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GRADUATE RESEARCH ARTICLE

Study of the Effect of Crude Oils on the Metallic Corrosion

Suresh Aluvihara[1*], Jagath K. Premachandra[2]

[1] Department of Science and Technology, Uva Wellassa University, Badulla, Sri Lanka.
[2] Department of Chemical and Process Engineering, University of Moratuwa, Katubedda, Sri Lanka.

*Corresponding Author email: sureshaluvihare@gmail.com

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Student(s)

- Suresh Aluvihara

Academic Year: 2016-17
Course Level: Bachelor
Course Name: B.Sc.
Course year: 4th Year

Mentor(s)

- Jagath K. Premachandra

ABSTRACT

Corrosion is a severe matter regarding the most of metal using industries such as the crude oil refining. The formation of the oxides, sulfides or hydroxides on the surface of metal due to the chemical reaction between metals and surrounding is the corrosion that highly depended on the corrosive properties of crude oil as well as the chemical composition of ferrous metals since it was expected to investigate the effect of Murban and Das blend crude oils on the rate of corrosion of seven different ferrous metals which are used in the crude oil refining industry and investigate the change in hardness of metals. The sulfur content, acidity and salt content of each crude oil were determined. A series of similar pieces of seven different types of ferrous metals were immersed in each crude oil separately and their rates of corrosion were determined by using their relative weight loss after 15, 30 and 45 days. The corroded metal surfaces were observed under the microscope. The hardness of each metal piece was tested before the immersion in crude oil and after the corrosion with the aid of Vicker's hardness tester. The metallic concentrations of each crude oil sample were tested using atomic absorption spectroscopy (AAS). The Das blend crude oil contained higher sulfur content and acidity than Murban crude oil. Carbon steel metal pieces showed the highest corrosion rates whereas the stainless-steel metal pieces showed the least corrosion rates in both crude oils since that found significant Fe and Cu concentrations from some of crude oil samples. The mild steel and the Monel showed relatively intermediate corrosion rates compared to the other types of ferrous metal pieces in both crude oils. There was a slight decrease in the initial hardness of all the ferrous metal pieces due to corrosion.

Keywords: Crude oils, Corrosive properties, Ferrous metals, Corrosion, Weight loss method, Hardness

1 Introduction

Corrosion is a natural phenomenon that usually found in most of industries related with the typical applications in a few of tasks of ferrous metals such as the industry of crude oil refining. As essential components of the corrosion the metal need to exposure either some oxidizing agent that stronger than the Fe^{2+} or any environment which is consisted with water and oxygen and the corrosion process is modified by the organic acids and salts presence in the surrounded environment [1]-[5]. According to the general definitions regarding the corrosion it's known as the destruction of materials as results of either chemical or electrochemical reaction between materials and the surrounding environment. Also regarding the metallic corrosion, a process chemical oxidizations has been found as the way of destruction the metal surface with the formations of metal sulfides, oxides or hydroxides on the metal surface itself [2]-[12]. The metallic corrosion can be divided into a few of sub categories such as the general corrosion or rust, pitting corrosion, galvanic corrosion and the stress corrosion based on their features and their chemical compositions [1]-[3]. The surrounded environment plays a major role of the metallic corrosion because of the consistency corrosive properties of such environment. According to the chemical compositions of crude oils usually it's known as a mixture of hydrocarbons also found elemental sulfur, active sulfur, organic acids and salts in trace amounts and which are known as the compounds that more prone on the metallic corrosion [4]-[15]. There have been done most of researches and experiments recently and previously to investigate the effect of such corrosive properties on the metals, strength of such properties at different environmental conditions such as the temperature and the prevention or deduction options of the metallic corrosion such as the introductions of less corrosive alloy metals which are having some specific chemical compositions and doping with some trace elements [1]-[15]. In the current research there were expected to speculate the stability of the stainless steels against the corrosion of some ferrous metals which are used in the crude oil refining industry due to the effects of crude oils when comparing with other metals, the contributions of elemental sulfur, active sulfur compounds, organic acids and salts on the metallic corrosion at the normal environmental conditions, the identifications and analysis of the formed corrosion compounds under such processes based on their visible appearances and investigations of the changes of some surface properties of those metals such as the hardness.

2 Materials and Methodology

2.1 Materials

2.1.1 Metals

Seven different types of ferrous metals were selected as the samples that including three different types of carbon steels, three different types of stainless steels and Monel which has a trace amount of ferrous while having copper and nickel as the majority [1] [3] [6]. Those metals are used in the essential purposes in the crude oil refining industry as given in the below.

