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CORROSIVE TRENDS OF MINERAL OILS AND EFFECTS OF SUCH DISTINCTIVE COMPOSITES ON THE METALS

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Abstract

The investigations of the impact of corrosive properties in mineral oils on the corrosion rates of different types of metals and the nature of such corrosion with respect to the corrosive compounds of mineral oils and the metal types were the aims and objectives of the research. The corrosive properties of two different types of selected crude oils and the chemical compositions of seven different types of ferrous metals were tested by the standard instruments and methodologies. The corrosion rates of the similar sized prepared metal coupons were determined by the relative weight loss method after certain immersion time periods in both crude oils. In addition, the corroded surfaces were observed by the optical microscope and analysed, decayed ferrous and copper amounts from metals into crude oils were tested by the atomic absorption spectroscopy (AAS) and the reductions of the initial hardness of the metal coupons were tested by the Vicker's hardness tester. As the results ultimately there were found the higher corrosion rates from carbon steels, lower corrosion rates from stainless, intermediate corrosion rates from Monel metal, higher impact from salts on the metallic corrosion when comparing with other compounds, formation of FeS, Fe_2O_3 rarely, corrosion cracks and pitting corrosion, higher amounts of decay of ferrous and copper from some metals into crude oils and slight reductions of the initial hardness of most of metals.

Keywords: *mineral oils, corrosive trend, ferrous metals, weight loss, decay, hardness.*

Introduction

Corrosion is a severe impact that found regarding the most of industrial applications of the ferrous metals which is usually explained as the formation of the ferrous oxides, sulfides or the hydroxides on the metal surface itself as a result of either chemical or electrochemical process that occur with the interaction of the relevant metal and the corrosive aided environment that rich in various corrosive compounds and basically the metal need to expose either some stronger oxidizing agent or water and oxygen composed environment. As the special things regarding the corrosion it is possible to emphasize the linkage between the parameters of the chemical compositions of the relevant metals and the strength of the contributed corrosive compounds of such processes [1-6]. According to the behaviors of such parameters and the supplementary conditions for the corrosion it can be categorized into a few of corrosion types such as general corrosion, pitting corrosion, galvanic corrosion and the thermal corrosion. Mineral oils is a mixture of hydrocarbons also having trace amounts of corrosive compounds such as the elemental sulfur, organic acids, active sulfur compounds and salts since the geological occurrence of such mineral oils and the compositions may be varied with the location or region of occurred [3-18].

In the most of researches with respect to the mineral oils and material engineering there have been investigated the effects and behaviors of mineral oils on the decay of metals and the stability of the metals regarding various mineral oils, various corrosive compounds and various metals as well. According to the scope of the current research there were expected to investigate

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the impact of two different types of mineral oils which are having different chemical compositions including the corrosive properties on the corrosion rates of seven different types of ferrous metals which are frequently used in the various sections of the industry of mineral oil or petroleum refining industry.

Materials and Methods

By considering the aims, objectives of the research and the availability of the materials there were selected two different types of mineral oils namely as Murban and Das Blend which are different in the chemical compositions and usually Das Blend is categorized as a “sour” mineral/crude oil because of the relatively higher sulfur content of that crude oil [2-9]. The dominant corrosive properties of both Murban and Das Blend mineral oils were tested by the standard methodologies and instruments as summarized in the Table 1.

Table 1. Test methodologies of corrosive compounds of 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.	End point
Mercaptans content	Each sample was dissolved in sodium acetate and titrated with silver nitrate.	End point
Salt content	Each sample was dissolved in organic solvent and exposed to the cell of analyzer.	Direct reading

In addition, seven different types of ferrous metals were selected to test the interaction with the selected two types of mineral oils also such metals are typically used in various sections the industry of petroleum refining as the equipment or materials. The chemical compositions of the selected metals were tested by the XRF detector. The selected ferrous metals and their typical applications are given in the below.

