

Annotation of the Corrosive Ardour of Petroleum Oils

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PAPER INFO	ABSTRACT
<p>Chronicle: Received: 16 August 2018 Revised: 20 October 2018 Accepted: 20 November 2018</p>	<p>Crude oil is an energy resource that mostly is composed of the hydrocarbons and trace corrosive compounds as well has found some cruxes in the petroleum refining industry because of the vast applications of the different types of metals. There is expectation to investigate the effect of two different types of crude oils on the corrosion of seven types of the selected ferrous metals in the pertinent research. The concerned corrosive properties of both crude oils have tested by standard methods and instruments, also the chemical compositions of the selected ferrous metals have tested. The corrosion rates of a series of the similar size prepared metal coupons have tested after limited immersion time periods as three trials in crude oils by the weight loss method since aiding the microscopic analysis to identify the corrosion compounds. Apart from those speculations, the decayed metallic elemental mounts from the metals into crude oils and the reductions of the initial hardness of metal coupons have tested. The obligatory results shows the relatively higher corrosion rates from carbon steels, relatively lower corrosion rates from stainless steels in both crude oils, formation of FeS in most of observations, decay of ferrous and copper into crude oils from some metals, and slight reductions of the initial hardness of metal coupons due to the formation of the corrosion on the metal surfaces.</p>
<p>Keywords: Crude Oils. Corrosive Compounds. Ferrous Metals. Corrosion. Weight loss.</p>	

1. Introduction

Crude oil is mostly used energy resources in the consumptions of the energy for the industrial purposes, such as the power generations, automobiles, and even the domestic purposes. Although the crude oils are impossible to use in their raw forms for such applications and these crude oils need to be refined into various components which are having some economic value, such as petroleum gases, diesel, gasoline, and some by products. Usually, the raw crude oils may be composed of a large number of different hydrocarbons and some of the trace compounds including corrosive compounds which are tend to cause a severe impact on the ferrous metals foremost in the industry of crude oil refining because of the significant applicability of metals for some of special tasks in the industry of crude oil refining under various conditions [1-6]. Usually, the corrosion is a result of either chemical or electrochemical process on the metals by the strong oxidizing agent or any environment that contained with both water and oxygen; also the processes may be modified by the acids and salts presence in the system [1-15].

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According to the major corrosion compounds and the features of such corrosion compounds, the corrosion phenomenon can be sub divided into several categories such as the galvanic corrosion, pitting corrosion, general corrosion, and thermal corrosion [1, 4, 6].

In the current research, the scope had been scheduled to investigate the effects of corrosive properties of two different types of crude oils on the corrosion of seven different types of selected ferrous metals including the manual determination of the corrosion rates of such metals, microscopic analysis of the formed corrosion compounds on the metal surfaces and the variations of the surface properties of such metals due to the formation of the corrosion such as the variations of the initial hardness of such metals.

2. Materials and Methodology

In the selections of the metals, there were considered the vast range of applications of such metals in the various units of the crude oil refining process under different conditions of such crude oils and their final products. The industrial applications of seven selected types of ferrous metals have been given in the below.

- Carbon Steel (High) – Transportation tubes.
- Carbon Steel (Medium) – Transportation tubes.
- Carbon Steel (Mild Steel) – Storage tanks.
- 410-MN: 1.8 420-MN: 2.8 (stainless steel) – Crude distillation column.
- 410-MN: 1.7 420-MN: 1.7 (stainless steel) – Crude distillation column, heat exchangers.
- 321-MN: 1.4 304-MN: 1.9 (stainless steel) – Pre heaters, crude distillation unit, desalting units.
- Monel 400 – Heat exchangers.

The chemical compositions of such selected ferrous metals were tested by the XRF detector. The XRF detector is an instrument which is designed based on the principle of the X-ray penetration through the materials and it allows to be read the percentages of every composite metallic element and most of nonmetals excluding the carbon [1, 4, 5]. In additions, two different types of frequently used crude oils were selected as the samples or corrosive agent to investigate the corrosion rates of such metals with respect to such crude oils, namely Murban and Das Blend. By referring there chemical compositions, the Das Blend crude oils have been categorized as a ‘sour’ crude oil because of its higher sulfur content and an essential factor regarding the cause of metallic corrosion. Murban is a matter of fact crude oil with the balanced chemical composition [4-18].

The dominant corrosive properties of both Murban and Das Blend crude oils which are important for the current experiment were tested by the standard methodologies and instruments as summarized in the Table 1.