- Carbon Steel (High) – Transportation tubes
- Carbon Steel (Medium)- Storage tanks
- Carbon Steel (Mild Steel)- Storage tanks
- 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

2.1.2 Crude Oils

Two different types of crude oils were selected as the samples. Those are Murban and Das Blend which are slightly or extensively different and also off- time used in the crude oil refining industry in Sri Lanka. Usually Das Blend is known as a "sour" crude oil because of the high sulfur content of that crude oil and Murban is mediocre crude oil which is used in a few refineries of the world [2].

2.2 Methodology

The procedure of the entire research has been subdivided into several categories based on the performed order of such experiments.

2.2.1 Determinations of the Corrosive Properties of Crude Oils

The corrosive properties of both crude oils were investigated by the standard test methods and instruments. A brief description of the determinations of the corrosive properties of crude oils is given in the Table 1.

Table 1: *Testing procedures 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

2.2.2 Determination of the Chemical Compositions of Metals

The elemental composition of each ferrous metal was determined by the XRF detector. According to the working principles behind the XRF detector it allows to determine the percentages of the dominant metallic elements and some of non-metallic elements as well exclusive of carbon.

2.2.3 Weight Loss Method and Corrosion Rates of Metals

A batch of similar sized metal coupons was prepared from seven different types of metals as six metal coupons from each type of metal as all were in equal dimensions and their surfaces were cleaned until free of corrosion as shown in the Figure 1.

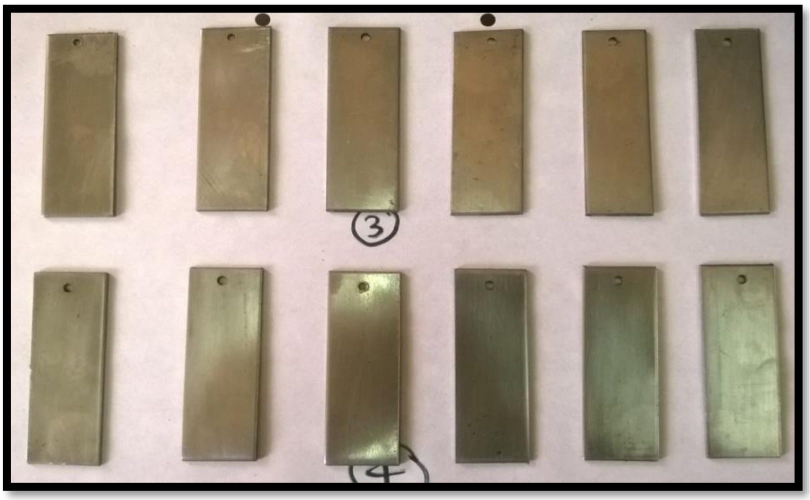

Figure 1: *Prepared metal coupons*

The dimensions and weights of all metal coupons were measured for the further requirements. The prepared metal coupons were totally immersed in both Murban and Das Blend crude oils separately according to the metal type as three homogeneous metal coupons in one crude oil container either Murban or Das Blend as shown in the Figure 2.

Figure 2: *(a) Samples and (b) set up of apparatus*

After 15 days from the immersion a metal coupon was taken out from each crude oil container and the corrosion rates of such metal coupons were determined by the weight loss method [10]. The mathematical expression and the terms of that expression have been given in the below.

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

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

The corroded metal surfaces were observed by the laboratory optical microscope. The corroded particles were removed from the metal surfaces by the sand papers and isooctane as an instant corrosive protection agent and the final weight of each metal coupon was measured by the electronic balance according to the necessity of the determination of the weight loss due to the corrosion [10].

The same procedure was repeated for another two sets of remaining metal coupons in the crude oil containers in order of after 30 and 45 days from the immersion to determine the corrosion rates of such metal coupons that same as the previous set of metal coupons.

2.2.4 Qualitative Analysis of Corrosion

As a confirmation stage of the formation of the corrosion compounds on the metal surfaces the qualitative analysis part was performed. According to the qualitative analysis the corroded surfaces of each metal coupon was observed under the 400X magnification lens of an optical microscope. The corrosion compounds were identified based on their physical and visible appearances foremost of the color and the surface properties such as the cavities**.**

2.2.5 Decay of Metals in Crude Oils

The decayed ferrous concentrations of crude oil samples which were exposed to stainless steels and carbon steels and also the decayed copper concentrations of crude oil samples which were exposed to Monel metal were measured by the atomic absorption spectroscopy (AAS) after the interacted with such metal coupons. Based on the observation of the invisible weight loss of some metal coupons after the immersion in crude oils this part was added to the scope of the experiment. According to the procedure of sample preparation for the atomic absorption spectroscopic (AAS) analysis 1 ml of each crude oil sample was diluted with 9 ml of 2-propanol and the precipitations were filtered out.

