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Research paper

The influences of the crude oils on the decay of metals

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Crude oils are the essential resources for the usages of industrial purposes in various forms and the refining is a key process of separating the mixture of raw crude oils. In the existing research there were expected to investigate the impact of salts, organic acids, mercaptans and elemental sulfur of crude oils on the corrosion rates of seven different types of ferrous metals in both qualitatively and quantitatively. The chemical compositions of such selected ferrous metals and the above mentioned corrosive properties of two different types of crude oils were measured by the standard instruments and methodologies. A set of similar sized metal coupons were prepared from seven different types of metals and the corrosion rates of such metals were determined by the relative weight loss method. In addition, that the corroded metal surfaces were analyzed under the microscope, decayed metal concentrations and deductions of the initial hardness of metal coupons were measured. According to the obtained results that there were observed the lower corrosion rates from stainless steels with at least 12% of chromium and nickel, higher corrosive impact from salt, formations of FeS, Fe_2O_3, corrosion cracks and pitting corrosion.

Introduction

Crude oils are the mixture of hydrocarbons that containing some of various traces compounds such as corrosive compounds since the occurrence. In fact, that the corrosive compounds play a considerable role in the most of metal based industries such as the crude oils refining because there are vast range of applications of metals. Usually the term of corrosion is defined as the formations of metal oxides, sulphide or the sulphide on the metal surfaces as a result of either chemical or electrochemical process. According to the mechanism of such corrosion processes the metal need to expose either strong oxidizing agent or any kind of medium which is containing both oxygen and water even in the form of moisture [1–6]. According to the chemical compositions of crude oils they are containing various corrosive compounds foremost of the sulphur com-

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pounds, salts and organic aids in various forms since the occurrence of crude oils because of the abundance of such compounds in the various interior parts of the earth. The impact of such corrosive compounds may be varied with the different physical and chemical conditions such as the temperature and concentrations [2–15].

In the existing research there were expected to investigate the impact of the salts, organic acids, elemental sulphur and mercaptans of two different types of crude oils on the corrosion rates of seven different types of ferrous metals which are applicable in the industry of crude oil refining for the various tasks under different conditions mainly under different temperature and concentrations. The major experiments were based on the analysis of corrosion of such metals as both qualitatively and quantitatively [1–18].

1. Materials and methods

According to the scope of the research and the requirements seven different types of metals were selected as the samples which are applicable in the industry of crude oil refining at around different units as given in the below:

– Carbon Steel (High) – transportation tubes, storage tanks;

– Carbon Steel (Medium) – storage tanks, vessels;

– Carbon Steel (Mild Steel) – storage tanks, crude distillation units;

– 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-Salter.

The chemical compositions of such metals were detected by the X-ray fluorescence (XRF) detector as the percentages of each composite metal and most of nonmetals excluding carbon.

In the selections of the crude oils two different types of crude oils were selected as the fluids namely as Murban and Das Blend which are slightly different in their chemical compositions including corrosive compounds. Regarding the classifications Das Blend is known as a "sour" crude oil because of the high sulphur content of such crude oils which a predominant factor for the corrosion [2; 4; 9]. The concentrations of the foremost corrosive compounds of both crude oils were measured by the stan-

dard methodologies and instruments as discussed in the Table 1.