Table 1. Test methodologies of corrosive properties of crude oils.

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

A batch of the similar sized metal coupons was prepared from the seven types of selected ferrous metals as six metal coupons from each type of metals and altogether forty two metal coupons from seven different types of ferrous metals. According to the necessities of the further calculations and precautions, the conditions of such metal coupons were managed as following.

- Similar sized.
- Equal dimensions and equal surface area.

The surfaces of such metal coupons were cleaned well while observing through the 400X lens of an optical microscope until getting free of any corroded particle or any heterogeneous material on such metal surfaces. Apart from that, the dimensions of each metal coupon and the initial weight of each metal coupon were tested.

The prepared metal coupons were immersed in both Murban and Das Blend crude oil samples separately as three homogeneous metal coupons in each crude oil sample altogether seven Murban crude oil samples and seven Das Blend crude oil samples according to the seven different types of ferrous metals as given in the Fig. 2.

After fifteen days from the immersion one metal coupon from each crude oil sample was taken out and the suddenly the corroded metal surfaces were observed by the 400X lens of an optical microscope. The corroded metal surfaces of the metal coupons were cleaned by the sand papers and isooctane until getting free of any corroded particle on that metal surfaces and finally the remaining weight of each metal coupon was measured and the corrosion rate of each metal coupons was determined by the weight loss method as explained in the below [10, 18].

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

Where, W= weight loss in grams, k= constant (22,300), D= metal density in g/cm³, A= area of metal piece (inch²), t= time (days), and CR= corrosion rate of metal piece.

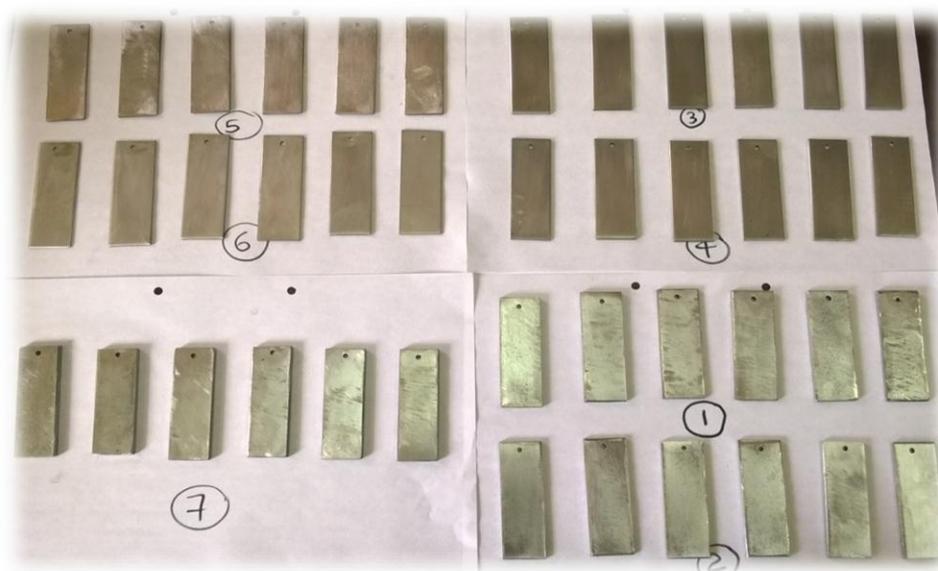


Fig. 1. Prepared metal coupons.



Fig. 2. (a) Metal coupons and (b) the immersed metal coupons.

The same procedure was repeated for another two similar batches of remained metal coupons in crude oil containers in order of after thirty and forty-five days from the immersion, and their corrosion rates were determined by the same methodology.

The microscopic analysis was performed regarding the qualitative analysis of the formed corrosion compounds on the metal surfaces by the 400X lens of the laboratory optical microscope.

Based on the on the observations of the invisible weight losses of some metals coupons when determining the corrosion rates of such metal coupons the decayed ferrous and copper concentrations from metals into crude oils samples were tested by the Atomic Absorption Spectroscopy (AAS). According to the sample preparation methodology for the atomic absorption spectroscopic test 1 ml of each crude oil sample was diluted with 9 ml of 2-propanol and filtered. Finally there were tested the

reductions of the initial hardness of metal coupons due to the formations of the corrosion compounds on the metals surfaces by the Vicker’s hardness tester. This is an instrument which is used to measure the hardness of some particular point on the metal surface. According to the necessity and the accuracy of the results, the hardness were tested at least three positions of each metal coupon and the average values were interpreted as the hardness of such metals with respect to each moment namely as the initial hardness and the hardness after formation of the corrosion compounds on the metals surfaces [1, 3, 4, 5, 6].