2.2.6 Testing Some Variations of the Properties of Metals

Corrosion is a surface-based phenomenon of the metals and the hardness is a foremost physical property of metal which is depended on the conditions of the metals surface and also it is much sensitive with the variations of the surface conditions of such metals [1][3]. In the current research the initial hardness and the hardness after the corrosion of each metal coupon were tested by the Vicker's hardness tester. According to the working principles of such instrument hardness were tested at least in three points at once on each metal coupon and the average value was suggested as the hardness of such metal coupon.

3 Results and Discussion

3.1 Corrosive Properties of Crude Oils

The values of the corrosive properties of both crude oils are given in the Table 2. According to the obtained results there were found higher amounts of elemental sulfur, mercaptans, organic acids and relatively lower amount of salts in Das Blend crude oil than the Murban crude oil. Acidity is a property of crude oil which meant the content of organic acids present in such crude oil and also called as naphthenic acids with the general formula of RCOOH [2] [9].

Table 2: *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

Also, this phenomenon was investigated in a few of past researches perfectly [15]. The chemical reactions of the formation of ferrous sulfides on the metal surfaces are given in the Equation 2, Equation 3 and Equation 4.

$$Fe + 2RCOOH \rightarrow Fe(RCOO)_2 + H_2 \quad \text{(Eq.2)}$$

$$FeS + 2\,RCOOH \rightarrow Fe(COOR)_2 + H_2S \quad \text{(Eq.3)}$$

$$Fe(COOR)2 + H_2S \rightarrow FeS + 2\,RCOOH \quad \text{(Eq.4)}$$

The salts in the crude oils play a dominant role in the cause of metallic corrosion. The value of the salt content of some particular crude oil denotes the accumulation of the amounts of NaCl, MgCl2 and CaCl2 present in such crude oil [7]. Due to the chemical reaction between those salts and water presence in crude oil at some higher temperatures salts tend to be broken into HCl since system approaching into those HCl will be behaved as highly corrosive compounds and tend to be formed the metal sulfides on the surfaces of relevant metals as explained in the Equation 5, Equation 6 and Equation 7.

$$CaCl_2 + H_2O \rightarrow CaO + 2HCl \quad \text{(Eq.5)}$$

$$HCl + Fe \rightarrow FeCl_2 + H_2 \quad \text{(Eq.6)}$$

$$FeCl_2 + H_2S \rightarrow FeS + 2HCl \quad \text{(Eq.7)}$$

According to the obtained results for the elemental sulfur and mercaptans contents in both crude oils Das Blend crude oil was stronger regarding the ability of corrosion than Murban crude oils. The elemental sulfur and active sulfur compounds in crude oils are much of corrosive compounds because of the reactivations of either fractions or functional groups that can be reacted with metals also called "sulfidation" [2] [6] [8]. Even though an active reaction needs some higher temperature conditions approximately more than 230 ^{0}C for the proper progress of corrosion. The corrosion caused by the elemental sulfur is called as the "localized corrosion" usually occurred at about 80 ^{0}C also investigated regarding some previous researches [11] [13]. The general chemical formula of the processes of "sulfidation" and "localized corrosion" are given in the Equation 8 and Equation 9.

$$S_{8(S)} + 8\,H_2O\,(l) \rightarrow 6\,H_2S\,(aq) + 2\,H_2SO_4 \quad \text{(Eq.8)}$$

$$8\,Fe + S_8 \rightarrow 8\,FeS \quad \text{(Eq.9)}$$

Therefore, the corrosive strength of Das Blend crude oil is higher than the corrosive strength of Murban crude oil based on the assumptions that all of corrosive processes were happened properly with the contribution of supportive conditions foremost the temperature.

3.2 Chemical Compositions of Metals

According to the results of the XRF detector the elemental compositions of the metals are given in the Table 3. According to the obtained results there were found some higher amounts of ferrous in three types of carbon steels, moderate amounts of ferrous in three types of stainless steels and trace amount of ferrous in Monel while composed higher amounts of nickel and copper. Especial three types of stainless steels were significantly composed with other trace elements including metals and non metals such as chromium, nickel and silicon based on the purposes of enhancements of the essential properties such as the hardness and the reduction of the corrosion as well [1] [3].