Table 1

Analysis of the 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

A batch of similar sized metals coupons was prepared by the seven different types of selected ferrous metals as six metal coupons from each metal type and altogether forty two metal coupons also the surfaces of such metal coupons were cleaned, weighted, measured the dimensions and observed by the optical microscope until free of any other compounds such as the corroded particles. The prepared metal coupons are shown in the Figure 1.
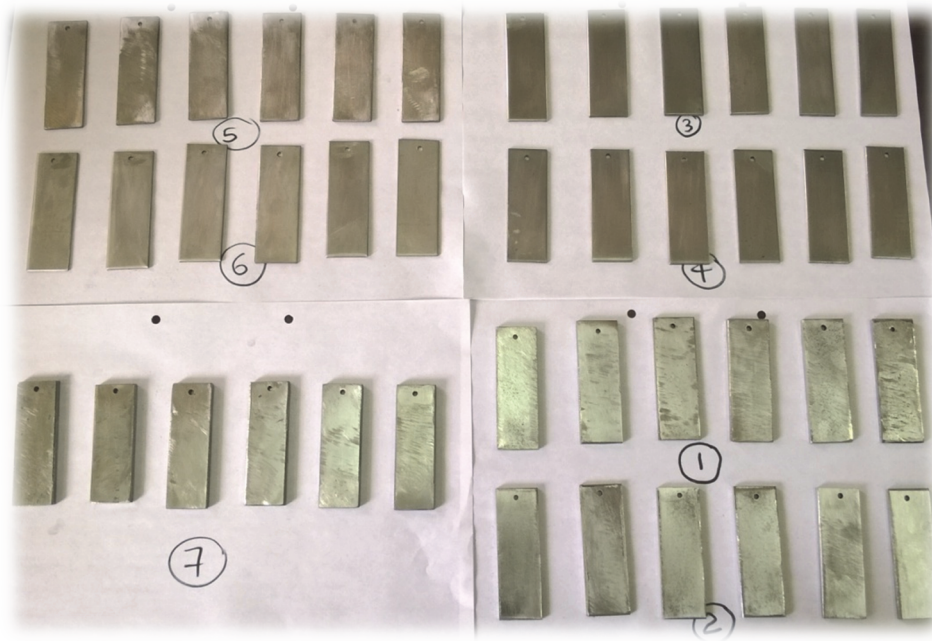
The prepared metal coupons were immersed in both crude oil containers separately as three homogeneous metal coupons per one crude oil container. The apparatus setup is shown in the Figure 2.

After the 15 days from the immersion one metal coupon from each crude oil container was taken out altogether fourteen metal coupons from all crude oil containers. The corroded metal surfaces were observed under the 400X lens of an optical microscope and the corroded metal surfaces were cleaned by isooctane and sand papers and also measured the final weight of each metal coupon. The corrosion rate of each metal was determined by the relative weight loss method as explained in the below [9; 10].

$$CR = W * k/(D * A * t),$$

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

The same procedure was repeated again twice for another two similar batches of metal coupons in order of after 30 and 45 days immersion time periods to determine the corrosion rates of such metal coupons.

Figure 1. Prepared metal coupons

Figure 2. Apparatus setup

Apart from the determinations of the corrosion rates of metals the decay of metallic elements from metal coupons during the immersion into crude oils were measured by the atomic absorption spectroscopy (AAS). According to the purpose of the current research the decayed ferrous and copper concentrations from metals in to crude oils were measured. In the sample preparation 1 ml of each crude oil sample was diluted with 9 ml of 2-propanol and filtered.

Finally, the variations of the initial hardness of metal coupons due to the corrosion of the metals were analyzed. The initial hardness and the hardness after the corrosion of each metal coupon were measured by the Vicker's hardness tester. The working principles of Vicker's hardness tester are shown in the Figure 3 [1; 3; 5].
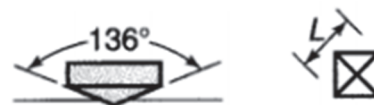
Figure 3. Indenter of the Vicker's hardness tester

$$HV = 1.854 * P^2/L^2,$$

where P – applied load on the surface of metal; L – diagonal length of square; HV – hardness.

For the one measurement of hardness at least three positions of the metal surfaces were selected randomly and measured the hardness of such points and the average values were interpreted.

2. Results and discussion

The obtained results for the analysis of chemical compositions of selected ferrous metals by the X-ray fluorescence (XRF) have been shortlisted in the Table 2.

According to the results mainly there were identified relatively higher ferrous compositions from car-bon steels, moderate ferrous concentrations in stain-less steels and trace ferrous concentrations in Mo-nel. As a special observation the trace amounts of *d*-block elements were observed from stainless steels such as nickel and chromium. The doping of trace metals into the major raw metal is based on some enhancements of the essential properties of those metals such as the improvements of the strength and the reduction of the corrosion [1; 3–6].

