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Study of the Incipient Components of the Mineral Oils on the Crumble of Metals

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A B S T R A C T

Ferrous metals play a dominant role in the industry of crude oil refining industry by covering basic units and essential equipments since facing some adverse impacts from such crude oils due to their corrosive composites. In the existing research there were expected to investigate the nature of the corrosion of seven different types of ferrous metals that applicable in the industry of crude oil refining industry due to the impacts of elemental sulfur, Mercaptans, organic acids and salts that presence in crude oils since the occurrences of such crude oils. The elemental sulfur contents, Mercaptans contents, acidities, salt contents and also the chemical compositions of selected ferrous metals were tested by in order of XRF analyzer, titration methods, salt analyzer and digital XRF detector. A set of similar sized prepared metal coupons were immersed in both crude oil containers separately as three homogeneous metal coupons per each crude oil container. The corrosion rates of each type of metal with respect to each crude oil type were determined by the immersed metal coupons in order of after 15, 30 and 45 days immersion time periods. As the further analysis the surfaces of corroded metal coupons, decayed ferrous and copper concentrations from metals into crude oils and the variations of the initial hardness of metal coupons were analyzed. Basically there were found the higher corrosive protection regarding the stainless steels when comparing with other metals especially having at least 12% of chromium and sufficient amounts of nickel, approximately higher contribution of salts in the metallic corrosion, formations of FeS, Fe_2O_3, corrosion cracks and pits, significant decays of ferrous during the corrosion, massive decays of copper from some metal which showed higher corrosion rates excluding stainless steels and the slight reductions of the initial hardness of most of metals due to the corrosion.

1. Introduction

Mineral oils are the vital earth resources which is an essential component of the providing of energy although it is required to be refined into various sub components. According to the chemical compositions of mineral oils that mainly composed with various hydrocarbons and some kinds of trace compounds while having different distinguish properties foremost of the corrosive properties. In the process of mineral oil refining there may be a vast range of applications in metals to cover various tasks including the manufacturing of the devices and units. Therefore, the metallic corrosion of mineral oils is closely linked with the industry of mineral oil refining. When considering the material engineering explanations of the corrosion usually it is defined as the formations of the metallic sulfide, metallic oxide or metallic hydroxide on the metal surface itself as the results of either chemical or electrochemical process in between the metals and surrounded environment which is mostly composed either strong oxidizing agent or some amounts of both water and oxygen [1-12]. According to the occurrences of mineral oils usually most of mineral oils are composed with some trace amount of water because of the impact of underground water bodies which are associated with some of bed rocks where the mineral oils are formed. In addition that it is possible to composed some various organic compounds in mineral oils that having the corrosive properties such as the organic acids, salts and a series of various sulfur compounds [3-15]. Regarding the relevant studies and investigations of the corrosive compounds and their impacts mainly there were found the higher impact from organic acids, salts, sulfur compounds and some of microorganisms especially at different conditions such as the various temperatures while forming various corrosion compounds on the metal surfaces [3-18].

In the existing research there were expected to investigate the nature of the corrosion of seven different types of ferrous metals which are widely applicable in the industry of mineral oils refining with respect to two different types of mineral oils that having slightly different chemical compositions including the corrosive compounds. The investigations were based on the analysis of the corrosion of such metals in both qualitatively and quantitatively.

2. Materials and Methodology

By considering the scope of the existing research and the availability of the mineral oils two different types of mineral oils were selected as the corrosive agents. Those mineral oils are namely as “Murban” and “Das Blend” which are slightly different in their chemical compositions including the corrosive compounds. Usually the “Das Blend” is known as a “sour” crude oil because of the relatively higher sulfur content of that crude oil [2-9]. The dominant corrosive

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compounds of such crude oils were analyzed by the standard instruments and methodologies as explained in the Table 1.

Table 1: Investigations of the corrosive properties 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 the selections of the metal types for the experiments mainly it was considered the applicability of such metals in the industry of mineral oil refining for various tasks and seven different types frequently applicable ferrous metals were selected as the samples which are given in the below with their applications.