Table 3: *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	-	-	-	-	-	-

3.3 Corrosion Rates of Metals

The average corrosion rates of metal coupons with respect to the both crude oils are given in the Figure 2.

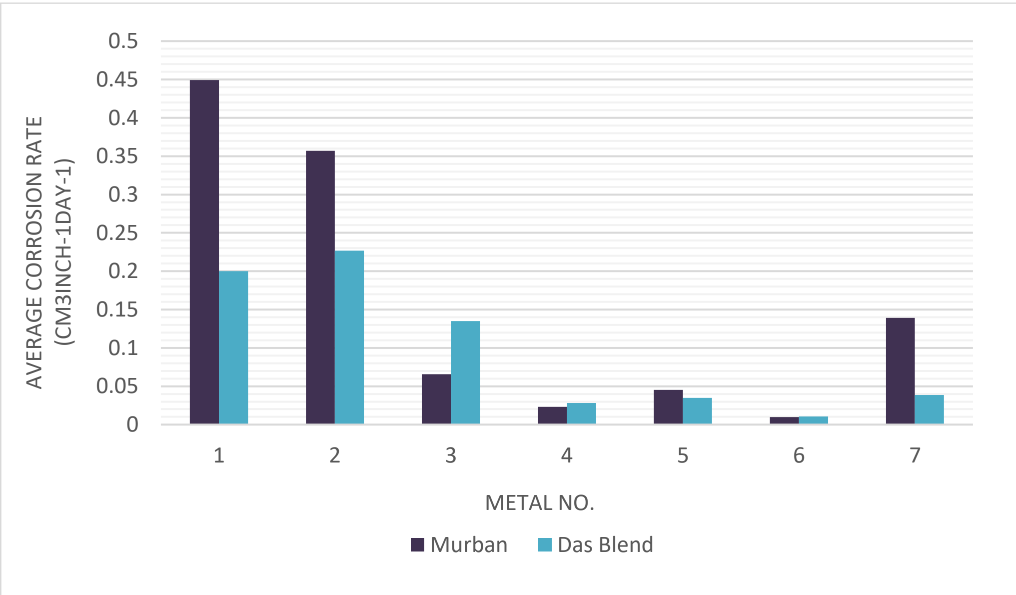

Figure 3: *Average corrosion rates of metals in both crude oils*

According to the distribution graph of the corrosion rates of ferrous metals in both Murban and Das Blend crude oils there were found the least corrosion rate from 321-MN: 1.4 304-MN: 1.9 (Stainless Steel) in both crude oils while finding relatively lower corrosion rates from three types of stainless steels, relatively higher corrosion rates from three types of ferrous metals and also a moderate corrosion rate from the Monel 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% of nickel. Regarding the chemical compositions of stainless steels at least 12% of chromium and sufficient amount of nickel tend to from a self-protection film on the

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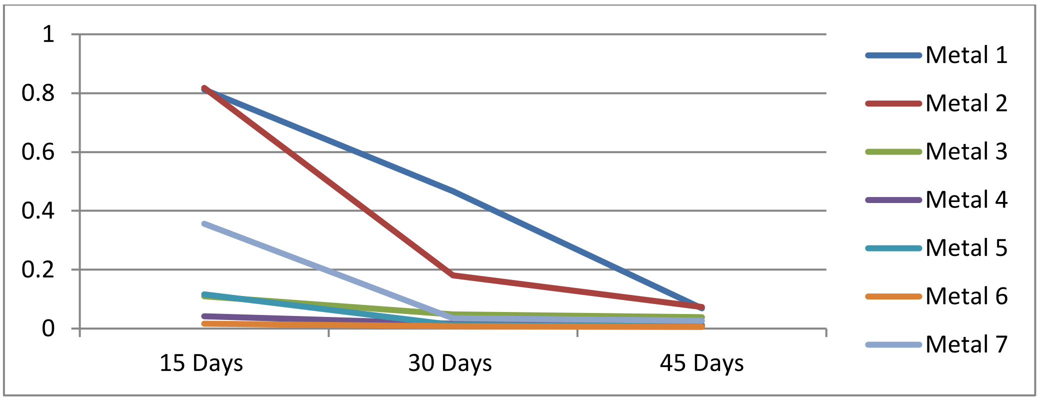
surface of the metal itself [1][4][5]. Apart from that other two types of stainless steels which are having 12% and 13% of chromium also showed some relatively lower rates of corrosion. Therefore, it can be concluded that the above self-protection film tend to be reduced the corrosion rate of a metal significantly and also the higher performances of such protection film can be expected at the recommended chemical compositions of both chromium and nickel [1] [4].