- Carbon Steel (High) – Transportation tubes
- Carbon Steel (Medium) – Transportation tubes, storage tanks
- Carbon Steel (Mild Steel) – Transportation tubes
- 410-MN: 1.8 420-MN: 2.8 (Stainless Steel) – Crude distillation unit
- 410-MN: 1.7 420-MN: 1.7 (Stainless Steel) – Heat exchangers
- 321-MN:1.4 304-MN:1.9 (Stainless Steel) – Crude distillation head
- Monel 400 – Pre heaters

A batch of similar sized metal coupons were prepared from such selected types of ferrous metals as six metal coupons from each type of metal and altogether there were forty-two metal coupons from seven different metal types and their surfaces were well cleaned as free of corrosion or another heterogeneous material on the surfaces of such metal coupons. As the necessity of the calculations the initial weights and dimensions of each metal coupon were tested by the electronic balance and the micrometer. The prepared metal coupons have been shown in the Fig. 1.

The prepared metal coupons were dipped in both mineral oil samples separately as three homogeneous metal coupons in one mineral oil sample and altogether fourteen mineral oil samples with the combination of seven Murban mineral oil samples and Das Blend mineral oil samples as shown in the Fig. 2.

After fifteen days from the immersion one metal coupon was taken from each mineral oil sample and the surfaces of such metal coupons were observed through 400X lens of the laboratory optical microscope. Also the corroded particles on the metal surfaces of such metal coupons were removed by the sand papers and isooctane for the ultimate requirements of the determination of the corrosion rates of such metal coupons. The mathematical expression (equation 1) of the weight loss method has been given in the below [9 - 10].

$$CR = W * k/(D * A * t) \quad (1)$$

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;

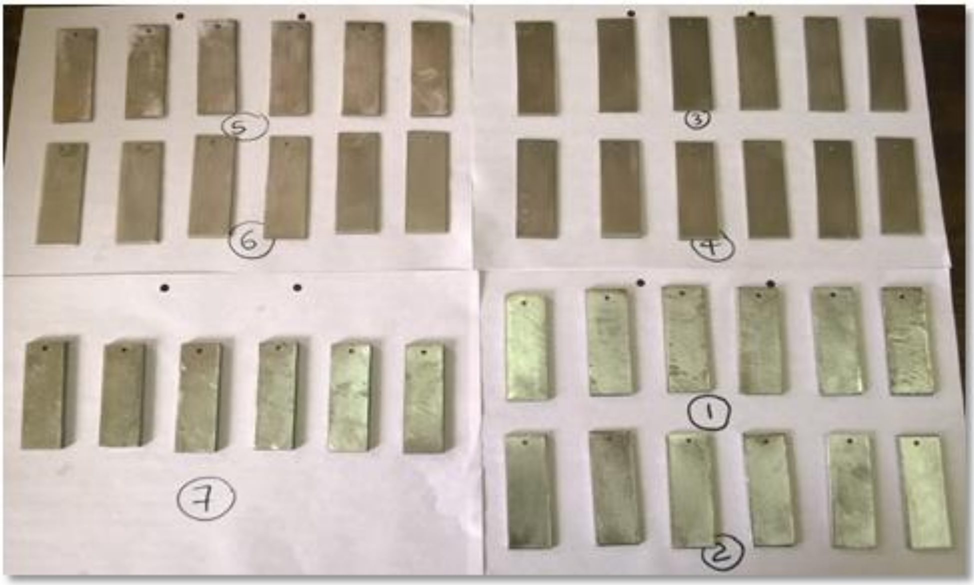

Fig. 1. Prepared metal coupons in similar dimensions

Fig. 2. Apparatus setup

The corroded metal surfaces were observed by the 400X lens of the laboratory optical microscope and based on this analysis there were expected a few of important information as given in the below.

- Confirmation of the corrosion free metal surfaces before the immersion in mineral oils;
- Identifications of the corrosion compounds based on their visible features;
- Confirmation of the corrosion free metal surface after cleaning the corroded particles.

Based on the observations of the invisible weight losses of some metal coupons that there were observed during the determinations of the corrosion rates of the metal coupons the decayed ferrous and copper concentrations from the metals into mineral oil samples were tested by the atomic absorption spectroscopy (AAS). According to the sample preparation methodology of that instrument 1 ml of each mineral oil sample was diluted with 9 ml of 2-propanol and filtered.