- Carbon Steel (High) – Transportation tubes, pre heaters
- Carbon Steel (Medium)- Storage tanks, transportation tubes
- Carbon Steel (Mild Steel)- Storage tanks, heat exchangers
- 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- Pre heaters, de-salters

The chemical compositions of the selected metals were detected by the X-ray fluorescence technique as the percentages of all metallic elements and most of non-metals excluding carbon. A batch of similar sized metal coupons was prepared from seven different types of selected ferrous metals as six metal coupons from each metal type altogether forty two metal coupons from seven types of metals. The surfaces of metal coupons were cleaned well by the sand papers and isooctane until it will get free from any heterogeneous materials on the surfaces. The initial weight dimensions of each metal coupon were measured by in order of analytical balance and micrometer. The prepared metal coupons have been shown in the Fig. 1.

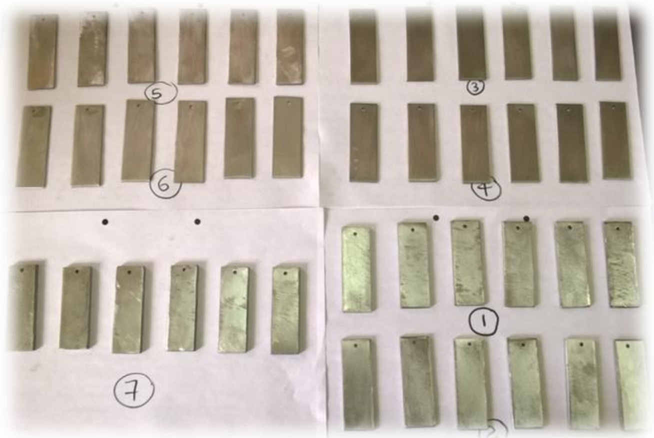

Fig. 1. Prepared metal coupons for the experiment

The prepared metal coupons were dipped in crude oil containers separately as three homogeneous metal coupons per each crude oil container that altogether seven Murban crude oil containers and seven Das Blend crude oil containers as shown in the Fig. 2.

Fig. 2. Apparatus and experimental setup

After fifteen days from the immersion one metal coupon was taken from each crude oil container altogether as a bath of fourteen metal coupons from all crude oil containers. The corroded metal surfaces were observed through the 400X lens of the optical microscope. The corroded metal surfaces were cleaned by the isooctane and sand papers and the final weights of such metal coupons were measured by analytical balance and ultimately the corrosion rates of such metal coupons were determined by the weight loss method. The mathematical expressions and the definitions of the weight loss method is 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

By following the similar methodology the corrosion rates of another two similar bathes of metal coupons in order of after thirty and forty five days immersion periods.

According to the qualitative analysis of the corrosion the formed corrosion compounds were basically observed through the 400X lens of an optical microscope and distinguished such compounds based on their visible appearances.

Based on the requirements of the clarifications of observations that the observed invisible weight losses of some metal coupons when determining the corrosion rates of such metal coupons the decayed ferrous and copper concentrations from metal coupons into crude oil samples were analyzed using the atomic absorption spectroscopy (AAS). In the sample preparation regarding the relevant analysis 1 ml of each crude oil sample was diluted with 9 ml of 2-propanol and filtered.

As a confirmation stage of the corrosion the variations of the initial hardness of metal coupons were measured by the Vicker's hardness tester. The working principles of Vicker's hardness tester are shown in the Fig. 3.

Fig. 3. 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

As the essential measures the initial hardness and hardness after formations of the corrosion of each metal coupon were measured by the above instrument.

3. Results and Discussion

The obtained results for the analysis of the chemical compositions of the metals by the X-ray fluorescence technique have been given in the Table 2.

Table 2: Chemical compositions of metals

Metal	Fe (%)	Mn (%)	Co (%)	Ni (%)	Cr (%)	Cu (%)	P (%)	Mo (%)	Si (%)	S (%)	Ti (%)	V (%)
(1)Carbon Steel (High)	98.60	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	0.10	0.16	-	0.11	-	-	-
(5) 410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	87.44	0.30	-	-	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	1.40	0.84	0.11	64.36	<0.04	33.29	-	-	-	-	-	-

According to the above analysis of the chemical compositions of metals carbon steels were composed higher ferrous amounts, stainless steels were composed intermediate ferrous amounts and Monel metal was composed trace amount of ferrous while having significantly higher amounts of nickel and copper. Especially there were fond relatively higher amounts of some d-block metallic elements such as molybdenum, chromium and nickel. The doping of the metals with such d-block elements is usual done based on some of expectations as given in the below [1, 3, 4, 5, 6].

