911	0.2269396
(3) Carbon Steel (Mild Steel)	0.162883	0.141093	0.100635	0.1348702
(4) 410-MN: 1.8 420-MN: 2.8 (Stainless Steel)	0.044146	0.034035	0.006149	0.0281102
(5) 410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	0.053701	0.034841	0.016363	0.0349681
(6) 321-MN:1.4 304-MN:1.9 (Stainless Steel)	0.022894	0.006503	0.002825	0.0107404
(7)Monel 400	0.061554	0.037655	0.016067	0.0384254

The average corrosion rates of each type of metals with respect to both petroleum oils are shown in figure 5.

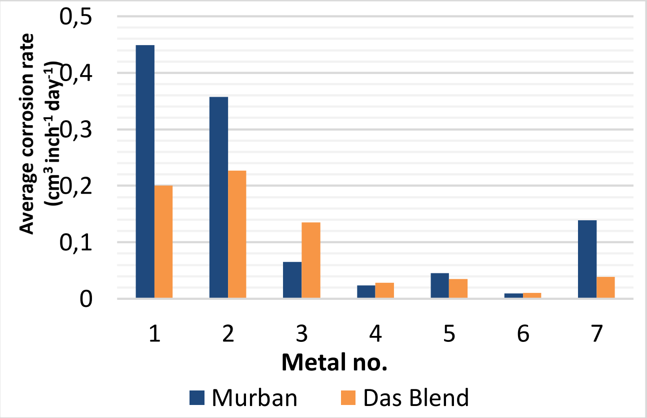

Fig.5. Average corrosion rates of metals

According to the obtained results for the average corrosion rates of metals clearly, they can be observed lower corrosion rates from stainless steels, higher corrosion rates from carbon steels and intermediate corrosion rates from Monel metal which is having a trace amount of ferrous. The least corrosion rates were obtained from 321-MN: 1.4 304-MN: 1.9 (Stainless Steel) which has the chemical composition ~18% of chromium and ~8% of nickel. Also according to the theoretical explanation of the self corrosive protection film of chromium and nickel of stainless steels the minimum requirement is ~12% of chromium and a sufficient amount of nickel. By referring the chemical composition of the 321-MN: 1.4 304- MN: 1.9 (Stainless Steel) it can be confirmed the efficiency of the corrosion protection film at higher compositions of both chromium and nickel. When comparing the other stainless steels 410-MN: 1.7 420 - MN: 1.7 (Stainless Steel) was composed ~12% of chromium although lack of nickel and showed higher corrosion rates among other stainless steels in both petroleum oils. Also 410-MN: 1.8 420 -MN: 2.8 (Stainless Steel) metal was composed with ~ 11% of chromium and ~0.2% of the nickel which was shown an intermediate corrosion rate among stainless steels. There can be concluded the necessity of both chromium and nickel on the formation of the self corrosive protections film as recommended in the theory for the high efficiency (Khana et al., 2009).

Regarding the contributions of the corrosive properties of petroleum oils on the corrosion rates of such metals, four types of metals were shown their higher corrosion rates in Murban petroleum oil since Das Blend petroleum oil has some higher corrosive strength according to the theoretical explanation and the presence of corrosive compounds in that petroleum oil. By considering the possible obtained results and the limitations of the corrosion reaction processes there can be concluded the improper progress of both "sulfidation" and "localized corrosion" processes regarding the sulfur and active sulfur compounds at the room temperature and also the progress of the corrosion process due to the salts is stronger than the progress of the corrosion process due to the organic acids or naphthenic acids on the destructions of the metals at the room temperatures (Speight et al., 1999).

The variations of the corrosion rates of metals in both Murban Das Blend petroleum oils with the exposure time period are shown in figure 6 and 7.