Finally there were performed the investigation of the reductions of the initial hardness of metal coupons due to the formations of the corrosion compounds on such metal surfaces. According to the scope the initial hardness and the hardness after formation of the corrosion of each metal coupon were measured by the Vicker's hardness tester. The working principles and the determination formula (equation 2) of the hardness by the Vicker's hardness tester have been explained in the below [1-5].

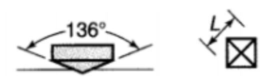

Fig. 3. The indenter of the Vicker's hardness tester

$$HV = 1.854 * P^2/L^2 \quad (2)$$

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

By considering the accuracy each hardness value was interpreted after measuring the hardness at least three points on each metal surface at once.

Results and Discussion

The obtained results for the analysis of the chemical compositions of the selected ferrous metals by the XRF detector have been given in the Table 2.

Table 2. Elemental compositions of the selected ferrous metal

Metal	Fe (%)	Mn (%)	Co (%)	Ni (%)	Cr (%)	Cu (%)	P (%)	Mo (%)	Si (%)	S (%)	Ti (%)	V (%)
(1)Carbon Steel (High)	98.6	0.43	-	0.17	0.14	0.37	0.12	0.086	0.09	-	-	-
(2)Carbon Steel (Medium)	99.36	0.39	-	-	-	-	0.109	-	0.14	<0.02	<0.04	-
(3) Carbon Steel (Mild Steel)	99.46	0.54	<0.30	-	<0.07	-	-	-	-	-	<0.19	<0.07
(4) 410-MN: 1.8 420-MN: 2.8 (Stainless Steel)	88.25	0.28	-	0.18	10.92	0.1	0.16	-	0.11	-	-	-
(5) 410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	87.44	0.3	-	-	11.99	-	0.18	-	0.09	-	-	-
(6) 321-MN:1.4 304-MN:1.9 (Stainless Steel)	72.47	1.44	-	8.65	17.14	-	0.18	-	0.12	-	-	-
(7)Monel 400	1.4	0.84	0.11	64.36	<0.04	33.29	-	-	-	-	-	-

By referring the above results there can be concluded that the carbons steels are composed with relatively higher amount of ferrous, stainless steels are composed with moderate amount of ferrous and the Monel metal is composed with significantly trace amount of ferrous. Especially there were observed the trace compositions of nickel and chromium in stainless steels. According to the material engineering theoretical concepts the combinations of the d-block trace metallic elements with the ferrous it may give various advanced properties for the ferrous metals such as the enhancement of the strength and the reduction of the tendency for the corrosion [1-9].

According to the analysis of the dominant corrosive compounds in the crude oils the obtained results have been shortlisted in the Table 3.

By referring the obtained results it seems the Das Blend crude oils was composed relatively higher amounts of organic acids, elemental sulfur and Mercaptans since it was composed relatively lower amount of salts. Although some chemical reactions regarding such corrosive compounds require specific supporting conditions for the proper progress.

Table 3. Corrosive properties of 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

Organic acids gained the significant status when discussing the metallic corrosion of crude oils. Also known as the “naphthenic acids” which are having a general formula of “RCOOH” and the summation of such acids of some particular crude oil are known as the “acidity” or “total acid number (TAN)” of such crude oil [2-18]. The general chemical reactions (equation 3) of such naphthenic acids with the ferrous metals are given in the below.

$$\begin{gathered} \mathrm{Fe} + 2\mathrm{RCOOH} \rightarrow \mathrm{Fe(RCOO)_2} + \mathrm{H_2} \\ \mathrm{FeS} + 2\,\mathrm{RCOOH} \rightarrow \mathrm{Fe(COOR)_2} + H_2\mathrm{S} \\ \mathrm{Fe(COOR)_2} + \mathrm{H_2S} \rightarrow \mathrm{FeS} + 2\mathrm{RCOOH} \end{gathered} \tag{3}$$

Salts are the abundant compound in various crude oils because of the naturally occurring process of such crude oils is taking a place in the interior part of the earth and also the earth crust is composed with such halides. Usually in the crude oils there were found NaCl, $CaCl_2$ and $MgCl_2$ as the salts and the summation of such slat is denoted as the salt content of a crude oil. In the heating conditions such salts tend to be broken into HCl molecules although has never found any corrosive behavior from such molecules at that stage. Although when the system is approaching towards the lower temperatures such HCl molecules tend to react with the water and the moisture that composed in such crude oils and formed hydrochloric acids which is known as the highly corrosive compound [2-7]. The important chemical resections with respect to the mechanism of the formation of the corrosion due to the salts have been given through the following chemical reactions (equation 4) [2].

