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AN EXPERIMENTAL STUDY OF THE CORROSIVENESS OF PETROLEUM OILS

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Abstract: Petroleum oils are usually composed with various hydrocarbons and also various composites in different forms and phases such as the corrosive compounds which are directly affected on the metallic destruction. In this research the major attention was gone on the investigation of the impact of elemental sulfur, Mercaptans, organic acids and the salts of two different types of petroleum oils on the corrosion rates of seven different types of ferrous metals. The chemical compositions of ferrous metals and the major corrosive properties of both petroleum oils were tested by the standard methodologies and instruments. The corrosion rates of prepared similar sized metal coupons were determined by the weight loss method after certain immersion time periods in both petroleum oils. The corroded surfaces were observed by an optical microscope. Basically there were found the least corrosion rates from stainless steels, relatively higher impact from salts on the corrosion rates of metals when comparing with other corrosive compounds at the room temperatures and the formation of FeS, rarely Fe_2O_3, corrosion cracks and the cavities on the metal surfaces as the predominant corrosion compounds.
Keywords: petroleum oils, corrosiveness, ferrous metals, weight loss, decay

1. INTRODUCTION

Petroleum oils are the naturally occurring chemical compounds which are having most of hydrocarbons and some specific trace compounds apart from such hydrocarbons. Corrosive compounds are predominantly considering in the petroleum oil refining process and regarding other industrial plants because of the severe impacts on the durability of the metals. The term of corrosion is known as the formation of such metal oxides, sulfides or the hydroxides on the metal surfaces as the result of chemical or electrochemical process on the metal surface with the aid of corrosive compound. The essential requirements the metals need to be exposed to either stronger oxidizing agent or some certain medium which is consisted with both water and oxygen [1–6]. According to the chemical compositions of crude oils those are highly composed with various corrosive compounds apart from the hydrocarbons such as the elemental sulfur, active sulfur compounds, organic acids and the salts also the impact of such corrosive compounds have been investigated and emphasized in previous studies with respect to various purposes [3–15].

In the existing research there were expected to investigate the impact of elemental sulfur, Mercaptans, organic acids and salts on the corrosion of seven different types of ferrous metals which are used in the industry of crude oils refining. Under the current studies basically there were expected to investigate the nature of the corrosion of ferrous metals in crude oils, the comparison of the strengths and progresses of the corrosive properties of crude oils and the stability of the types of metals against the corrosive environments [1–21].

2. MATERIALS AND METHODOLOGY

By considering the availability and the corrosive compounds of crude oils two different types of crude oils were selected that namely as Murban and Das Blend. According to the chemical composition of Das Bend crude oil it is known as a "sour" crude oil because of the relatively higher sulfur content of that crude oil which a key factor for the corrosion [2], [9], [15], [21]. The elementary corrosive properties of both Murban and Das Blend crude oils were tested by the standard methods and instruments as explained in the Table 1.

Table 1. Analysis of the corrosiveness of petroleum oils

Property	Method	Readings
Sulfur content	Directly used 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

As the next stage seven different types of ferrous metals were selected by considering the applicability of such metals in the industry of crude oils refining. The typical applications of such metals have been given in the below:

(1) Carbon Steel (High) – Transportation tubes, storage tanks
(2) Carbon Steel (Medium)– Storage tanks
(3) Carbon Steel (Mild Steel)– Storage tanks
(4) 410–MN: 1.8 420–MN: 2.8 (Stainless Steel)– Heat exchangers, pre heaters
(5) 410–MN: 1.7 420–MN: 1.7 (Stainless Steel)– Crude distillation columns
(6) 321–MN:1.4 304–MN:1.9 (Stainless Steel)– Crude distillation columns
(7) Monel 400 – Pre heaters, heat exchangers

The chemical compositions of such metals were detected by the XRF detector as percentages of composite metals and some of nonmetals excluding carbon.

