Study on the Relationship between the Corrosive Nature of Crude Oils and the Corrosion of Metals.

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Abstract

Crude oils are primarily found in the interior of the earth and consist of various compounds, including corrosive composites like sulfur compounds, salts, and organic acids, which contribute to the degradation of metals. The main focus of this study was to investigate the impact of these corrosive compounds on the corrosion of seven different types of ferrous metals. The chemical compositions of the chosen ferrous metals and the corrosive properties of two different crude oils were analyzed using recommended instruments and standard methodologies. Metal coupons of the same size were prepared from the seven types of metals, and their corrosion rates were determined using the weight loss method. Additionally, the concentrations of corroded ferrous and copper were examined, and the variations in the initial hardness of the metals were measured. The findings revealed lower corrosion rates in stainless steels, higher corrosive effects from salts, the formation of FeS and Fe2O3, corrosion-related cracks and cavities on the metal surfaces, significant decay of ferrous and copper from certain metals into the crude oils, and minor reductions in the initial hardness of the metals as a result of corrosion.

Keywords: Crude oils, Corrosiveness, Metals, Decay, Weight loss, Corrosion

1. Introduction

Crude oils are essential natural resources containing a mixture of hydrocarbons and trace compounds like sulfur compounds, organic acids, and salts. Corrosion refers to the formation of metal oxides, sulfides, or hydroxides on metal surfaces due to chemical or electrochemical reactions. Corrosion typically occurs when metals are exposed to strong oxidizing compounds or environments containing water and oxygen. The corrosion process and resulting compounds can vary depending on the type of oxidizing chemicals present. Specific types of corrosion include general corrosion, pitting corrosion, thermal corrosion, and galvanic corrosion. Various chemical engineering studies and literature reviews have examined the correlations between specific corrosive compounds found in crude oils and the corrosion of different metals under various conditions. Previous research has focused on analyzing the effects of sulfur compounds including sulfoxides, thiophenes, hydrogen sulfides, and elemental sulfur. Furthermore, it has been noted that organic acids and salts are particularly corrosive compounds (Ajimotokan, Badmos & Emmanuel, 2009). "In this study, the focus was on examining the effects of elemental sulfur, mercaptans, organic acids, and two types of salts found in selected crude oils on the corrosion of seven different types of ferrous metals commonly used in the crude oil refining industry at ambient temperature. Both qualitative and quantitative analyses of corrosion were conducted as part of the investigation."

2. Materials and Methodology

In light of the criteria set forth in previous research, two distinct varieties of crude oil were chosen as the corrosive substances for the study, specifically known as Murban and Das Blend. These varieties exhibit slight variations in their chemical compositions and corrosive attributes. Das Blend typically contains a higher concentration of sulfur, making it particularly suitable for corrosion studies. The levels of elemental sulfur, mercaptans, organic acids, and salts in both types of crude oil were analyzed using the instruments and analytical techniques listed in Table 1.

Table 1. Test methodologies for 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 light of the utilization of metals in the crude oil refining industry, seven distinct varieties of ferrous metals were chosen for examination regarding their susceptibility to corrosion caused by crude oils. The chosen metals and their common uses are outlined below.

- Carbon Steel (High) – Transportation tubes, storage tanks
- Carbon Steel (Medium)- Storage tanks, transportation tubes

- Carbon Steel (Mild Steel)- Storage tanks
- 410-MN: 1.8 420-MN: 2.8 (Stainless Steel)- Heat exchangers, pre heaters
- 410-MN: 1.7 420-MN: 1.7 (Stainless Steel)- Crude distillation columns, pre heaters
- 321-MN:1.4 304-MN:1.9 (Stainless Steel)- Crude distillation columns
- Monel 400- Pre heaters, heat exchangers , de-salting units

The X-ray fluorescence detector was used to determine the chemical compositions of these metals, revealing the percentage of each metal present as well as the absence of most non-metals, with the exception of carbon. Metal coupons of various metals were prepared in identical dimensions, and their surfaces were meticulously cleaned using sandpaper and isooctane until they were devoid of any extraneous compounds. This cleaning process was conducted under the observation of a 400X lens of an optical microscope. The initial weights of these metal coupons were accurately measured using an analytical balance and duly documented. These prepared metal coupons are depicted in Figure 1.