- Enhancing the mechanical properties of the raw metal such as the strength and the hardness.
- Reducing the corrosion tendency of such metals or enhancing the corrosive protection ability of metals.

According to the much important factor that the reductions of the corrosive ability the most important thing when having a combination of at least 12% of chromium with sufficient amount of nickel is tend to be formed a self-corrosive protection layer on the metal surfaces and such layer is acting as a barrier for the further corrosion. Therefore, the corrosion rates of such metals will be reduced [1, 3,4,5,6, 17].
The obtained and determined results for the analysis of the corrosive properties of both Murban and Das Blend crude oils the relevant values have been interpreted in the Table 3.

Table 3: 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

In the evaluations of the impact of such corrosive compounds only considering the magnitude of the concentrations of such corrosive compounds the Das Blend crude oils was composed relatively higher amounts of organic acids, elemental sulfur, Mercaptans and lower amount of salts when comparing the relevant parameters of Murban crude oil. But according to the real estimation of the impacts of the corrosive compounds on the metallic corrosion there must be considered the supportive conditions for the occurrences of the relevant chemical reaction foremost of the temperature simultaneously with the magnitudes of such concentrations.
Organic acids are the traces compounds that presence in crude oils since the occurrences of crude oils also having the ability of corrosive causing which are also known as the "naphthenic acids" that denoted by the chemical formula of "RCOOH". Usually such naphthenic acids may have the ability of acting as a strong oxidizing agent for metals and some of metallic compounds such as the sulfides.

Also the organic acids are remaining in the same form at the end of the chemical reaction and therefore it is categorized as a catalyst for this chemical process [2, 4, 8, 12, 15, 18]. The general chemical reactions for the relevant process have been given in the Equation 3, Equation 4 and Equation 5.

$$Fe + 2RCOOH = Fe(RCOO)2 + H2 \tag{3}$$

$$FeS + 2\,RCOOH = Fe(COOR)2 + H2S \tag{4}$$

$$Fe(COOR)2 + H2S = FeS + 2\,RCOOH \tag{5}$$

Salts are the trace compounds that found in crude oils since the occurrence because of the abundance of salts in the interior part of the earth in the forms of NaCl, $MgCl_2$ and $CaCl_2$. When increasing the temperature of the system such salts molecules are converted into HCl molecules although show some inert conditions or non-reactive conditions at the same temperatures and with the decreasing of the temperature such HCl molecules tend to be reacted with the water of even moisture presence in crude oils and ultimately produced highly corrosive hydrochloric acids as shown in the Equation 6 [2, 7, 14, 15].

$$CaCl2 + H2O = CaO + 2HCl \tag{6}$$

Such hydrochloric acids are strong oxidizing agents because of the behaviours of strong acidic. Mainly in the process of corrosion with hydrochloric acids the elemental sulfur which are presence in crude oils play a significant role while happening the reaction since the hydrochloric acids are holding the role of catalyst as shown in the Equation 7, Equation 8 and Equation 9 [2,4,7,17].

$$HCl + Fe = FeCl2 + H2 \tag{7}$$

$$H2 + S = H2S \tag{8}$$

$$FeCl2 + H2S = FeS + 2HCl \tag{9}$$

Sulfur is a frequently found compound in crude oils since the occurrences in various forms because of the abundance of sulfur in the interior parts of the earth. Most of sulfur compounds have been identified as the corrosive compounds because of the high reactivity of the relevant functional groups such as the elemental sulfur, Mercaptans, sulfoxides, thiophenes and hydrogen sulfides. The relevant chemical processes and relevant products may be varied with the types of function groups and types of bonds that associated with them [2, 4, 7, 8, 13, 14, 18].