The obtained results for the analysis of major corrosive properties of both crude oils have been interpreted in the Table 3.

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	–	–	–	–	–	–

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

According the obtained results the Das Blend crude oils was composed relatively higher amounts of sulphur, mercaptans, organic acids and relatively lower amount of salts. But according to the most of related researches there were investigated the impact of such compounds may not depend only on the amount or the concentrations and they are highly sensitive in the environmental conditions as well such as the temperature. Therefore, in the discussions of the impacts of such corrosive compounds on the metallic corrosion the supportive conditions also mandatory to be mentioned with the observations.
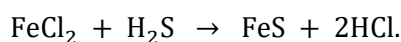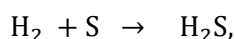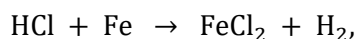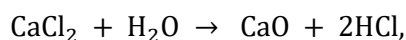
Organic acids are the major corrosive compounds that presence in the crude oils since the occurrences also known as the naphthenic acids which are having a formula of RCOOH. The total acid content of some crude oil is known as the acidity or total acid number (TAN) of such crude oil [2; 4; 9; 12; 15].

$$Fe + 2RCOOH \rightarrow Fe(RCOO)_2 + H_2,$$

$$FeS + 2RCOOH \rightarrow Fe(COOR)_2 + H_2S,$$

$$Fe(COOR)_2 + H_2S \rightarrow FeS + 2RCOOH.$$
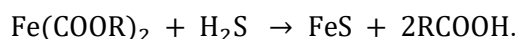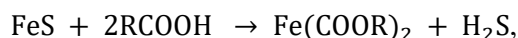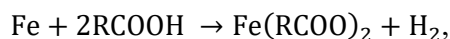
Salts are the dominant corrosive compounds found in crude oils since the occurrences because of the abundance of the interior part of the earth. Basically, such

salts are found from crude oils in the forms of NaCl, $MgCl_2$ and $CaCl_2$ and also the total amount of such halides presence in some crude oil is known as the salt content of that crude oil. When increasing the temperature of crude oils such salts tend to be broken into HCl molecules although show some inert properties at that occasion. But later on with the reductions of the temperature such HCl molecules react with water and even moisture presence in crude oils and tend to form hydrochloric acid. It is highly corrosive compound because of the strong oxidizing ability of that and on the other hand it is a strong acid. The general chemical reactions for the corrosion process of metals due to the slats are given in the following chemical reactions [2; 4; 7; 18].

$$CaCl_2 + H_2O \rightarrow CaO + 2HCl,$$

$$HCl + Fe \rightarrow FeCl_2 + H_2,$$

$$H_2 + S \rightarrow H_2S,$$

$$FeCl_2 + H_2S \rightarrow FeS + 2HCl.$$

Sulphur is an element that abundance in the interior part of the earth and possible to find from most of various crude oils in different forms such as the elemental sulphur, sulfoxides, thiophenes, mercaptans and hydrogen sulphides. Most of them are corrosive compounds and their corrosion process also may be varied with the functional groups and reactivity of that functional groups. The corrosion process due to the elemental sulphur is known as the "localized corrosion" which is occurred at about 80 °C properly and formed FeS as the major corrosion compound. Mercaptans are the active sulphur compounds that presence in crude oils which are having a molecular formula of "RSH" and the corrosion process due to the mercaptans is known as the "sulfidation" also happened in the temperature range between 230–460 °C. The initiations of above corrosion process is normally happened with the contributions of water or even moisture to produce the sulphuric acid or hydrogen sulphides which are corrosive compounds and strong oxidizing agents. The general chemical reactions for above processes are given in the following equations [2; 4; 13; 14; 16; 17].

$$S_8(s) + 8\,H_2O\,(l) \rightarrow 6\,H_2S\,(aq) + 2\,H_2SO_4\,(aq),$$

$$8\,Fe + S8 \rightarrow 8\,FeS.$$
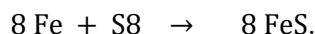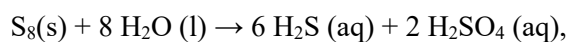
By considering the progress of corrosion of above corrosion reactions it is impossible to concluded or estimate the overall corrosive impact of all corrosive compounds because the required conditions are dissimilar and their might be affected some various corrosive compounds on the metallic corrosion which were not investigated in the current research.