When considering the corrosion rates of metals with respect to the crude oils there were found higher corrosion rates of four types of metals out of seven types of metals in Murban crude oil than the corrosion rates of same metals in Das Blend crude oils since Das Blend was stronger in corrosive properties than Murban. There can be concluded the improper progress of sulfur and active sulfur compounds at the room temperature and also the contribution of the salts on the metallic corrosion was much stronger than the contribution of the organic acids on the metallic corrosion [6] [8] [14] [15].

The variations of the corrosion rates of metals with the exposure time with respect to both crude oils are given in the Figure 3.

(a)



(b)

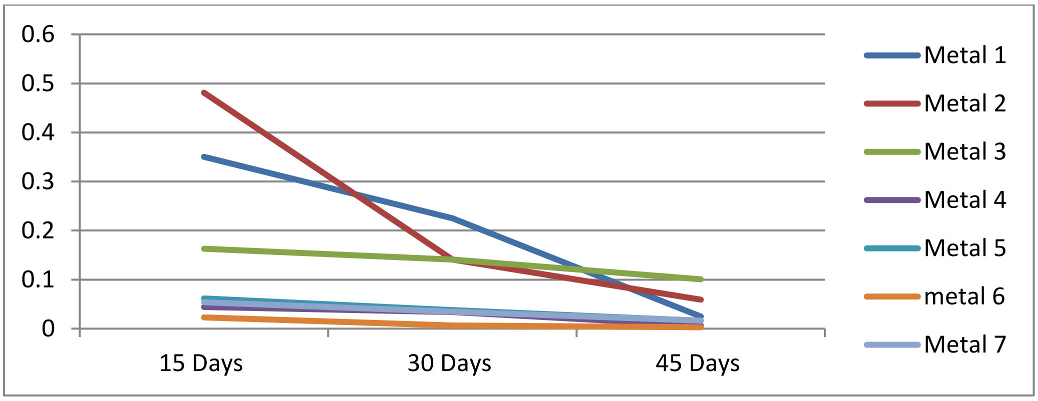


Figure 4: *Variations of the corrosion rates of metals with the exposure time in (a) Murban and in (b) Das Blend*

The variations of the corrosion rates with the exposed time in both crude oils usually found the sequence of reduction of the corrosion rate of each metal coupon with the exposed time period regarding both Murban and Das Blend crude oils. When causing the corrosion on the surfaces of metal coupons there can be suggested some self corrosive barriers will be occurred from the corroded surface on the formation of the corrosive compounds forever. There can be identified the proof of the weight loss method under the inversely proportional relationship between the corrosion rate and the exposed time period in front of the relevant corrosive surrounding [10].

3.4 Microscopic Review

Regarding the microscopic analysis of the corroded metal surfaces some of distinctive appearances of corroded surfaces of stainless steels, carbon steels and Monel are given in the Figure 4.