$$\begin{gathered} \mathrm{CaCl_2} + \mathrm{H_2O} \rightarrow \mathrm{CaO} + 2\mathrm{HCl} \\ \mathrm{HCl} + \mathrm{Fe} \rightarrow \mathrm{FeCl_2} + \mathrm{H_2} \\ \mathrm{H_2} + \mathrm{S} \rightarrow \mathrm{H_2S} \\ \mathrm{FeCl_2} + \mathrm{H_2}S \rightarrow \mathrm{Fe}S + 2\mathrm{HCl} \end{gathered} \tag{4}$$

Same as the above discussed corrosive compounds sulfur has a dominant significance in the cause of corrosion on the metal surfaces with the aid of some specific environmental conditions that mainly identified some certain requirements of the temperature and the chemical structure of the sulfur consisted compounds. According to the classifications of the sulfur compounds basically those compounds were categorized into a few of various groups such as the elemental sulfur, hydrogen sulfide, mercaptans, thiophenes and sulfoxides and also most of them are highly corrosive compounds because of the higher oxidation ability of bonded functional groups with the atoms of sulfur while having a series of various mechanisms that based on the types of such bonded functional group [2-18]. When discussing within the scope of existing research that is emphasized both possible mechanisms to cause the corrosion due to the impact of elemental sulfur is “localized corrosion” which is usually happening properly at about 80°C and the corrosion mechanism due to the impact of Mercaptans is “sulfidation” which is happening properly in the range of temperature 230°C- 460°C. The initiation chemical reactions (equation 5) for most of above corrosion processes have been given in the following chemical reactions [2] [12].

$$\begin{gathered} S8(s) + 8\,H_2O\,(l) \rightarrow 6\,H_2S\,(aq) + 2\,H_2SO_4\,(aq) \\ 8\,Fe + S8 \rightarrow 8\,FeS \end{gathered} \tag{5}$$

By considering the resonance impact altogether with the all corrosive compounds on the formations of the corrosion of metals in the existing experiments the obtained results must be analyzed within the required environmental conditions for the happening of such chemical reactions that related with each considering corrosive compounds.

The determined results for the corrosion rates of metal coupons with respect to both crude oils by the weight loss method after 15, 30 and 45 days with the finalized average corrosion rates of each type of metal with respect to both crude oils have been interpreted in the Table 4 and Table 5.

Table 4. Corrosion rates of metals 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 (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

By using the results for the average corrosion rates of used metals with respect to both crude oils have been shown in the Fig. 4.

When considering the average corrosion rates of used metal types in the experiments that it is possible to identified the relatively higher corrosion rates from carbon steels, moderate corrosion rates from Monel and least corrosion rates from stainless steels as expected before the experiments. As the analysis of the variations of the corrosion rates of the used three types of stainless steels with the chemical compositions of such stainless steels the lowest corrosion rates were recorded from 321-MN:1.4 304-MN:1.9 (Stainless Steel) which is having ~18% of chromium and ~8.7% of nickel. By considering these data that it is possible to emphasized the higher corrosion protection by the self corrosive protection film that formed with the combinations of both chromium and nickel at least 12% of chromium with sufficient amount of nickel. The maximum corrosion rates were observed from 410-MN: 1.7 420-MN: 1.7 (Stainless Steel) among used three different types of stainless steels which is having ~12% of chromium although lack of nickel [1] [3-6]. Therefore, these results showed the better performances of the nickel chromium corrosion protection film at the recommended compositions of both nickel and chromium.