According to the determinations of the corrosion rates of metals in order of after 15, 30 and 45 days from the immersion time have been interpreted in the Tables 4 and 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-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 in Das Blend

Metal	Corrosion rate after 15 days (cm³ inch⁻¹ day⁻¹)	Corrosion rate after 30 days (cm³ inch⁻¹ day⁻¹)	Corrosion rate after 45 days (cm³ inch⁻¹ day⁻¹)	Average corrosion rate (cm³ 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

As the summary of above interpreted results the average corrosion rates of metal coupons with respect to crude oils have been shown in the Figure 4.
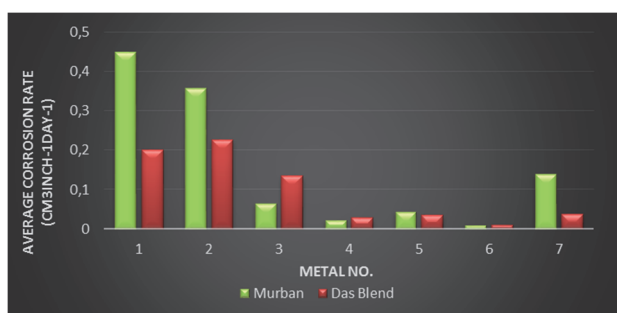

Figure 4. Average corrosion rates of metals

According to the above conclusion there were observed higher corrosion rates from carbon steels, moderate corrosion rates from Monel metal and least corrosion rates from stainless steels. Among the lower corrosion rates of stainless steels, the least corrosion rates were observed from 321-MN: 1.4 304-MN: 1.9 (Stainless Steel) with respect to both crude oils. When considering the chemical composition of that metal it was composed ~18% of chromium and ~8.5% of nickel. According to the combination of both nickel and at least 12% of chromium tends to be created some self-corrosive protection film on the metal surfaces and it is able act as a barrier of corrosion. Therefore, with the obtained results that it can be concluded the higher performance of the corrosion protection when having the higher amount of chromium with sufficient amount of nickel [1; 3; 5; 6; 17].

When comparing the corrosion rates of metals with respect to the crude oils four types of 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. The comparison of the corrosive strengths of crude oils and corrosion rates of metals in both crude oils it seems the corrosive impact of salts is high at the room temperatures because of the less progressiveness of sulphur compounds at the lower temperatures. By referring the basic results it is possible to recommend to be continued even the same methodology for the investigation of the more different corrosive compounds in various crude oils and also it is better to perform the experiments to investigate the impact of such corrosive compounds at different temperatures for the further research works for more advanced results [2; 4; 9; 12; 15; 18].

The variations of the corrosion rates of metals with the exposure time in both crude oils have been interpreted in the graphs which are shown in the Figures 5 and 6.
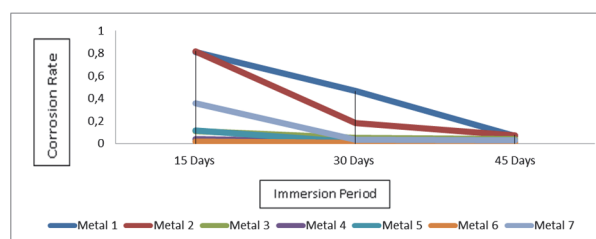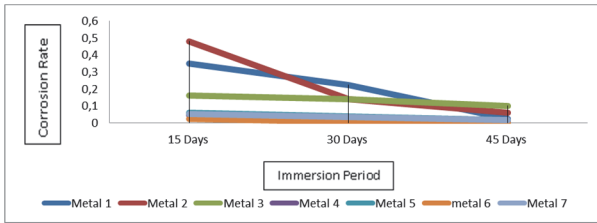