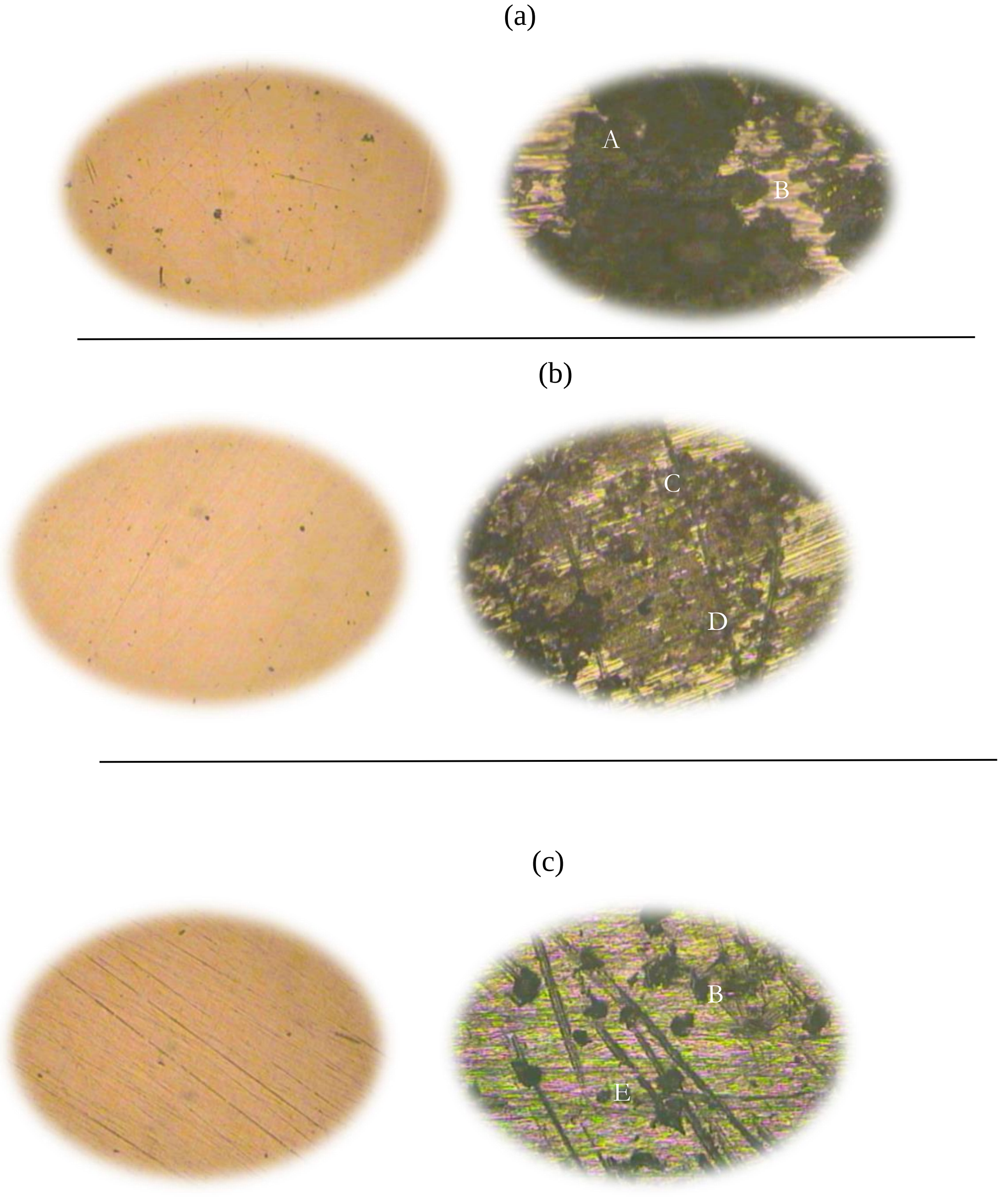


Figure 5: *Corroded surfaces of (a) 410-MN: 1.7 420- MN: 1.7 (Stainless Steel) in Murban, (b) Carbon Steel (Mild Steel) in Murban and (c) Monel in Murban*

When considering the visible features of the corroded surface under the microscope there were identified some important properties that relevant corrosion compounds that have been formed on the metal surfaces foremost the color [1][3][4]. The list of some major corrosion compounds has been given in the below.

A- Ferrous Sulfides and Trace Compounds
B- Pitting Corrosion
C- Corrosion Cracks
D- Ferrous Oxides and Trace Compounds
E- Copper Sulfides and Trace Compounds

According to the microscopic analysis there were identified some important properties of corrosion compounds since those observations can be used as the confirmation stage of the formation of corrosion in the current experiment. Some of important physical appearances of the corrosion compounds have been given in the Table 4.

Table 4: Appearances of the corrosion compounds

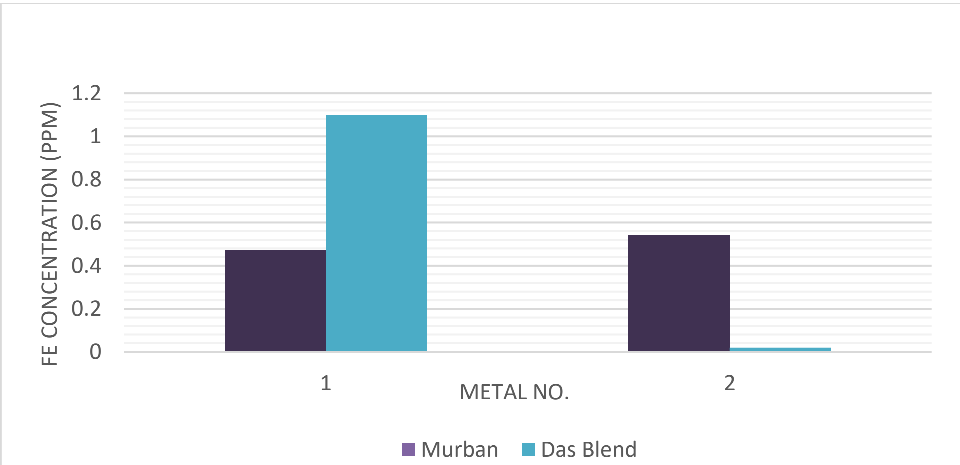
Compound	Appearances	Observations
FeS	Black, brownish black, property of powder, pitting, cracks	Observed most of features in each metal piece.
Fe_2O_3	Rusty color	Observed rarely.
CuS	Dark indigo/ dark blue	Unable to specify.