A batch of similar sized metals coupons was prepared from selected metals as six metal coupons from each metal type and forty two metal coupons from seven types of metals. The initial weights of each metals coupon and the dimensions of each metals coupon were measured y in order of electronic balance and the micrometer. The prepared metal coupons have been shown in the Figure 1.

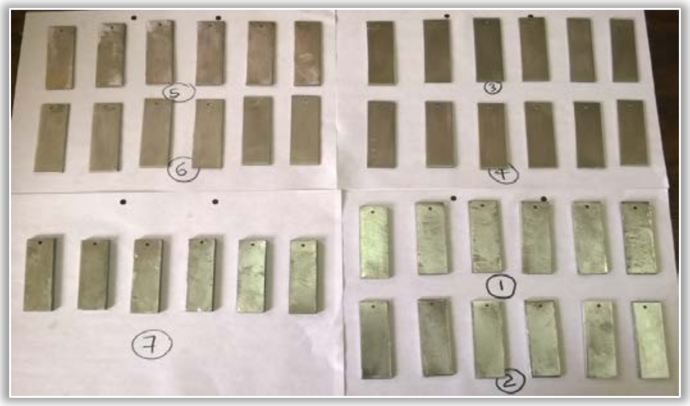

Figure 1. Metal coupons

The prepared metal coupons were immersed in both crude oil samples separately as three homogeneous metal coupons for each crude oil container as shown in the Figure 2.

Figure 2. Samples and the apparatus

After 15 days from the immersion one metal coupon was taken out from each crude oil sample as fourteen metal coupons from all of crude oil samples and the corroded surfaces of such metal coupons were observed by the 400X lens of an optical microscope. The corroded metal surfaces were cleaned by the sand papers and isooctane and also the final weight of each metal coupon was measured by the electronic balance. The corrosion rates of such metal coupons were determined by the relative weight loss method as explained in the Equation 1 [9–10].

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

where: W = weight loss 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 same procedure was performed twice again for another two similar batches of metal coupons which are remaining in the crude oil containers for the determinations of the corrosion rates of such metal coupons in order of after 30 and 45 days immersion periods.

Apart from that the decayed ferrous and copper concentrations from each metal into each crude oil sample were measured by the atomic absorption spectroscopy (AAS). According to the sample preparation methodology for that instrument 1 ml of each crude oil sample was diluted with 9 ml of 2–proponol and filtered. The decayed copper and ferrous concentrations into each crude oil sample were measured separately by using sufficient portions from each diluted crude oil sample for each element.

Finally the reduction of the initial hardness of each metal coupon was tested by the Vicker's hardness tester. As the essential measurements the initial hardness and the hardness after formation of the corrosion on the metal surface were measured by the Vicker's hardness tester.

3. RESULTS AND DISCUSSION

Accordingly the results of the chemical compositions of selected ferrous metals have been shortlisted in the Table 2.

By referring the obtained results for the chemical compositions of the selected ferrous metals that there can be seen the higher ferrous amounts in carbon steels, moderate ferrous amounts in stainless steels and the trace amount of ferrous in Monel metal. In addition there were observed the trace compositions of nickel and chromium in the stainless steels. According to the alterations of the metal manufacturing a few of advantages can be expected through this doping of trace elements with the ferrous metals as given in the below.

- Enhancements of the strength of such metals
- Reduction of the corrosive tendency of such metals

The combination of at least 12% of chromium and sufficient amount of nickel tend to be formed a self–corrosive protection layer for the relevant metal. This is an important phenomenon regarding the stability of the metals [1], [3–5], [8].

The obtained results for the elementary corrosive properties of both Murban and Das Blend crude oils have been interpreted in the Table 3.

Table 2. Chemical compositions of the ferrous 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	–	–	–	–	–	–

Table 3. Corrosive properties in both petroleum 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

When comparing the obtained results for the corrosive compounds in both crude oils there can be seen the Das Blend crude oil was composed relatively higher amounts of sulfur, Mercaptans and organic acids since the Murban crude oil was composed relatively higher amount of salts.