Figure 5. Variations of the corrosion rates of metals with the exposure time in Murban

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

By referring the above variations that it is possible to conclude the applicability of the weight loss method for the various kinds of materials because in here there were observed approximately similar curves for the corrosion rates of each metal. In each distribution it can be found the inversely proportional relationship between the two parameters of the corrosion rate and the exposure time [9; 10].
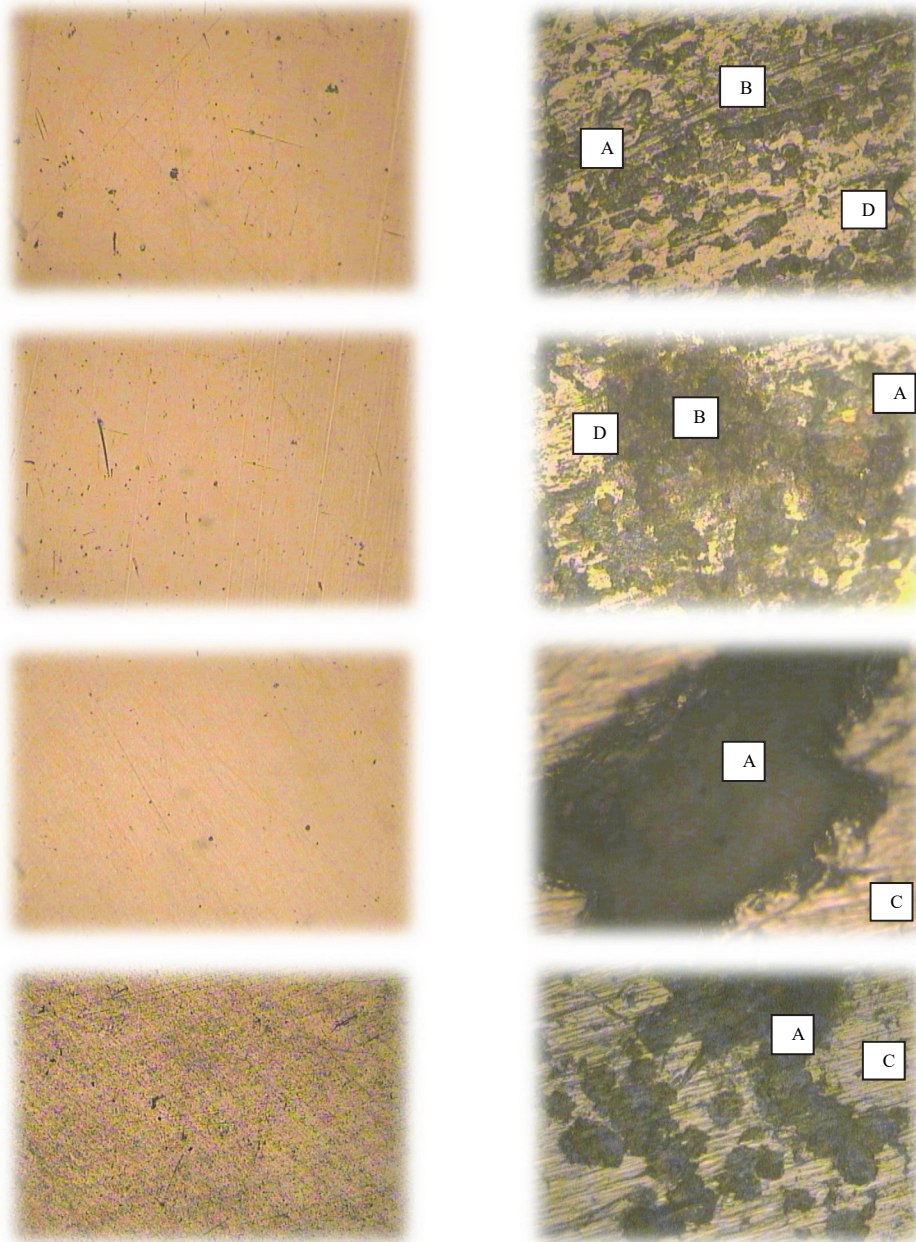
Figure 7. Corroded metal surfaces that observed under the microscope:
A – ferrous sulphide (FeS); *B* – ferrous oxides (Fe₂O₃); *C* – corrosion cracks; *D* – pits and trace compounds