Table 5. Corrosion rates of metals 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

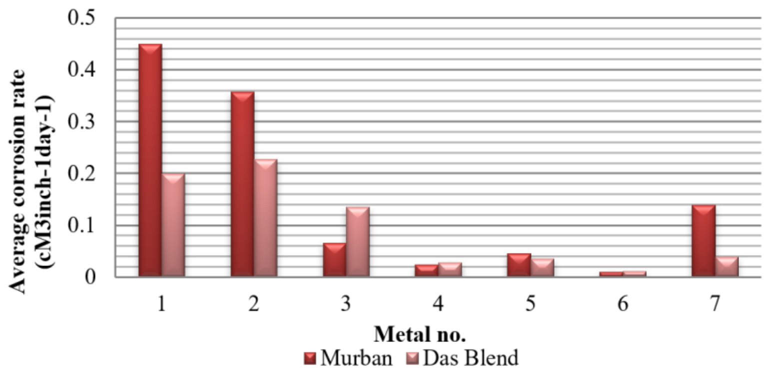

Fig. 4. Corrosion rates of metals in both crude oils

Also regarding the analysis of the impact of corrosive compounds on the corrosion rates of metals that four types of metals showed their higher corrosion rates in Murban crude oils while other three types of metals showed their higher corrosion rates in Das Blend crude oils since Das Blend was composed relatively higher amounts of elemental sulfur, Mercaptans, organic acids and lower amount of salts when comparing with such composites of Murban crude oil. In the comparisons of the obtained results for the corrosion impacts of both crude oils with the measured corrosive composites of such crude oils it can be concluded that there might be occurred some higher impact from salts rather than the impact of organic acids because of the less progressiveness of the corrosion process of sulfur compounds at the lower temperatures [12-18].

As the future works of these kind of researches it is recommended to analyze a large number of corrosive composites from various types of crude oils and the impacts of such compounds at the various environmental conditions as possible such as the various temperatures.

The variations of the corrosion rates of metals with the exposure time in the crude oils have been shown in the graphs below.

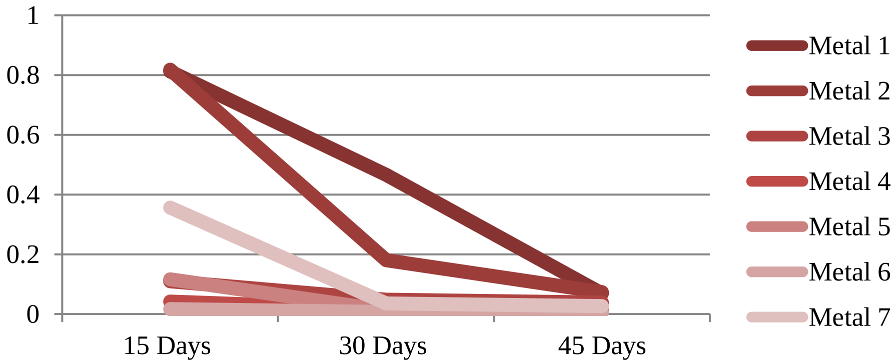

Fig. 5. Variations of the corrosion rates in Murban with the exposure time

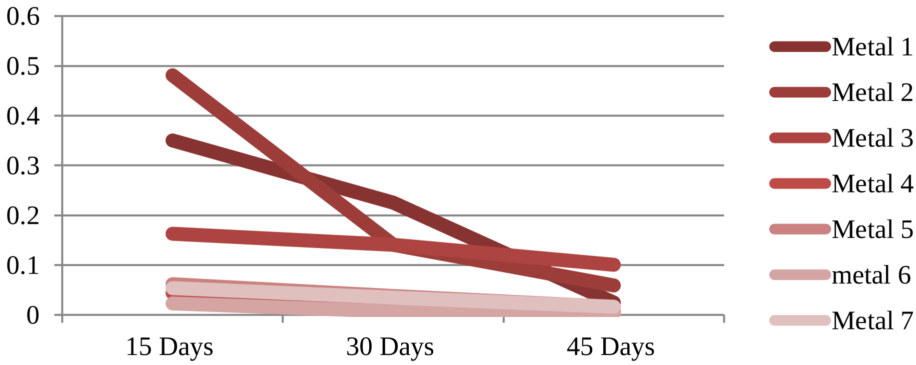

Fig. 6. Variations of the corrosion rates in Das Blend with the exposure time

The similar variations of the corrosion rates of metals with the exposure time period can be seen in the above graphs. Therefore, with these interpretations that it can be concluded the validity of inversely proportional relationship between the both parameters of corrosion rate and exposure time regarding the explanations of the weight loss method for various types of metals [9-10].