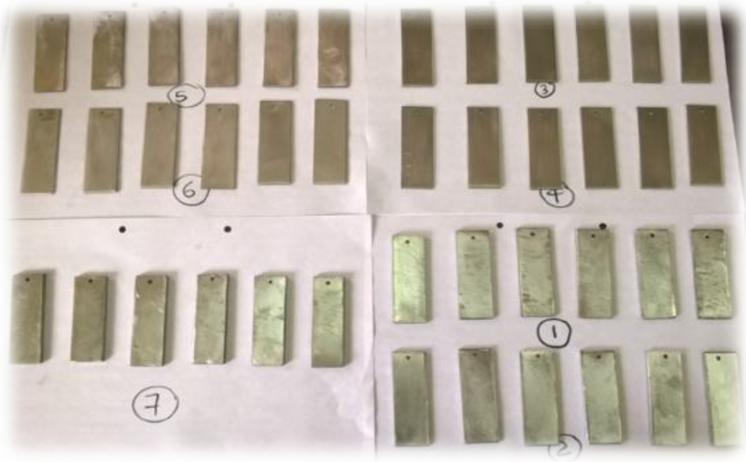

Figure 1. Prepared metal coupons

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

Figure 2. Setup of apparatus

After the 15-day immersion period, one metal coupon was retrieved from each crude oil container, and the corroded surfaces were visually examined under a 400X optical microscope for qualitative analysis. Subsequently, the corroded particles were eliminated from the metal surfaces using sandpapers and isooctane, and the final weight of each metal coupon was determined using an analytical balance to assess the weight loss. The corrosion rate of each metal coupon was then calculated using the weight loss method (Okpokwasili and Oparaodu, 2014).

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

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

The same procedure was repeated two more times to ascertain the rates of corrosion of two additional sets of metal coupons remaining in crude oil containers after 30 and 45 days of immersion. The degradation of metal elements while immersed in crude oils was examined using atomic absorption spectroscopy (AAS) to explain the unexplained weight loss of metal coupons during corrosion rate measurements through the weight loss method. In the sample preparation process, 1 ml of each crude oil sample was mixed with 9 ml of 2-propanol and then filtered. In order to confirm the formation of corrosion, the effect of corrosion on the original hardness of metal samples was evaluated using the Vicker's hardness tester. The operational principles of this device are outlined in Figure 3 and the subsequent equation.

Figure 3. Indenter of the Vicker's hardness tester

$$\mathrm{HV} = 1.854 * \mathrm{P}^2/\mathrm{L}^2 \tag{2}$$

Where;

P= Applied Load on the surface of metal
L= Diagonal length of square
HV= Hardness

Regarding the essential measurements both initial hardness and hardness after the formations of the corrosion on the metal surfaces were measured by such instrument. For each measurement of hardness at least three hardness values were measured on each meal coupon and the average values were recorded.

3. Results and Discussion

According to the results for the analysis of the chemical compositions of selected metals by the X-ray fluorescence have been interpreted in the Table 2.

Table 2. Chemical compositions of the ferrous metals

Metal	**Fe (%)**	**Ni (%)**	**Cr (%)**	**Cu (%)**
Carbon Steel (High)	98.60	0.17	0.14	0.37
Carbon Steel (Medium)	99.36	-	-	-
Carbon Steel (Mild Steel)	99.46	-	<0.07	-
410-MN: 1.8 420-MN: 2.8 (Stainless Steel)	88.25	0.18	10.92	0.10
410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	87.44	-	11.99	-
321-MN:1.4 304-MN:1.9 (Stainless Steel)	72.47	8.65	17.14	-
Monel 400	1.40	64.36	<0.04	33.29

The results presented above indicate higher levels of iron in carbon steels, moderate levels of iron in stainless steels, and a minimal amount of iron in Monel metal. Moreover, trace amounts of other metallic elements such as nickel, copper, molybdenum, and chromium were detected in stainless steels. These additional elements contribute to the higher strength and corrosion resistance of stainless steels. The purpose of incorporating d-block elements with iron in these metals is to enhance their strength and reduce their susceptibility to corrosion. By combining a minimum of 12% chromium with a sufficient amount of nickel, a stable protective layer against corrosion is formed on the surface of the metal when exposed to corrosive environments (Khana et al., 2009). Based on the examination of the corrosive substances present in the crude oils, the findings have been explained in Table 3.