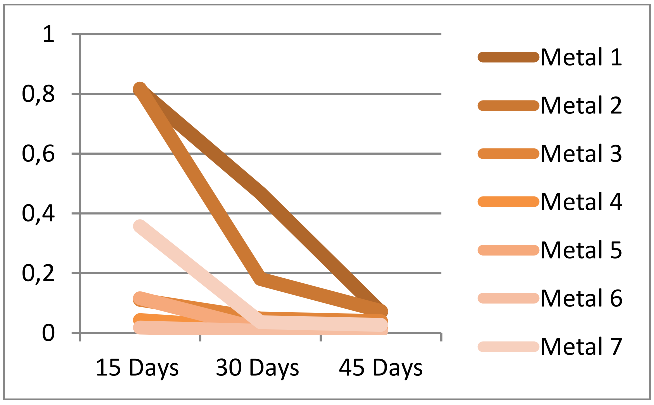

Fig. 6. Variations of the corrosion rates of metals in Murban

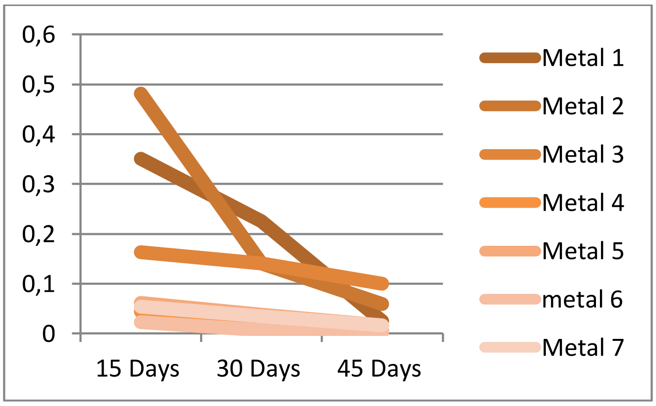

Fig. 7. Variations of the corrosion rates of metals in Das Blend

By observing the variations of the corrosion rates of metals in both Murban and Das Blend petroleum oils there can be emphasized the similar variations regarding each type of metals with respect to both petroleum oils. There were observed the reductions of the corrosion rates with the exposure time period and it can be confirmed the inversely proportional relationship between the corrosion rate and the exposure time which was discussed in the weight loss method forever.

According to the microscopic analysis, the observed features have been shown and highlighted in figure 8.

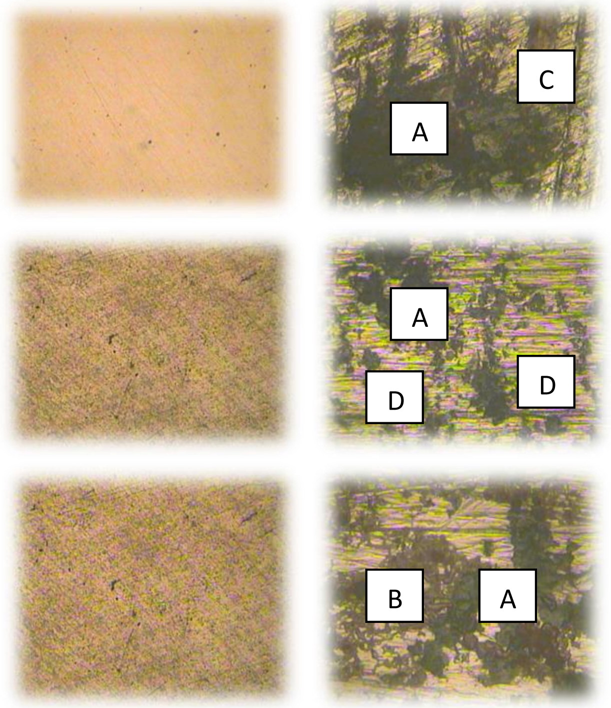

Fig. 8. Corroded metal surfaces

The microscopic analysis and identifications were based on the visible features of such compounds including the color and surface changes. The observations and the distinguishing features from the microscopic analysis have been shortlisted below (Singh et al., 2006).

- A- Black color compounds
- B- Reddish black color compounds
- C- Cavities
- D- Cracks and irregular pits

A brief description of such observations and the theoretical explanations of such observations have been given in Table 7 (Luther and Rickard, 2007).

Table 7. Visible appearances of coroison compounds

Compound	Appearances	Observations
FeS	Black, brownish black, the property of powder, pitting, cracks	Observed most of the features in each metal piece.
Fe_2O_3	Rusty color	Observed rarely.
CuS	Dark indigo/ dark blue, the property of the powder	Unable to specify.