According the microscopic analysis of the corroded metals that there were identified some specific features and formed compounds on the metal surfaces qualitatively with the aid of some well defined distinguished properties of such corrosion compounds as emphasized in the Fig. 7.

Among the various observations on the corroded metal surfaces it is possible to conclude and detect some common specific corrosion compounds as given in the below.

A - Ferrous Sulfide

B - Ferrous Oxides and Trace Compounds

C - Corrosion Cracks

D - Pits and Cavities

The specific features of such corrosion compounds have been compared with the existing observations in the Table 6 [1] [3] [5 - 6].

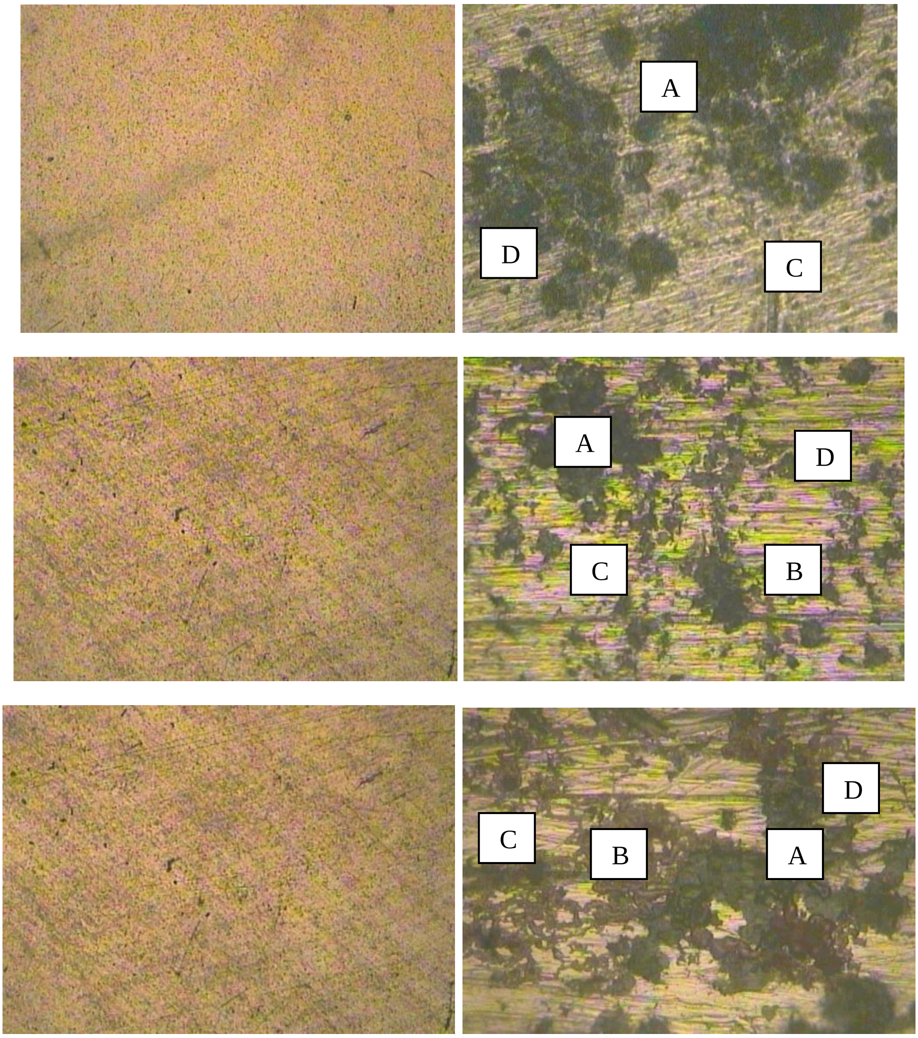

Fig. 7. Comparison of some specific corroded metal surfaces with the same initial metal surfaces

Table 6. Features of specific corrosion compounds

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

By comparing the observations and the relevant appearances of observed compounds that there can be concluded the formations of FeS, rarely Fe_2O_3, corrosion cracks and some pits on the metal surfaces asymmetrically. In some observations on the Monel metal that it was observed some similar corrosion compound with same appearances also possible to suggest as CuS although it is better to recommend some advanced chemical compositional analysis of such corrosion compound such as the X-ray diffraction (XRD) to distinguish such corrosion compounds.