According to the microscopic analysis of the corroded metal surfaces simultaneously with the determinations of the corrosion rates of metals the specific observations have been shown in the Figure 7.

Among such results some of special observations have been discussed with the absolute features of the observed features in the Table 6 [1; 3– 6; 17].

Table 6

Visible appearances of the corrosion compounds

Compound	Appearances	Observations
FeS	Black, brownish black, property of powder, pitting, cracks	Observed most of features in each metal piece
Fe₂O₃	Rusty color	Observed rarely.
CuS	Dark indigo/dark blue, property of powder	Unable to specify

Table 7

Decays of the metallic elements 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: 1.8 420-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

Regards the explanations and comparisons between the features of corrosion compounds basically it is possible to conclude the formations of some specific corrosion compounds during the current experiment such as the FeS, Fe₂O₃, corrosion cracks and pitting corrosion on the metal surfaces. In addition, that a black color compounds which is similar to FeS was indicated on the Monel metal surface although impossible to conclude as CuS only having visible features and better to recommend some advanced com-

positional analysis method for the analysis of corrosion compounds such as X-ray diffraction (XRD).

The obtained results for the experimental analysis of the decayed ferrous and copper from metals into crude oils by the atomic absorption spectroscopy (AAS) have been interpreted in the Table 7.

The obtained results for the analysis of decayed ferrous and copper amounts from metals into crude oils by the atomic absorption spectroscopy (AAS) have been shortlisted and interpreted in the following graphs that shown in the Figures 8 and 9.
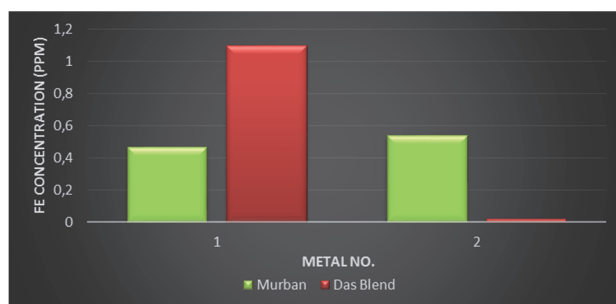

**Figure 8.** Decayed ferrous concentrations from metals into crude oils

Figure 9. Decayed copper concentrations from metals into crude oils

The above graphs showed some significant decay of copper from Monel metal and relatively higher decay of ferrous from carbon steels into both crude oils during the immersion. As special observations there were not observed any decay concentration of metal from any stainless steel into crude oils also found least corrosion rates from stainless steels. After the formations of the corrosion compounds on the metal surfaces such corrosion compounds tend to be removed from the relevant metal surfaces either partially or completely because of the repulsive and attractive forces between the successive electrons and protons of relevant compounds [1; 3– 6]. Usually the corrosion compounds are the metal oxides, sulphides and other possible compounds. Therefore, the decay of metals into crude oils is feasible to happen due to the metallic corrosion.