3. Results and Discussion

According to the obtained results of the XRF detector, the chemical compositions of selected ferrous metals have been shortlisted in the Table 2.

Table 2. Chemical compositions of selected ferrous metals.

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 above results showed the some variations of the chemical compositions of carbon steels, stainless steels, and Monel metal especially there were found higher ferrous amounts from all of carbon steels, moderate amounts of ferrous in stainless steels, and trace amounts of ferrous from Monel metal. The special case that there was found the doping of some trace metallic elements in stainless steels apart from the ferrous and other major elements. These alterations of the compositions of the stainless steels are some enhancements of the important mechanical properties of such metals such as the hardness, strength, and also the reduction of the corrosion of such metals. According to the theoretical concepts of reductions of the corrosion, the most possible mechanism that the self-corrosion protection layer of the stainless steels is at ~12% of chromium and sufficient amounts of nickel which is defined especially for the stainless steels [1, 3, 5, 6, 16, 17].

According to the analysis of the corrosive compounds in both Murban and Das Blend crude oils, the observe results have been shortlisted with respect to the corrosive compounds in the Table 3.

Table 3. Corrosive properties of both Murban and Das Blend crude oils.

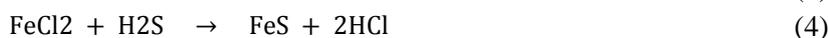
Property	Murban	Das Blend
Sulfur content (Wt. %)	0.758	1.135
Salt content (ptb)	4.4	3.6
Acidity (mg KOH/g)	0.01	0.02
Mercaptans content (ppm)	25	56

According to above results of the analysis of the corrosive compounds of both Murban and Das Blend crude oils there can be observed the higher elemental sulfur content, Mercaptans sulfur content, acidity, and the lower salt contents in Das Blend crude oil than the Murban crude oil, although these observations should be analyzed with the limitations of the causing of the corrosion due to the such corrosive compounds forever.

Salts are the dominant corrosive compounds that found in the crude oils in various amounts especially as a trace compound usually in the forms of NaCl, MgCl₂, and CaCl₂ since the occurrences of such crude oils. Also the measurement of the salt content is indicates the total amounts of the salts present in some certain crude oil. When increasing the temperature of the system such salts tend to be converted in to HCl as results of the reaction with the moisture and water presence in the system/crude oils as given in the Eq. (2) [2, 4, 7].



At the initial stage such HCl molecules are behaved as inactive compounds, although with the decreasing of the temperature such HCl molecules tend to react with water or moisture presence in crude oils and formed highly corrosive hydrochloric acids. The chemical reactions of the hydrochloric acids when causing the corrosion of the metals are given in the Eqs. (3) and (4) [4,7].



Also this process is some sort of reproducing mechanism of HCl and it's possible to expect some impact from the hydrogen sulfide on the metallic corrosion as a major byproduct during this mechanism; the impact of the hydrogen sulfide should be analyzed in another research descriptively.

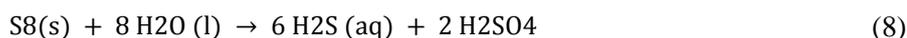
Organic acids are the major trace compounds that found in most of crude oils since the natural occurrences also ahead in the cause of the metallic corrosion. Usually, such organic acids are known as naphthenic acids under the common general chemical formula of 'RCOOH' and the term of acidity interprets the total amount of such acids in some particular crude oil or is possible to be named that term as the Total Acid Number (TAN) [2, 4, 9, 12, 15]. The general chemical reactions of the causing of the corrosion on the metals have been given in the Eqs. (5-7).



Also, in this mechanism it is possible to expect some long term impact from the hydrogen sulfide because in this mechanism the hydrogen sulfide is found as the byproduct and the reproducing of the

organic/naphthenic acids again [2, 9]. Although, it needs some investigation of the impacts of such hydrogen sulfides on the metallic corrosion.