Salts are the foremost corrosive compounds presence in the crude oils since the occurrences of the crude oils in the forms of $NaCl$, $CaCl_2$ and $MgCl_2$. The term of salt content interprets an idea about the summation of such halides presence in a particular crude oil. When heating the slat contained system such slats tend to be broken into HCl molecules even though behave as neutral compounds. But the system is approaching to the lower temperatures such HCl molecules are reacted with the water or the moisture presence in the existing system and ultimately formed the hydrochloric acids which are known as the highly corrosive compounds [2], [7], [9]. The general chemical reactions of the salts and the ferrous metals are given in the below.

$$\mathrm{CaCl2} + \mathrm{H2O} \rightarrow \mathrm{CaO} + \mathrm{2HCl} \tag{2}$$

$$\mathrm{HCl} + \mathrm{Fe} \rightarrow \mathrm{FeCl2} + \mathrm{H2} \tag{3}$$

$$\mathrm{FeCl2} + \mathrm{H2S} \rightarrow \mathrm{FeS} + \mathrm{2HCl} \tag{4}$$

Organic acids play a dominant background in the metallic corrosion regarding the crude oils and also known as the “naphthenic acids” which are having the general formula of RCOOH. The summation of such organic acids in some particular crude oil is known as the acidity of total acid number (TAN) of such crude oil [2], [9], [12], [15]. The general chemical reactions between the organic acids and the metals are given in the below.

$$\mathrm{Fe} + 2\,\mathrm{RCOOH} \rightarrow \mathrm{Fe(RCOO)2} + \mathrm{H2} \tag{5}$$

$$\mathrm{FeS} + 2\,\mathrm{RCOOH} \rightarrow \mathrm{Fe\,(RCOO)2} + \mathrm{H2S} \tag{6}$$

$$\mathrm{Fe(COOR)2} + \mathrm{H2S} \rightarrow \mathrm{FeS} + 2\,\mathrm{RCOOH} \tag{7}$$

Sulfur is a mainly found trace composite in crude oils since the occurrences in various forms such as the elemental sulfur, hydrogen sulfides, Mercaptans, thiophenes and sulfoxides. Most of them are known as the corrosive compounds because of the higher reactivity of fraction and the functional groups of such compounds. Mercaptans are highly corrosive compounds due to the reactivity of the functional group of “RSH”. The process of the corrosion formation is depending on the nature of the functional group of the relevant compound. The corrosion process due to the presence of the active sulfur compounds is known as the “sulfidation” which is usually happening at about 230^{0}C properly and the corrosion process due to the presence of the elemental sulfur is known as the “localized corrosion” which is happened at about 80^{0}C properly. The general chemical reactions for such corrosion processes are given in the below [2], [11], [13–14], [20].

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

$$8\,\mathrm{Fe} + \mathrm{S8} \rightarrow 8\,\mathrm{FeS} \tag{9}$$

The corrosion rates of the metal coupons in order of after 15, 30 and 5 days from the immersion in both crude oils and their average corrosion rates have been given in the Table 4 and Table 5.

Table 4. 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–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 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 valuses of the corrosion rates of metals with respect to both Murban and Das Blend crude oils have been interpreted in the Figure 3.