The obtained results for the analysis of the decayed metallic elements into crude oils from the metals by the atomic absorption spectroscopy (AAS) have been interpreted below in the Table 7.

Table 7. Decayed ferrous and copper concentrations from metals into crude oils

Metal no.	Metal	Crude Oil	Fe - Concentration/ppm	Cu - Concentration/ppm
1	**Carbon Steel (High)**	Murban	0.47	-
		Das Blend	1.10	-
2	**Carbon Steel (Medium)**	Murban	0.54	-
		Das Blend	0.02	-
3	**Carbon Steel (Mild Steel)**	Murban	-0.08	-
		Das Blend	-0.48	-
4	**410-MN: 1.8 420- MN: 2.8 (Stainless Steel)**	Murban	-0.65	-
		Das Blend	-0.78	-
5	**410-MN: 1.7 420-MN: 1.7 (Stainless Steel)**	Murban	-0.71	-
		Das Blend	-0.79	-
6	**321-MN:1.4 304-MN:1.9 (Stainless Steel)**	Murban	-0.44	-
		Das Blend	-0.17	-
7	**Monel 400**	Murban	-	10.47
		Das Blend	-	9.49

The obtained results for the analysis of the decayed metallic elements into crude oils from ferrous metals during the interactions with crude oils by the atomic absorption spectroscopy (AAS) have been summarized and interpreted in the following graphs.

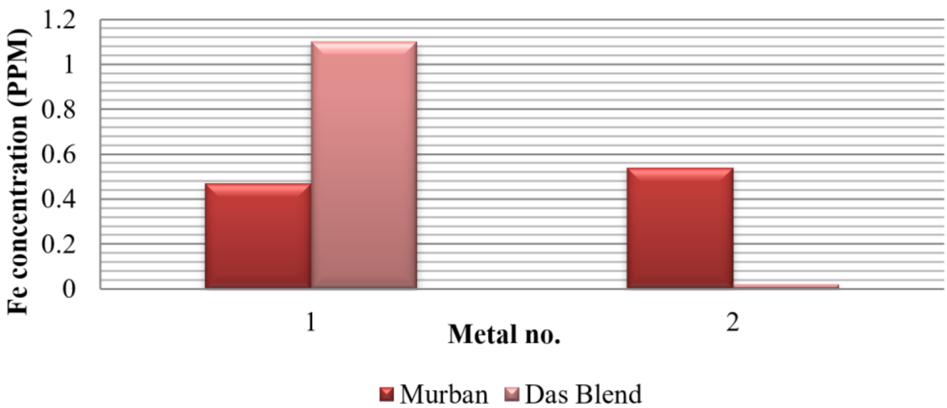

Fig. 8. Decayed ferrous concentrations into crude oils

As a summary of above interpretations there were observed the relatively higher decay of ferrous from high carbon steels and medium carbon steels since obtaining some negative results from the mild steels and stainless steels as simultaneous with the obtained results for the rate of corrosion of such metals. In addition that there were found some significant decay of copper from Monel metal also found some intermediate corrosion rates. According to the theoretical explanation that it is possible to explain with the concept that the repulsive and attractive forces between the successive electrons and protons of such metals and corrosion compounds those corrosion compounds tend to be removed from the metals surfaces as it is [1] [3] [6]. Among such removals it is possible to find FeS, Fe_2O_3 and various corrosion compounds. Therefore, it can be concluded the decay of ferrous and copper is happened simultaneous with the process of corrosion.