Apart from the analysis of corrosion compounds there can be concluded that the formation of pitting corrosion in most of metals and corrosion cracks especially on the surfaces of stainless steels forever. Finally it can be concluded the formation of FeS in most of observations as explained earlier and also the formations of CuS in Monel metal although difficult to distinguish only with optical appearances and required some advanced compositional analytical technique such as the X-ray diffraction (XRD) [1] [3] [6].

3.5 Metallic Concentrations in Crude Oils

The concentrations of ferrous and copper in crude oil samples which were interacted with metals are given in the Figure 5.

(a)



(b)

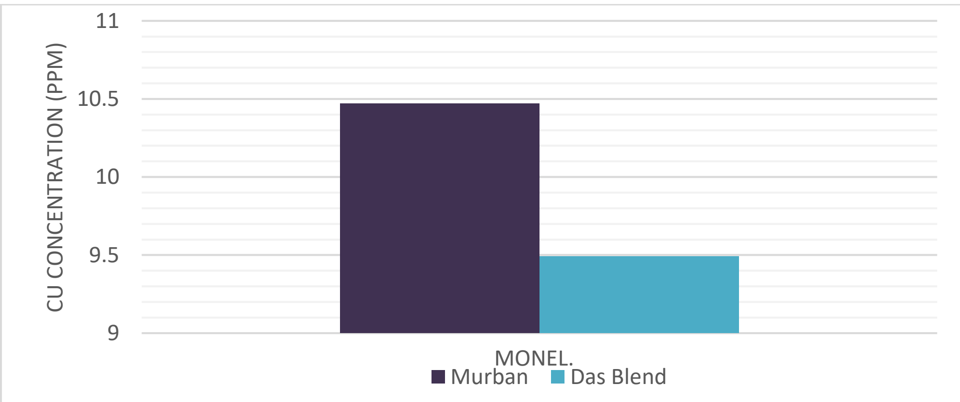


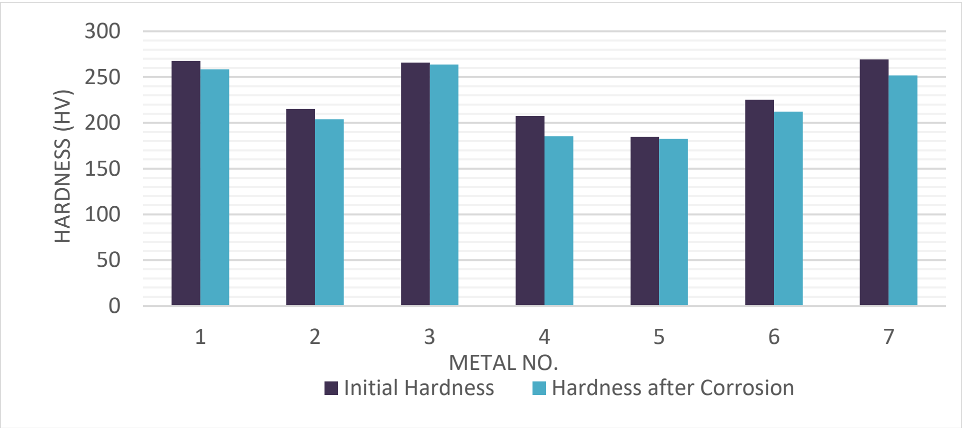
Figure 6: *(a) Ferrous concentrations and (b) copper concentrations in both crude oils*

According to the obtained results regarding the ferrous concentrations in carbon steels and stainless steels there were identified some significant concentrations of ferrous in both Murban and Das Blend crude oil samples which were exposed to high carbon steel and medium carbon steels also found highest corrosion rates simultaneously in these two types of metals and there were not found any amount o ferrous in both Murban and Das Blend crude oil samples which were exposed to neither mild steel or any type of stainless steel also showed the least corrosion rates in both Murban and Das Blend crude oils while finding relatively greater amount of copper in both Murban and Das Blend crude oil samples which were interacted with Monel metal since showing a moderate corrosion rate. The formation of metal oxides, sulfides, hydroxides or any certain compound is known as the corrosion. Due to the formation of corrosion compounds on the metallic surface it will be converted into a heterogeneous system although the diverse compounds tend to be removed from the initial metal surface with the effects of repulsive and attractive forces in between successive electrons and protons [1] [3]. The phenomenon of the invisible weight loss will be realized with this theory.