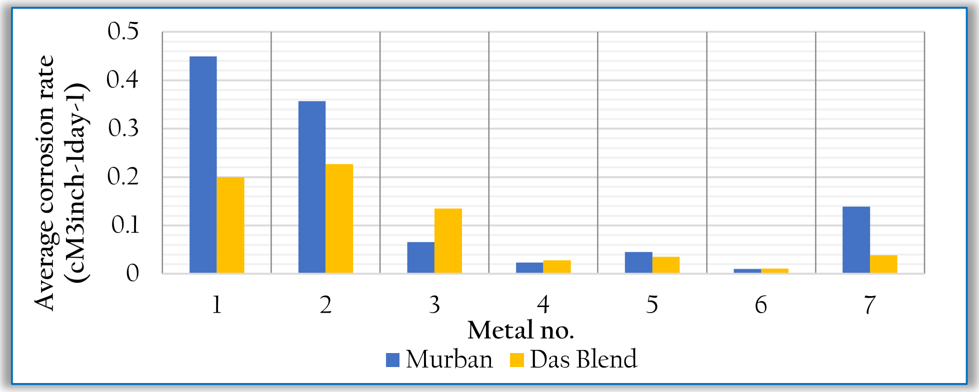

Figure 3. Average corrosion rates of metals in both petroleum oils

By observing above summarized results that there can be conclude the least corrosion rates from stainless steels, higher corrosion rates from carbon steels and moderate corrosion rates from Monel metal. When comparing the chemical composition of 321–MN: 1.4 304–MN: 1.9 (Stainless Steel) it can be seen that it was composed ~18% of chromium and ~8% of nickel which are the essential combination for the self–corrosive protection layer. Usually it is formed at least 12% of chromium with sufficient amounts of nickel. By comparing the corrosion rates and the corresponding chemical compositions of other metals that there can be concluded the high performances of the self–corrosive layer of stainless steels over the recommend chemical compositions [1], [3–5], [8].

When comparing the corrosion rates of metals with respect to both crude oils four types of metals have shown their higher corrosion rates in Murban crude oils since other three types of metals showed their higher corrosion rates in Das Blend crude oils. According to the corrosive compounds of both crude oil Das Blend crude oil was composed relatively higher elemental sulfur content, Mercaptans content and organic acid content while Murban was composed relatively higher amount of salts even though the progress of the process of sulfur and active sulfur compounds in the lower temperatures. Therefore, by considering observed results and the theoretical concepts it can be conclude the impact of salts on the metallic corrosion is greater than the impact of the organic acids on the metallic corrosion [2–18].

The variations of the corrosion rates of metal coupons with the exposure time with respect to both crude oils have been shown in the Figure 4 and Figure 5.

By referring the curves of corrosion rates of all types of metals with respect to the exposure time period against the corrosive medium that there can be concluded the similar distributions of each type of metal and also the accuracy of the weight loss method the inversely proportional relationship between two parameters of exposure time and the corrosion rates of metals for most of various materials [9–10].

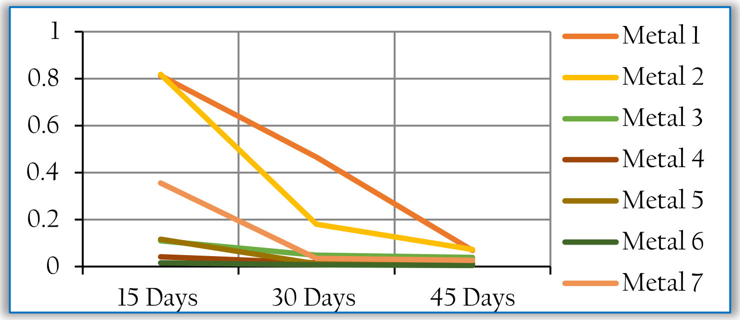

Figure 4. Variations of the corrosion rates with the exposure time in Murban

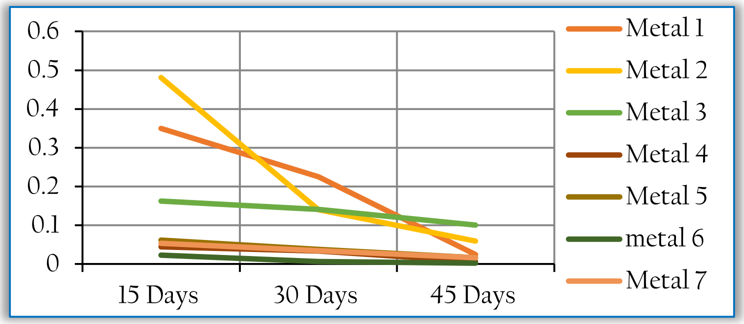

Figure 5. Variations of the corrosion rates with the exposure time in Das Blend

Figure 6. Distinguish corrosion compounds and features

According to the obtained results for the microscopic analysis of the corroded metals surfaces some specific and distinguished features have been listed and emphasized in the Figure 6.
Among the various results of the qualitative analysis of the corroded metals surfaces through the 400X lens of an optical microscope some specific features have been identified as follows.