The sulfur plays a dominant role in the metallic corrosion because it has several polymorphs and various active sulfur compounds and also most of them have been identified as the corrosive compounds due to the highly active functional groups and fractions of them, such as elemental sulfur, thiophenes, hydrogen sulfides, and Mercaptans which have the functional group of ‘RSH’ and it has strong affinity of electrons [2, 4, 8]. According to the type of the functional groups of the sulfur compounds, the corrosion mechanisms and limitations may be varied; commonly the corrosion due to the elemental sulfur is known as the ‘localized corrosion’ likely is happened at about 80⁰C and the corrosion process due to the most of active sulfur compounds is known as the ‘sulfidation’ usually is happened properly at about 230⁰C. The general chemical reactions for above corrosion processes have been given in the Eqs. (8) and (9) [2, 4, 8, 13, 14].



In the sulfidation process there may be formed both hydrogen sulfides and sulfuric acids as the results of the reactions of sulfur with water or moisture at some higher temperatures as mentioned above; both compounds are highly corrosive compounds although cannot make any forecast of the impact of hydrogen sulfides because of the volatility of the hydrogen sulfides, and the impact from the sulfuric acids can be expected as long as.

By referring the overall impact from various corrosive properties of crude oils on the metallic corrosion it is possible to keep much confidents on both organic acids and salts on the results of this experiment because of the less progressiveness of sulfur compounds on the metallic corrosion at lower temperatures.

The determined values for the corrosion rates of metals with respect to the Murban and Das Blend crude oils are interpreted in Table 4 and Table 5.

Table 4. Corrosion rates of metal coupons in Murban crude oil.

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		0.011968	0.007574	0.0452676

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)
(Stainless Steel)	0.11626			
(6) 321-MN: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 5. Corrosion rates of metal coupons in Das Blend crude oil.

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 Das Blend and Murban crude oils have been summarized in the bar chart of Fig. 3.

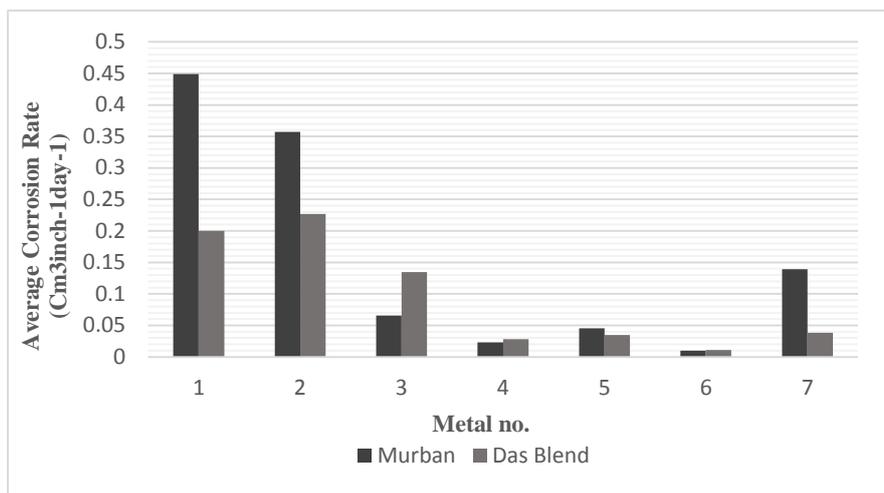


Fig. 3. Average corrosion rates of metal types in both Murban and Das Blend crude oils.

When considering the summary of the averages corrosion rates basically there can be found the relatively higher corrosion rates from carbon steels, intermediate corrosion rates from mild steels, and Monel metal; also significantly lower corrosion rates from the stainless steels. Based on the analysis of the corrosion rates of the three different types of stainless steels there can be clearly concluded that the higher performances of self-corrosive protection film are at least 12% of chromium and sufficient amount of nickel as an essential component because the least corrosion rates were found from 321-MN: 1.4 304-MN: 1.9 (stainless steel) which was composed 18% of chromium and 8% of nickel; among stainless steels, the highest corrosion rates were found from 410-MN: 1.7 420-MN: 1.7 (stainless steel) which was composed 12% of chromium. Although the lack of nickel and the intermediate corrosion rates were found from 410-MN: 1.8 420-MN: 2.8 (stainless steel) which was composed 11% of chromium and 0.2% of nickel [1, 3, 4, 5, 17].

By comparing the corrosion rates of metals with respect to both crude oils there was found the higher corrosion rates of four types of metals in Murban crude oils and higher corrosion rates of three types of metals in Das Blend crude oils as an asymmetric distribution of the corrosion rates of metals. Finally, it can be concluded a few possible reasons for the above variations as given in the below [2, 9, 15, 18].