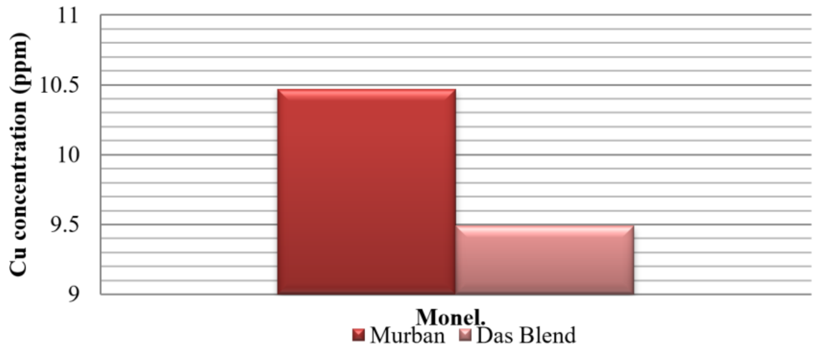

Fig. 9. Decayed copper concentrations into crude oils

The below graphs show the differences between the initial hardness and hardness after the corrosion on the metal surfaces with respect to both different types of crude oils regards the obtained results for the analysis of hardness of metals by the Vicker's hardness tester.

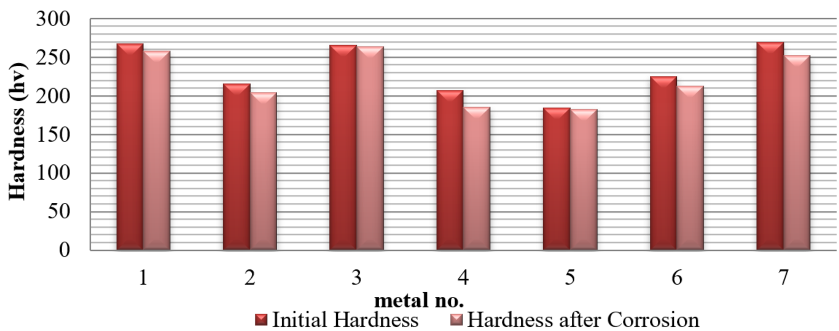

Fig. 10. Variations of the hardness of metals in Murban

The concluded results of the variations of the hardness of metals due to the corrosion showed a slight reduction of the initial hardness of each metal coupon after the corrosion. When considering the theoretical concepts regarding the material science that it is possible to explain under the concept of the electron and proton. After the formations of the corrosion compounds on the metals surfaces such heterogeneous compounds tend to be removed from the metal surface due to the repulsive and attractive forces between the successive electrons and protons of such compounds and metals also it is possible to be happened either completely or partially.

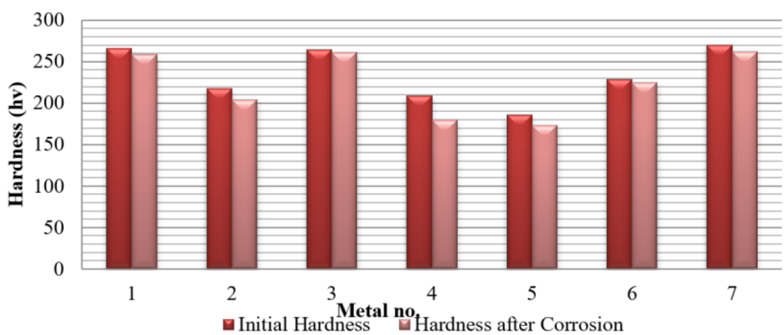

Fig. 11. Variations of the hardness of metals in Das Blend

According to those conditions the original metal surface may be contaminated or covered by the layers of such compounds [1] [3] [5-6]. Therefore, it is possible to find the dissimilarities of the initial properties of such metals and furthermore it is possible to conclude that the reductions of the initial hardness of metals were happened due to the formations of the corrosion.

Conclusion

Basically the results interpreted the more reasonable stuffs of the material science and engineering aspects such as the least corrosion rates of stainless steels due to the effects of the self corrosive protection layer of the stainless steels when having the combination of ~12% of chromium and sufficient amount of nickel, relatively higher impact from salts on the metallic corrosion at the lower temperatures when comparing with the impacts of other corrosive compounds especially the sulfur compounds, formations of the FeS, Fe_2O_3, corrosion cracks and pits on the metal surfaces, relatively significant decay of ferrous and copper from some of metals into crude oils while the interaction and the slight reductions of the initial hardness of most of metal coupons due to the formation of the corrosion on the metal surfaces.

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