A– Black/ brownish black compounds
B– Rusty/ reddish brown compounds
C– Corrosion cracks
D– Pits/ cavities

The comparison of the theoretical concepts regarding the corrosion compounds and the obtained results have been given in the Table 6 [1], [3–6].

Table 6. Features of major 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	Unable to specify.

According to the above comparison that there can be concluded the formation of FeS, Fe_2O_3 rarely, corrosion cracks and pitting corrosion regarding the most of observations. In addition that there were observed some black color compound on the Monel surfaces which is also similar with FeS. By the references it can be concluded as the CuS although better to recommend some advanced compositional analysis of such compounds such as X–ray diffraction (XRD) for the better analysis and the distinguishing.
The obtained results for the analysis of the decayed ferrous and copper concentrations into crude oils from the metals have been shortlisted in the Table 7.

Table 7. Decayed metal concentrations into petroleum 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 obtained results for the analysis of the decayed ferrous and copper concentrations from the metals into crude oils have been interpreted in the Figure 7 and Figure 8.

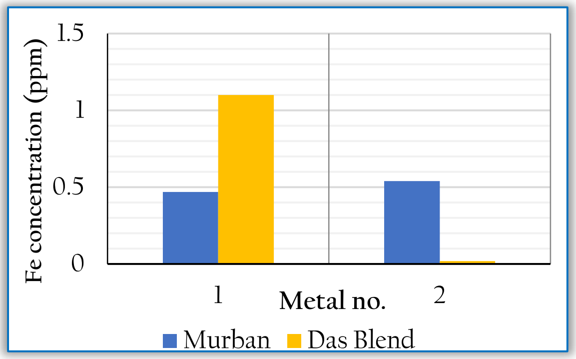

Figure 7. Decayed ferrous concentrations into petroleum oils

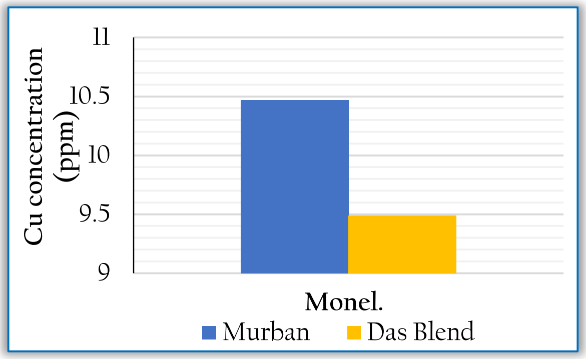

Figure 8. Decayed copper concentrations into petroleum oils

According to the obtained results for the decayed ferrous concentrations from metals into crude oils there were observed the significant decay of ferrous from both carbon steel (high) and carbon steel (medium) and also found the highest corrosion rates from those two metals in both crude oils. Also it was not found any amount of decayed ferrous from stainless steels into crude oils similar with the results of the least corrosion rates. In addition some significant decays of copper were found from Monel metal into crude oils which showed an intermediate corrosion rates in both crude oils. After formations of the corrosion compounds on the metal surfaces such compounds tend to be removed from the metallic surfaces due to the attractive and repulsive forces between the successive electrons and protons [1], [3–6]. The decay of metals into crude oils is usually happened due to this mechanism and the obtained results can be used as the confirmations of the formation of the corrosion on the metal surfaces.

The results of the variations of the initial hardness of metal coupons due to the formations of the corrosion on the metal surfaces have been interpreted with respect to both Murban and Das Blend crude oils in order of Figure 9 and Figure 10.