- Improper progress of sulfur and active sulfur compounds at lower temperatures.
- Salts are much stronger in the metallic corrosion rather than the organic acids.
- There may be affected some various corrosive compounds on the metallic corrosion in addition to the investigated corrosive properties of crude oils in this experiments.

The variations of the corrosion rates of metals with the exposure time period have been interpreted in the Fig. 4.

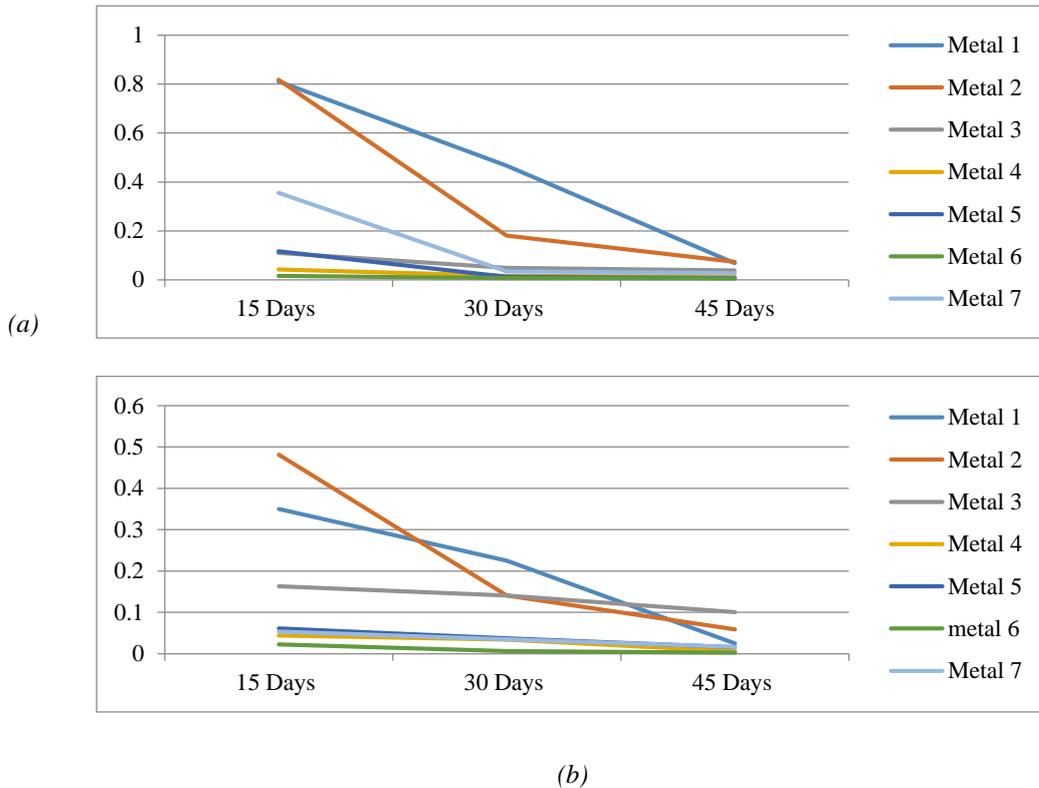
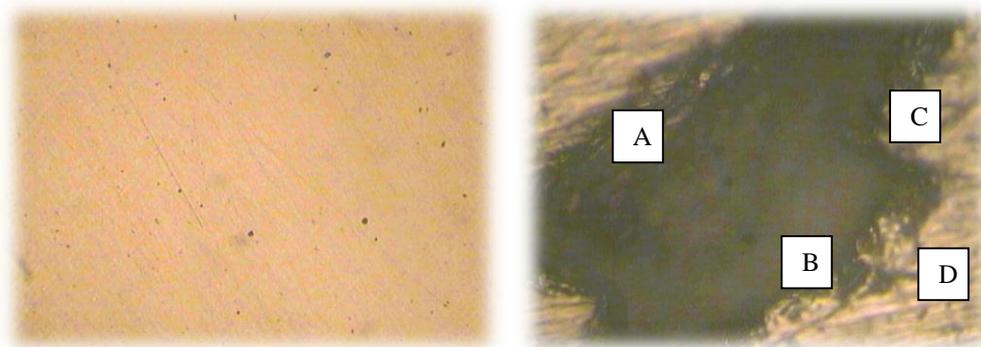


Fig. 4. Variations of the corrosion rates of metals with the exposure time period against the (a) Murban and (b) Das Blend crude oils.