The corrosion process due to the elemental sulfur is known as the "localized corrosion" which is likely to happen properly at about 80^0C. Mercaptans are the active sulfur compounds that having the chemical formula of "RSH" which is tend to show the proper progresses in the temperature range of 230^0C- 460^0C that defined as the "sulfidation". The initiation chemical reactions regarding such processes have been given in the Equation 10 and Equation 11 [2, 4, 9, 12, 15, 18].

$$S8(s) + 8\,H2O\,(l) = 6\,H2S\,(aq) + 2\,H2SO4\,(aq) \tag{10}$$

$$8\,Fe + S8 = 8\,FeS \tag{11}$$

When considering the overall corrosive impact of such corrosive compounds it is mandatory to consider about the required environmental conditions that associated with such corrosion processes. Therefore, it is better to recommend some analysis of the more corrosive compounds from different crude oils and their progresses in a few of different temperatures as a future recommendation work.

The determined results of the corrosion rates of metal coupons in order of after 15, 30 and 45 days from the immersion in both crude oils have been interpreted in the Table 4 and Table 5.

Table 4: Corrosion rates of metals after certain immersion periods 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 5: Corrosion rates of metals after certain immersion periods 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 concluded results of the average corrosion rates of meals with respect to both Murban and Das Blend crude oils have been shown in the Fig. 4.

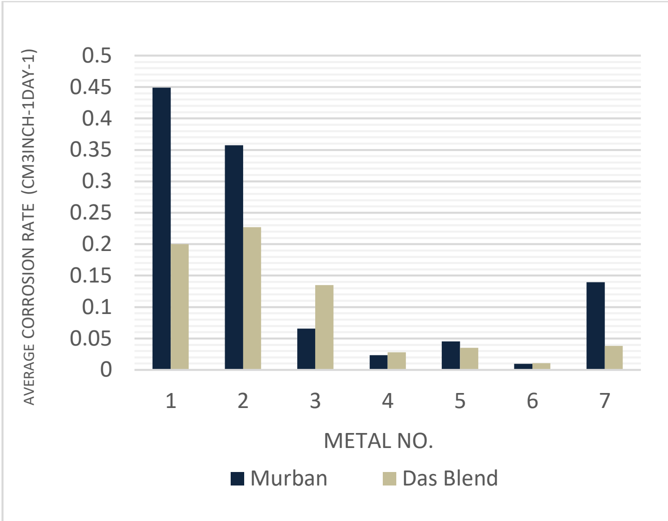

Fig. 4. Average corrosion rates of metals

According to the above interpretation the highest corrosion rates were observed from carbon steels, moderate corrosion rates were observed from mild steels and Monel while observing the lower corrosion rates from stainless steels as usual. Among the corrosion rates of stainless steels the least corrosion rates were obtained from 321-MN: 1.4 304-MN: 1.9 (Stainless Steel) in both crude oils. When considering the chemical composition of 321-MN: 1.4 304-MN: 1.9 (Stainless Steel) it was composed ~18% of chromium and ~8.65% of nickel which is much suitable combination for the formation if the self corrosive protection film on the metallic surfaces because the such contents meet the minimum requirements for such corrosive protection film that 12% of chromium and sufficient amount of nickel. Therefore, it is possible to conclude that the self corrosion protection film shows some higher performances when having more than 12% if chromium with sufficient amount of nickel [1, 3, 4, 5, 6, 17].

In the consideration of the corrosive properties of both crude oils and corrosion rates of metals with respect to both crude oils four types if metals showed their higher corrosion rates in Murban crude oil while other three types of metals were showing their higher corrosion rates in Das Blend crude oil. By comparing the corrosive compounds of both crude oils it is possible to emphasize that there might have some higher corrosive impact from salts when comparing the impact of other corrosive compounds. As the future recommendations there can be suggest a few of vast analysis of the corrosiveness of crude oils as given in the below.

- Investigation of the more corrosive compounds from different types oils with the supportive conditions such as the temperature
- Determinations of the corrosion rates of metals by using digital corrosion analysis instruments simultaneously with the determination by the weight loss method
- Analysis of the corrosion rates of metals at different temperatures because some of corrosive compounds may have their proper progress in higher temperatures such as the most of sulfur compounds

The variations of the corrosion rates of metals with respect to the exposure time period in both Murban and Das Blend crude oils have been shown in the Fig. 5 and Fig. 6.