According to the observations of the microscopic analysis, there can be concluded the formations of the compound of FeS in most of the observations as explained corrosive causing processes in the discussion and the formations of the corrosion cracks and pitting corrosion regarding most of the stainless steels apart from the formation of FeS. As a very special observation, it can be emphasized the formation of CuS on Monel metals although unable to specify because of the similar appearance with FeS in color and most of the appearences. Also, it can be recommended some compositional analysis method such as the X-ray diffraction (XRD) for the analysis of obtained corrosion compounds on the metal surfaces for the better analysis of the corrosion forever.

The obtained results for the analysis of the decayed metallic concentrations into both petroleum oils the ferrous concentrations in petroleum oil samples which were exposed to stainless steels and carbon steels also the decayed copper concentrations in petroleum oil samples which were exposed to Monel metal have been interpreted under the Table 8.

Table 8. Decayed metallic amounts into crude oils

Metal	Crude Oil	Fe Concentration /ppm	Cu Concentration /ppm
Carbon Steel (High)	Murban	0.47	-
	Das Blend	1.10	-
Carbon Steel (Medium)	Murban	0.54	-
	Das Blend	0.02	-
Carbon Steel (Mild Steel)	Murban	-0.08	-
	Das Blend	-0.48	-
410-MN: 1.8 420- MN: 2.8 (Stainless Steel)	Murban	-0.65	-
	Das Blend	-0.78	-
410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	Murban	-0.71	-
	Das Blend	-0.79	-
321-MN:1.4 304-MN:1.9 (Stainless Steel)	Murban	-0.44	-
	Das Blend	-0.17	-
Monel 400	Murban	-	10.47
	Das Blend	-	9.49

The concluded results of the above-obtained results the decayed ferrous and copper concentrations from metals into crude oils have been interpreted in Figure 9 and figure 10.

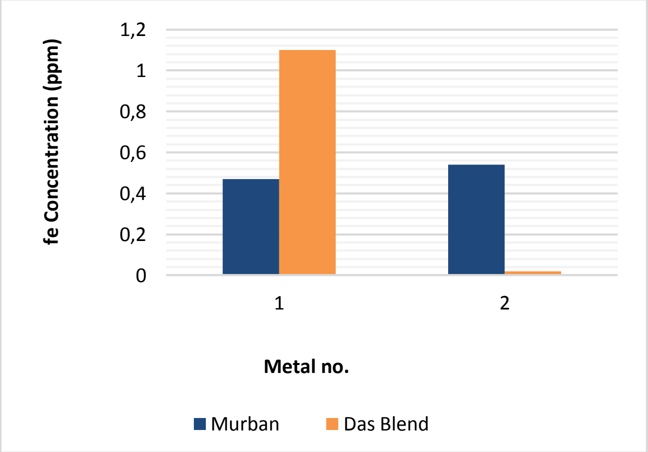

Fig. 9. Decayed ferrous concentrations from metals into crude oils

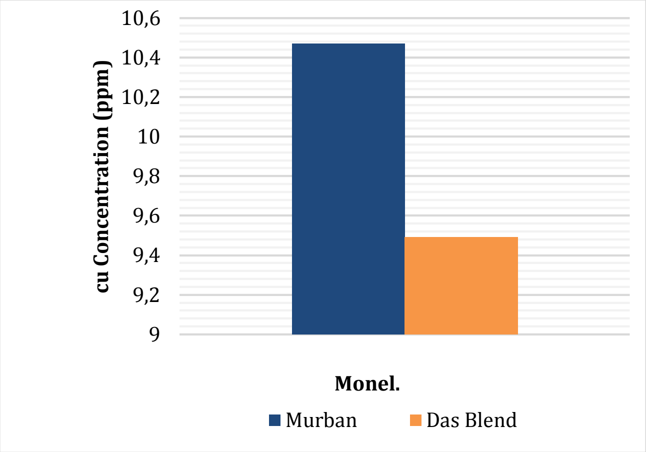

Fig. 10. Decayed copper concentrations from metals into crude oils

The obtained results showed some relatively higher decay of ferrous from carbon steel (high) and carbon steel (medium) also found the highest corrosion rates from these two metals in both petroleum oils. The special observation that it couldn't find any ferrous concentration in any petroleum oil sample related to any type of stainless steel also found least corrosion rates from stainless steels. Some higher decayed copper concentrations were found in petroleum oil samples related to Monel metal. After the formation of corrosive compounds on the metal surfaces such compounds and to be removed from the initial surfaces suddenly due to the attractive forces and repulsive forces between successive electrons and the nucleus of such atoms (Calister et al., 2003). The observations confirmed the formations of the corrosion and explained the reason for the invisible weight loss of metal coupons during the formation of corrosion.