By considering the summary of above distributions of the corrosion rates of metals there can be found some similarity of the variations of corrosion rates of each metal with the exposure time period [10, 18]. These observations can be used as the confirmations of the independency of inversely proportional relationship between the corrosion rate and the exposure time on the metal type forever.

As the results of the microscopic analysis of the corroded surfaces of the metal coupons there were observed some specific features as confirmations and as new observation that need to be analyzed in another research. The specific observations and most highlighted things have been shown in the Fig. 5.



(a)

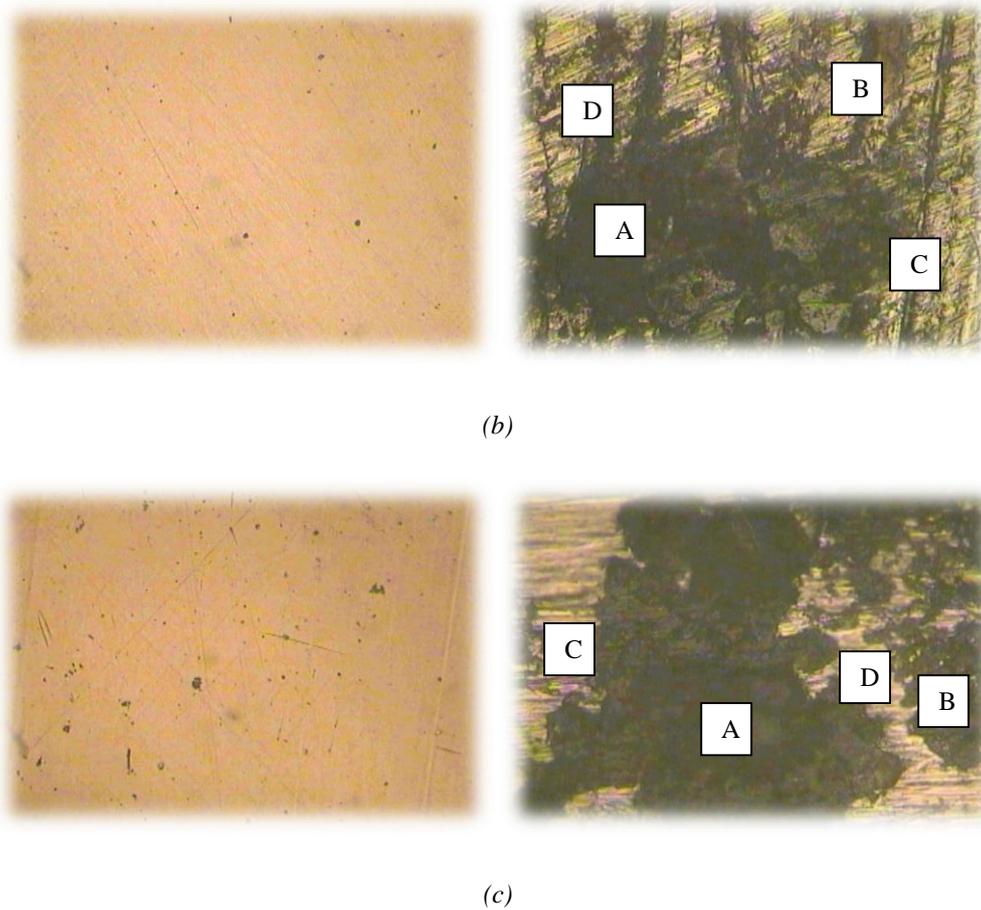


Fig 5. Corroded metal surface of (a) Carbon steel (high) in Das Blend, (b) carbon steel (mild steel) in Murban, and (c) 410-MN: 1.7 420- MN: 1.7 (stainless steel) in Murban.

The analysis of these observations and the identifications of the corrosion compounds were based on the visible appearances of such corrosion compounds foremost of the color and surface changes with the corrosion such as the cavities and cracks. The explanations of the visible appearances of the relevant corrosion compounds have been discussed in the Table 6 [1, 3, 4, 5, 17].

- A-Black color/ brownish black color.
- B-Rusty color/ brownish black color.
- C-Cracks.
- D-Cavities.

Table 6. Appearances of corrosion compounds

Compound	Appearances	Observations
FeS	Black, brownish black, property of powder, pitting, and cracks.	Observed most of features in each metal piece.
Fe ₂ O ₃	Rusty color.	Observed rarely.
CuS	Dark indigo/dark blue.	Unable to specify.