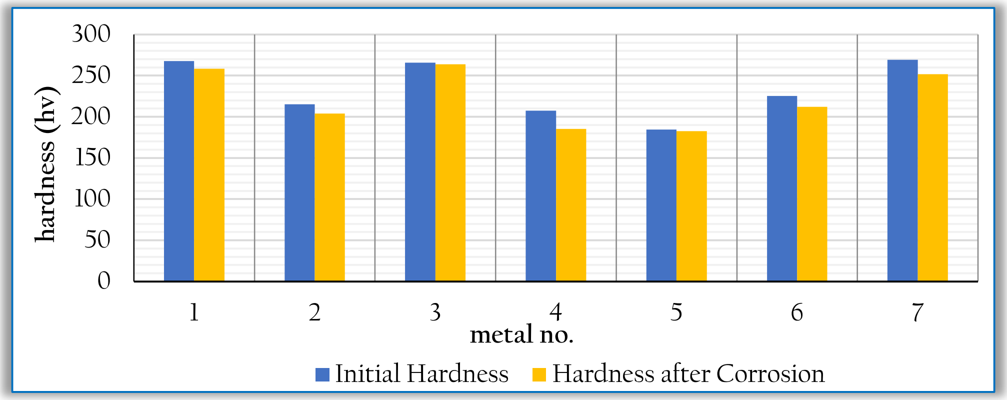

Figure 9. Deduction of the initial hardness of metal in Murban

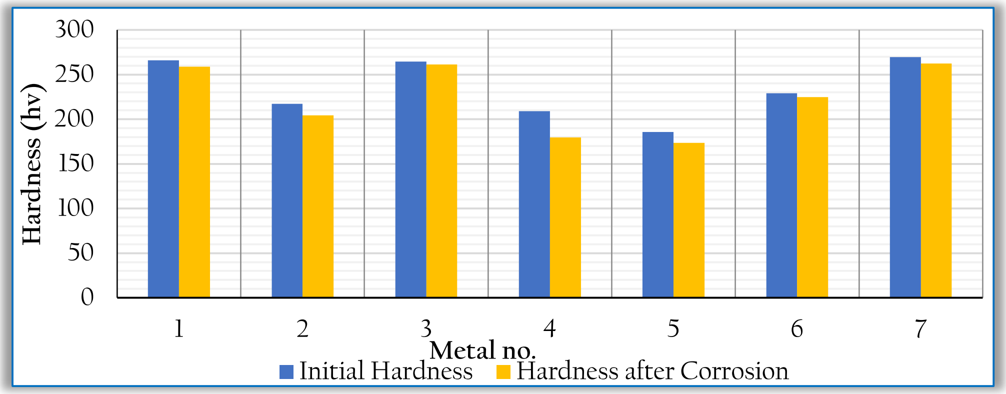

Figure 10. Deduction of the initial hardness of metal in Das Blend

The above results showed the slight reductions of the initial hardness of most of metals after the formations of the corrosion on the metallic surfaces with respect to both Murban and Das Blend crude oils. When forming the corrosion compounds on the metallic surfaces such compounds may have a less retention time period on the metal surfaces because of the impact of the repulsive and attractive forces between the successive electrons and protons of existing compounds [1], [3], [5–6]. This conditions together to create an instability on the metallic surfaces in various forms. The reductions of the initial hardness of the metals can be explained under this phenomenon.

4. CONCLUSION

As the summary of the obtained results that there can be concluded the least corrosive tendency of the stainless steels with the performances of the self–corrosive protection film of at least 12% of chromium with sufficient amounts of nickel, relatively higher impact of salts on the metallic corrosion at the lower temperatures when comparing with the impact of other elementary corrosive compounds, improper progress of the process of sulfur and sulfur compounds on the metallic corrosion at the lower temperatures, formations of FeS, Fe_2O_3, corrosion cracks and the pitting corrosion on the metal surfaces, significant decays of ferrous and copper from some of carbon steels during the interactions with crude oils and the lack of metal decay in detectably from stainless steels during the interaction with the crude oils and the slight reductions of the initial hardness of metal coupons due to the formations of the corrosion on the metallic surface itself.

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