The deductions of the initial hardness of the metal coupons after interacted with both Murban and Das Blend petroleum oils have been interpreted in graphs of figure 11 and 12.

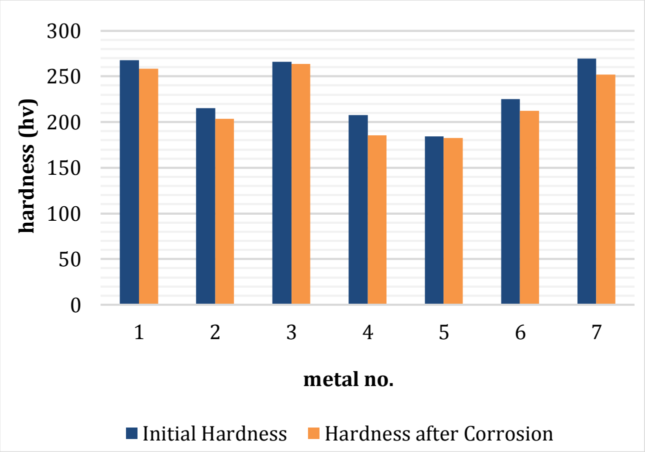

Fig. 11. Variations of the initial hardness of metals after corrosion in Murban

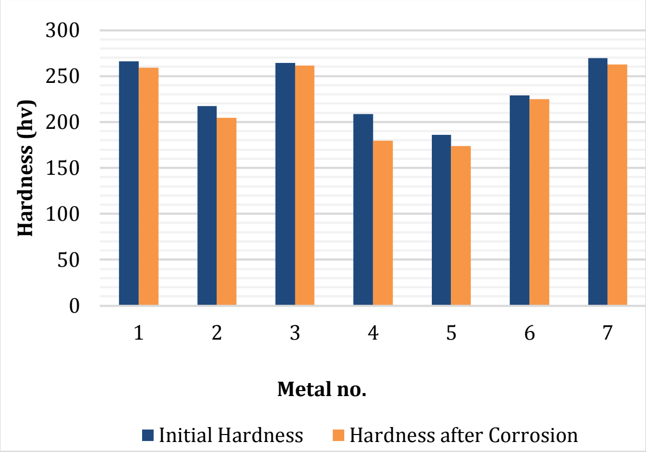

Fig. 12. Variations of the initial hardness of metals after corrosion in Das Blend

By referring the variations of the initial hardness of metal coupons after interaction with petroleum oils it was identified the reduction of the initial hardness of each metal coupon in a slight amount. During to the formation of the corrosion such corrosion compounds tend to be removed from the metal surface due to the effect of repulsive forces and attractive forces between successive electrons and protons of relevant atoms while creating some kind of instability and heterogeneous conditions on the initial metal surface (Calister et al., 2003). Therefore, the reduction of the initial hardness can be used as evidence for the formation of the corrosion compounds on the surfaces of such metal coupons due to the effects of petroleum oils.

4. CONCLUSION

The higher efficiency of corrosive protection film was found at ~18% of chromium and ~8% of nickel in stainless steels against the corrosive environment. The proper progress of both “sulfidation” and “localized corrosion” is impossible and the effect of salts on the metallic corrosion is stronger than the effect of organic acids on the metallic corrosion in the normal temperature conditions. The major corrosion compound that formed during the interaction between metals and petroleum oils would be FeS. There can be happened the decay of materials into the petroleum oils during the corrosion and also it is possible to change some surface properties of metals such as the hardness of the metals due to the corrosion.

5. ACKNOWLEDGMENTS

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