Basically, there were observed some FeS frequently, Fe₂O₃ rarely, cavities, and some asymmetric distributions of the corrosion cracks among the major corrosion compounds on the metal surfaces. It

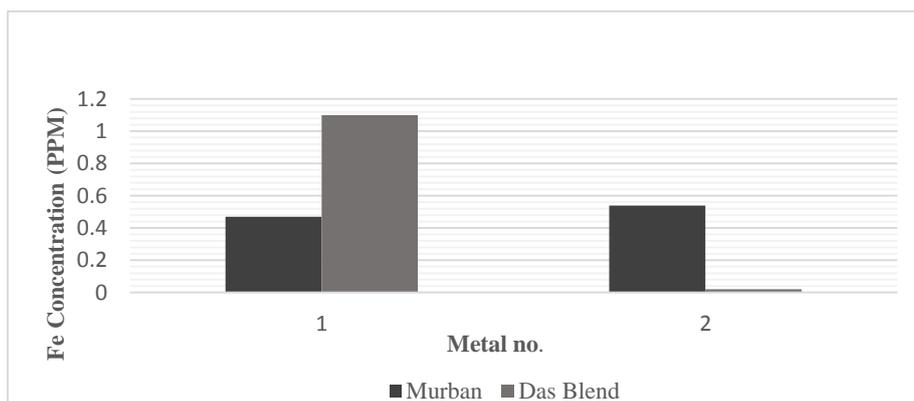
was observed uncertainly the black color corrosion compound on Monel metal surfaces which is similar to the FeS and may be possible to suggest as the CuS although need to be analyzed compositionally in advanced such as the method of X-ray Diffraction (XRD) rather than the analysis based on the visible appearances [1, 4, 5, 6, 17].

According to the obtained results for the analysis of the atomic absorption spectroscopic (AAS) the summarized results for the decayed ferrous and copper concentrations into crude oil samples have been interpreted in the Table 7 and Fig. 6.

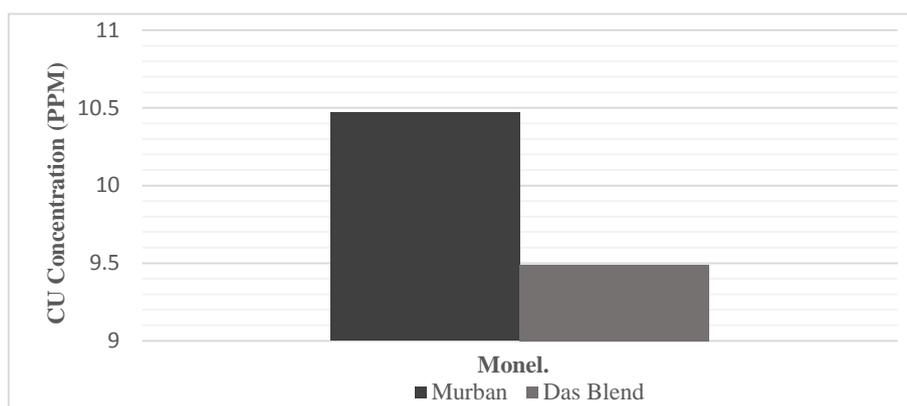
Table 7. The results of the atomic absorption spectroscopic (AAS) analysis of crude oils.

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

By referring the variations of the hardness values of metal coupons before the immersion in crude oils and after formation of the corrosion, there can be identified some sort of slight reduction of the initial hardness of each metal coupon after formation of the corrosion. According to the theoretical explanation of that incident that there can be explained and clarified after formations of the corrosion compounds on the metal surfaces such compounds play a state of unstable because of the attractive and repulsive forces between the successive electrons and protons and tendency to be removed from the initial metal surfaces as the formed compounds and to be extracted to the surrounded medium [1, 3, 4, 5]. Under such conditions, it is possible to create some unstable status because of heterogeneity of the relevant metal surfaces; also the reductions of the initial hardness of metal coupons were possible to explain with that incident.