According to the analysis of the variations of the initial hardness of metals by the Vicker's hardness tester the obtained results have been shortlisted in the Figures 10 and 11.
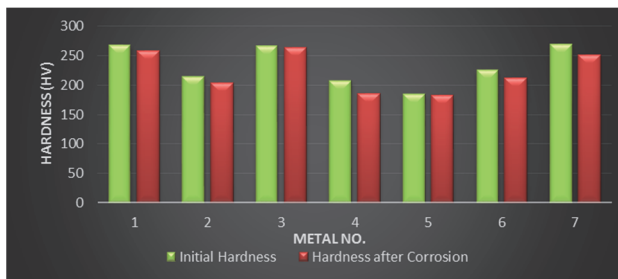
Figure 10. Variations of the initial hardness of metals in Murban
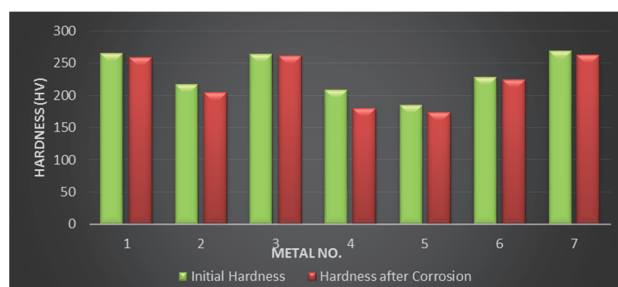
Figure 11. Variations of the initial hardness of metals in Das Blend

Those results showed slight reductions of the initial hardness of most of metals after the formation of the corrosion on the metal surfaces. The incident of the reductions of the initial hardness is possible to explain with the theory of electron repulsive because after the formations of corrosion compounds on the metal surfaces such corrosion compounds tend to be removed from the metal surfaces while creating unstable occasions on the metal surfaces because of the repulsive an attractive forces between the successive electrons and protons of existing compounds and also it is impossible to find a homogeneous metal surface after the formation of the corrosion compounds on the metal surfaces since they may be removed either partially or completely from the metal surfaces [1; 3; 5; 6]. Therefore, the initial conditions on the metal surfaces may be varied. As a result of the formation of the different corrosion compounds on the metallic surfaces, the most outer layer of the metal would be a heterogeneous one. The deduction of the initial hardness of might be caused due to the heterogeneity. However, the authentic reason for the reduction of the initial hardness must be investigated through some advanced material engineering theoretical explanations and experiments

Conclusion

As the important investigations of the existing experimental study that there were obtained and it is possible to conclude after analysis of the results it is emphasized the relatively lower corrosion tendency of stainless steels against the petroleum oils because of the self-corrosive protection film of the stainless steels when having at least 12% of chromium with sufficient amount of nickel, considerable progress of the process of corrosion regarding the salts when comparing with the impact of other corrosive compounds, irregular distributions of the corrosion, formations of the FeS, Fe_2O_3, corrosion cracks and pitting due as the corrosion compounds, destruction of the copper and ferrous from some of metals into petroleum oils and the deductions of the initial hardness of the metal coupons due to the formations of the corrosion compounds on such metal surfaces.

Recommendations for Future Improvements and Research Works: implementation of an appropriate experiment to measure or indicate the reduction of the hardness that associated with material engineering of friction science.

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Научная статья

Влияние сырой нефти на разрушение металлов

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Сырые нефти являются основным ресурсом для использования в промышленных целях в различных формах, а их переработка – ключевой процесс разделения смеси сырых нефтей. В проведенном исследовании предполагалось изучить влияние солей, органических кислот, меркаптанов и элементарной серы сырых нефтей на скорость коррозии семи различных типов черных металлов как в качественном, так и в количественном отношении. Химические составы отобранных черных металлов и вышеупомянутые коррозионные свойства двух различных типов сырой нефти измерялись с помощью стандартных приборов и методик. Набор металлических купонов одинакового размера был приготовлен из семи различных типов металлов, скорости коррозии металлов определялись методом относительной потери веса. Кроме того, под микроскопом анализировались корродированные металлические поверхности, измерялись концентрации распадающихся металлов и вычеты начальной твердости металлических купонов. Согласно полученным результатам, наблюдаются более низкие скорости коррозии купонов из нержавеющих сталей с содержанием хрома и никеля не менее 12 %, более высокое коррозионное воздействие – со стороны солей, образования FeS, Fe₂O₃, коррозионных трещин и питтинговой коррозии.

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