3.6 Variations of the Hardness

The initial hardness and the hardness after causing the corrosion on the surface of each metal coupon are given in the Figure 6.

(a)



(b)

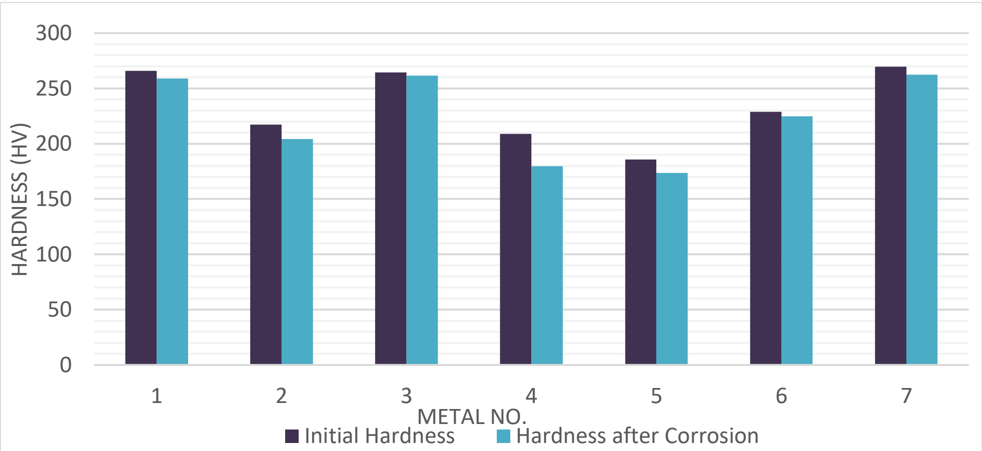


Figure 7: *Variations of the hardness of metals in (a) Murban and (b) Das Blend*

The interpretation of the values of initial hardness and hardness after the corrosion of metals there can be observed a slight reduction of the initial hardness of each metal coupon after formation of the corrosion. The hardness of a metal is a property which is depended on the surface conditions. As results of the formation of the corrosion compounds on the surfaces of metals the stability of the metal surface will be reduce due to the removing tendencies of the corrosion compounds from the metal surface [1] [2]. Therefore, it can be concluded the reduction of the corrosion was happened due to the uncertainty of the metal surface and due to the asymmetric variation of the formation of the corrosion compounds on the surfaces of metal coupons.

4 Conclusion

According to the obtained results the least corrosion rates were found from 321-MN: 1.4 304-MN: 1.9 (Stainless Steel) in both crude oils while finding relatively higher corrosion rates from carbon steels and moderate corrosion rates from Monel in both crude oils. The recent factor of the lowest corrosion rates of the stainless steels can be concluded the doping of at least 12% of chromium and a sufficient amount of nickel correspondence to that because of the formation of self-protection film against the corrosion. Some higher rates of corrosion were found regarding four types of metals out of seven in Murban crude oil than the Das Blend crude oil since Das Blend was stronger than Murban regarding the corrosive properties namely as acidity, sulfur content and mercaptans content while weaker in the content of salt. It can be concluded the improper progress of the mercaptans and elemental sulfur on the process of corrosion and higher contribution of salts on the rate of corrosion of metals than the contribution of organic acids on the metallic corrosion. The microscopic analysis interpreted and confirmed the formation of the corrosion and verified the corrosion compounds up to certain level of discussion based on the physical appearances of such corrosive compounds foremost of the color. The results of the atomic absorption spectroscopic (AAS) analysis showed some significant ferrous concentrations in both Murban and Das Blend crude oil samples which were exposed to high carbon steel and medium carbon steel also found highest corrosion rates in both crude oils since there was not found any amount of ferrous in any crude oil samples neither regarding any stainless steel or mild steel while finding higher concentrations of copper in both crude oil samples relevant to the Monel metal altogether it can be concluded the formations of FeS regarding stainless steels, carbon steels and CuS regarding Monel in most of occasions. Finally, the slight reduction of the hardness after the corrosion of each metal denoted the changes in metal surface due to the corrosion forever. There can be recommended a compositional analysis for the observed corrosion compounds during the microscopic analysis by the advanced analytical method such as the X-ray diffraction (XRD) for the better results forever.

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