(a)

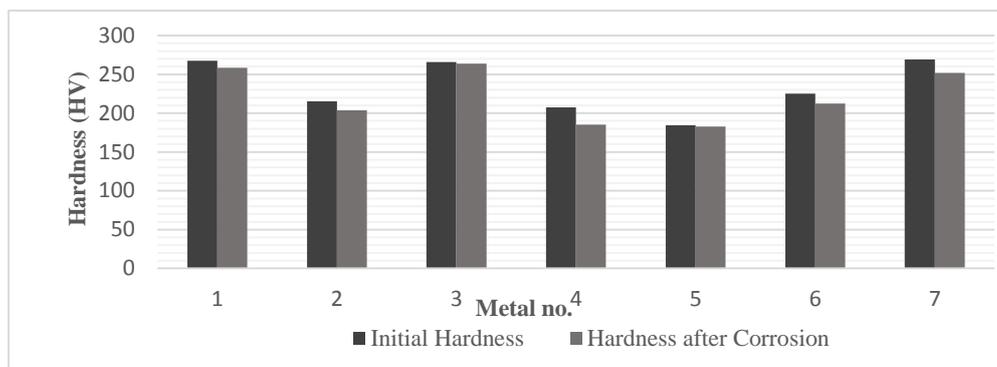


(b)

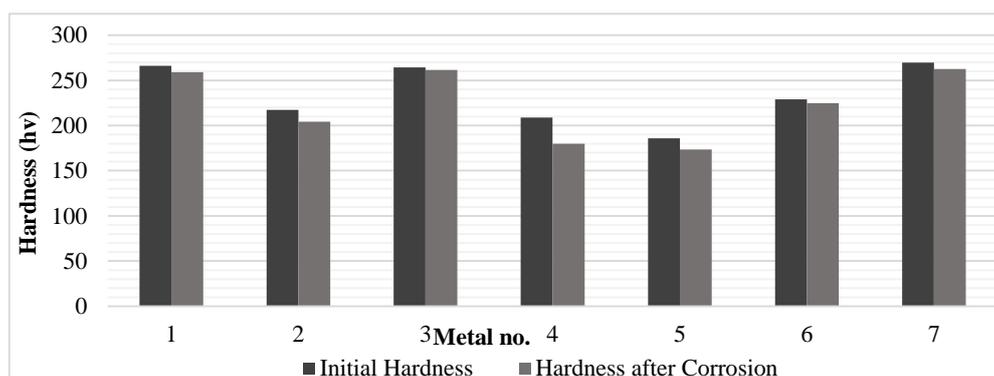
Fig. 6. (a) Decayed ferrous concentrations into crude oil samples and (b) decayed copper concentrations in to crude oil samples.

According to the above results, the higher amounts of ferrous were decayed into crude oils from both carbon steels (high) and carbon steels (medium), also highest corrosion rates in both crude oils were found since there was not observed decay of any amount of ferrous from any stainless steels or carbon steel (mild steel). Also there was found the significant decay of copper from the Monel metal into both crude oils when comparing the decayed amounts of ferrous. Regarding the common theories of the formations of the corrosion and special behaviors of such corrosion compounds specially after the formations of the corrosion compounds on the metal surfaces, such compounds tend to remove from the metal surface because of the strength of the repulsive and attractive forces between the successive electrons and protons as the formed compounds or some possible forms [1, 3, 4, 5]. Therefore, the invisible weight losses of some metal coupons can be clarified with this analysis and explanations.

The variations of the hardness of metal coupons which were obtained according to the observed results of the Vicker's hardness tester have been shown in the Fig. 7.



(a)



(b)

Fig. 7. Variations of the initial hardness of metal coupons in (a) Murban and (b) Das Blend crude oils.

4. Conclusion

Basically there were found the higher corrosion rates from carbon steels, lower corrosion rates from stainless steels, and intermediate corrosion rates from Monel in both Murban and Das Blend crude oils since the least corrosion rates were found from 321-MN: 1.4 304-MN: 1.9 (stainless steel) with respect to both Murban and Das Blend crude oils because of the corrosive protection chemical compositions ~18% of chromium and ~8% of nickel. In addition, the obtained results showed the higher corrosive strength from salts rather than the corrosive strength of organic acids since the improper progress of both process of 'sulfidation' and 'localized corrosion' at the low temperatures. The formation of the FeS as the major corrosion compound on the metallic surfaces with some of corrosion cracks, the cavities also rarely found Fe₂O₃. The decay of ferrous amounts from carbon steels and stainless steels into crude oils and the decay of copper amounts from Monel metal into crude oils were found in the Atomic Absorptions Spectroscopic (AAS) analysis. Eventually there were found the slight reductions of the initial hardness of metal coupons due to the formation of the corrosion on the metal surface.

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