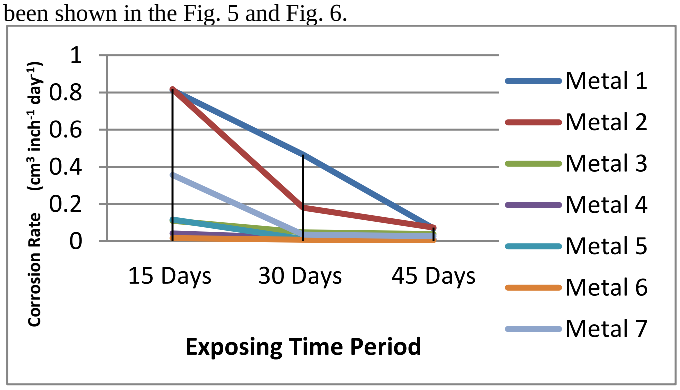

Fig. 5. Variations of the corrosion rates of metals with the exposure time in Murban

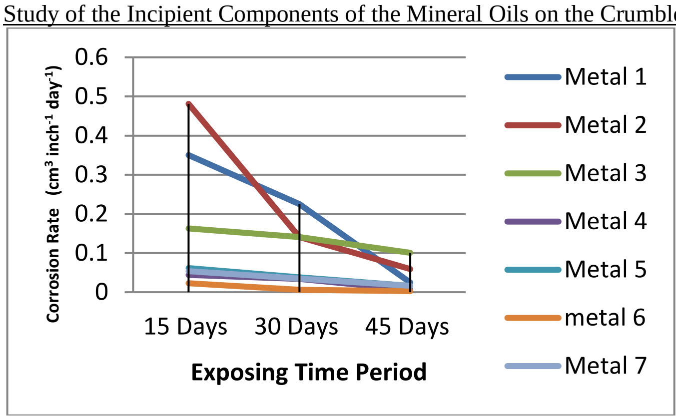

Fig. 6. Variations of the corrosion rates of metals with the exposure time in Das Blend

Above graphs showed some similar variations of the corrosion rates of different metals with respect to the exposure time in both crude oils. The obtained results are easily linked with the explanation of the weight loss method that which is the inversely proportional relationship between the corrosion rate and exposure time period. Therefore, the observed explained the applicability of the weight loss method for the various types of materials for the calculations of the corrosion rates [7, 9, 10, 12, 15].

According to the qualitatively analysis of the corroded metal surfaces through 400X lens of an optical microscope the most highlighted results have been shown in the Fig. 7.

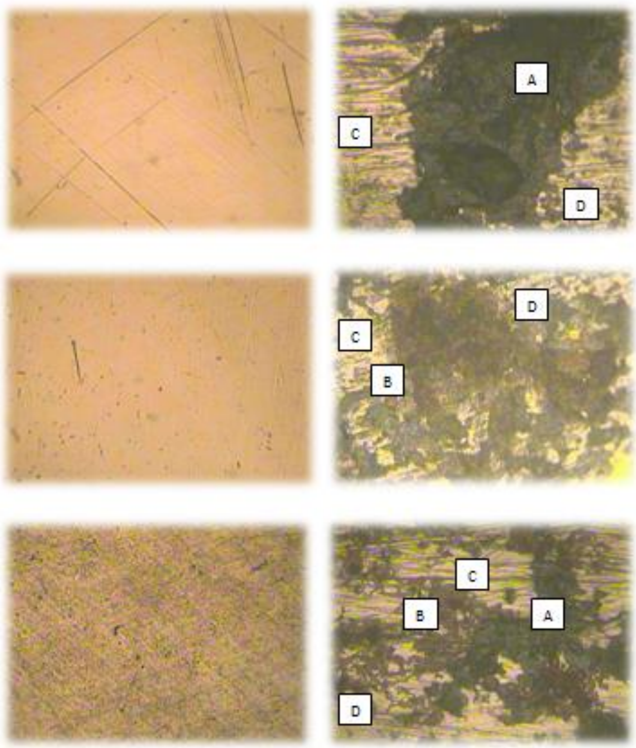

Fig. 7. Corroded metal surfaces of metals

The analysis of the corroded surfaces was based upon the visible features of such corrosive compounds foremost of the color. By using the unique identification features of such compounds there were distinguished some of specific corrosion compounds that also explained under the corrosion processes due to the different corrosive compounds. The visible observations and their linkage with the existing observations are given in the Table 6 [1, 3, 4, 5, 6, 9, 12, 13, 14, 15].

- A- Ferrous Sulfide (FeS)
- B- Ferrous Oxide (Fe_2O_3)
- C- Corrosion Cracks
- D- Cavities

Table 6: Visible appearances and their physical properties

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.

According to the existing explanations and the observations that it is possible to conclude the formations of the ferrous sulfides (FeS) on most of metal surfaces because of the oxidizing impact of corrosive compounds of crude oils, formations of the ferrous oxides (Fe_2O_3) or the rusty corrosion due to the impact of composed water in crude oils and dissolved oxygen, corrosion cracks and cavities due to the decay of metals which is theoretically explained in the next subsections.

As some of special observation it was observed black color corrosion compounds in Monel metal surfaces that similar with the features of ferrous sulfides (FeS) although impossible to conclude as CuS only considering the visible features. Therefore, as a future research work it is much adequate to recommend some compositional analysis of the corrosion compounds by the advanced analytical method such as X-ray diffraction (XRD) for better results.

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

Table 7: Decayed metallic elemental concentrations from metals 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	Murban	-0.65	-
420- MN: 2.8 (Stainless Steel)	Das Blend	-0.78	-
410-MN: 1.7	Murban	-0.71	-
420-MN: 1.7 (Stainless Steel)	Das Blend	-0.79	-
321-MN:1.4	Murban	-0.44	-
304-MN:1.9 (Stainless Steel)	Das Blend	-0.17	-
Monel 400	Murban	-	10.47
	Das Blend	-	9.49

The concluded results of the obtained results have been interpreted in the Fig. 8 and Fig. 9.

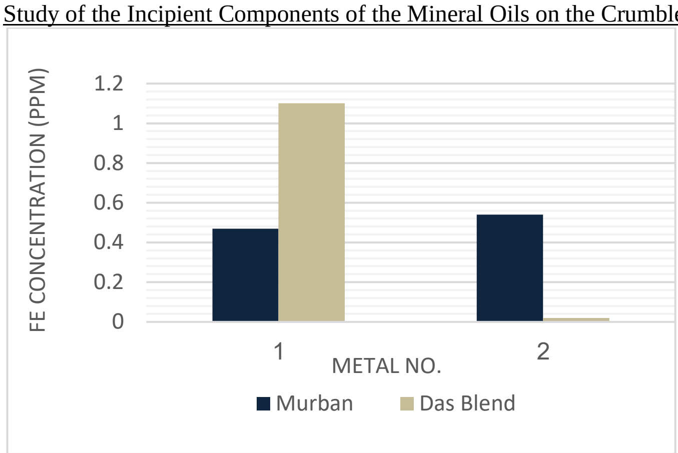

Fig. 8. Decayed ferrous concentrations from metals into crude oils

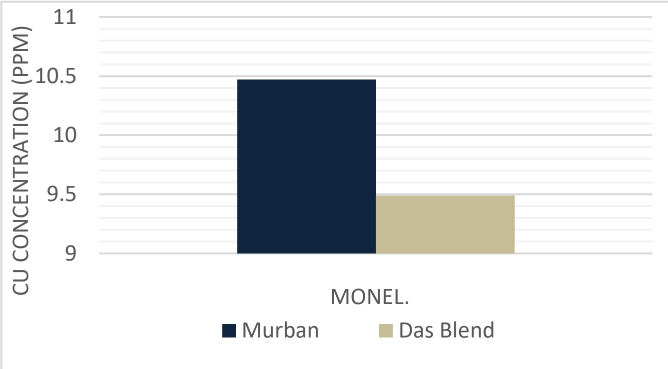

Fig.9. Decayed copper concentrations from metals into crude oils

According to the both of interpretations mainly there were observed relatively higher decays of ferrous from carbon steels also found the higher corrosion rates, significant decays of copper from Monel metal that found intermediate corrosion rates and lack of any decay if neither copper nor ferrous from any stainless steels into crude oils also found least corrosion rates in both crude oils. The decay of metallic elements is a characteristic phenomenon which is occurred during the process of corrosion that possible to explain with the theory of electron repulsive. After the formations of the corrosion compounds on the metal surfaces such compounds tend to be removed from the relevant metallic surfaces because of the repulsive and attractive forces between the successive electrons and protons of the relevant compound and metals [1,3,4,5,6,14]. Also due to the decay of matter from the metal surfaces is possible to emphasize for the reasonable evidences for the observations of the corrosion cracks and cavities that observed in the microscopic analysis.

Regarding the analysis of the variations of the initial hardness after formations of the corrosion on the metal surfaces by the Vicker's hardness tester the obtained results have been interpreted in the Fig. 10 and Fig.11.

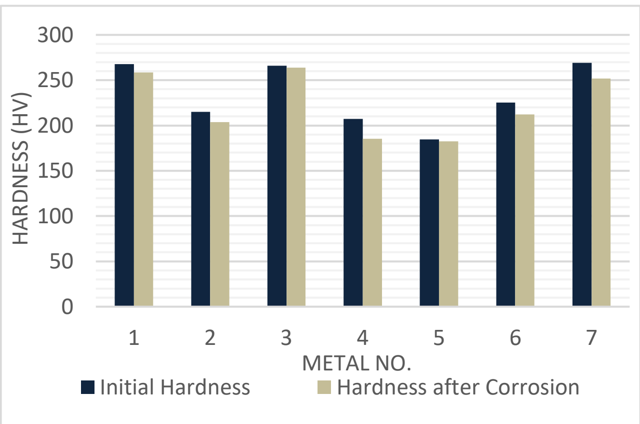

Fig.10. Variations of the initial hardness of metals in Murban

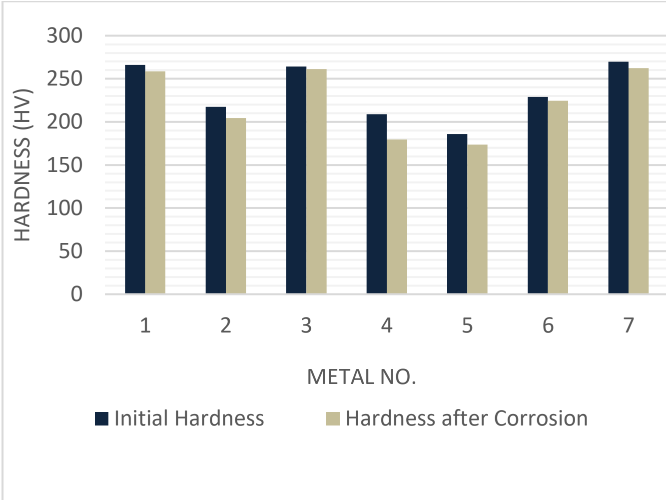

Fig.11. Variations of the initial hardness of metals in Das Blend

By considering the above interpretations slight reductions of the initial hardness were identified from most of metals after formations of the corrosion. The relevant incident is possible to be discussed under two of specific evidences as given in the below [1, 3, 4, 5, 17].

- Uncertainty of metallic surfaces on the mental surfaces due to the repulsive and attractive forces between the successive electrons and protons of the metals and relevant compounds
- Heterogeneity of the corroded surfaces because of the formations of the corrosion compounds and the partially removing of such compounds

4. Conclusion

Throughout the existing research there were able to investigate the relatively higher corrosive stability of stainless steels especially with the chemical compositions of at least 12% of chromium and sufficient amount of nickel, relatively higher contribution of the salts on the corrosion of metals at the lower temperatures while having less of progress of sulfur and active sulfur compounds on the metallic corrosion at the mower temperatures, formations of the ferrous sulfides (FeS), ferrous oxides (Fe_2O_3), corrosion cracks and pits, relatively higher ferrous destructions of some metals, significant decay of copper from Monel metal and lack of any decay of metals from any stainless steels during the immersion in crude oils and also the slight reductions of the initial hardness of most of metals after formations of the corrosion on the metal surfaces.

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