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

Upon reviewing the aforementioned findings, it can be inferred that Das Blend crude oils contained comparatively higher levels of elemental sulfur, mercaptans, and organic acids, as well as lower levels of salts when compared to Murban crude oil. Previous research has shown that recent findings indicate that the effects of corrosive compounds are not solely dependent on their concentrations, but also require an analysis of other factors such as the temperature of the relevant systems in order to understand their corrosive properties more comprehensively (Afaf et al., 2007). Organic acids are corrosive compounds present in crude oils, known as naphthenic acids with the general formula RCOOH, originating from natural sources in the Earth's crust. The overall concentration of acids in a particular crude oil is referred to as its acidity or total acid number (TAN) (Afaf et al., 2007).

These organic acids exhibit strong oxidizing properties, capable of oxidizing metals through a consistent mechanism involving the organic acids, as demonstrated in subsequent chemical reactions (Ahmed, Elnour & Ibrahim, 2014).

$$\mathrm{Fe + 2RCOOH \rightarrow Fe(RCOO)2 + H2} \quad (3)$$
$$\mathrm{FeS + 2\,RCOOH \rightarrow Fe(COOR)2 + H2S} \quad (4)$$
$$\mathrm{Fe(COOR)2 + H2S \rightarrow FeS + 2\,RCOOH} \quad (5)$$

Similar with the impact of organic acids on the metallic corrosion salts are the strong oxidizing agents that involve in the destructions of the metals. According to the recent studies regarding the impact of salts on the metallic corrosion that there were mainly investigated three types of slats presence in the crude oils namely as NaCl, $CaCl_2$ and $MgCl_2$ (Davis and Davis, 2003). When increasing the temperature of crude oils the molecules of those salts tend to be converted into HCl molecules which are having inert conditions at those conditions. With variations of the temperature such HCl molecules are reacted with water or even moisture presence in such crude oils and formed the hydrochloric acids which have been identified as highly corrosive compound because of the instability and oxidizing ability of hydrochloric acids. The chemical reactions of the corrosion process regarding the salts have been given in the following chemical reactions (Speight et al., 1999).

$$\mathrm{CaCl2 + H2O \rightarrow CaO + 2HCl} \quad (6)$$
$$\mathrm{HCl + Fe \rightarrow FeCl2 + H2} \quad (7)$$
$$\mathrm{H2 + S \rightarrow H2S} \quad (8)$$
$$\mathrm{FeCl2 + H2S \rightarrow FeS + 2HCl} \quad (9)$$

Sulfur compounds are highly corrosive substances that can be found in various forms throughout nature. These compounds exhibit greater corrosive potential compared to other types of corrosive substances due to the increased reactivity of their functional groups. Among the various sulfur compounds, elemental sulfur, Mercaptans, sulfoxides, thiophenes, and hydrogen sulfides have been identified as particularly corrosive due to the functional groups present in them. Mercaptans, in particular, are known for their high corrosiveness due to the reactivity of the "RSH" functional group. The corrosion process is influenced by the specific behavior of the sulfur compound in question and the characteristics of its functional group (Fang, Nesic & Young, 2008). The phenomenon of localized corrosion caused by elemental sulfur is commonly referred to as "localized corrosion" and is typically observed at temperatures around 800^0C. Similarly, the corrosion process resulting from Mercaptans is termed "sulfidation" and predominantly occurs within the temperature range of 230^0C-460^0C. The initial chemical reactions responsible for these corrosion processes are outlined in the following chemical reactions (Bota, Nesic, Qu and Wolf, 2010).

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

$$8\ Fe + S8 \rightarrow 8\ FeS \tag{11}$$

Therefore, the impact on the corrosion of such corrosive compounds of crude oils is possible to discuss with the experimental results with the aid of related environmental conditions unless discussing with past investigations.

According to the determinations of the corrosion rates of metals in order of 15, 30 and 45 days immersion time periods with respect to both crude oils have been interpreted in the Table 4 and Table 5.

Table 4. Corrosion rates of metals coupons in Murban crude oil

Metal	**Corrosion Rate after 15 Days ($cm^3\ inch^{-1} day^{-1}$)**	**Corrosion Rate after 30 Days ($cm^3\ inch^{-1}\ day^{-1}$)**	**Corrosion Rate after 45 Days ($cm^3\ inch^{-1}\ day^{-1}$)**	**Average Corrosion Rate ($cm^3\ inch^{-1}\ day^{-1}$)**
Carbon Steel (High)	0.811971	0.466425	0.068794	0.4490632
Carbon Steel (Medium)	0.817791	0.180339	0.073358	0.3571623
Carbon Steel (Mild Steel)	0.10973	0.048244	0.038592	0.0655217
410-MN: 1.8 420-MN: 2.8 (Stainless Steel)	0.041784	0.016075	0.011801	0.02322

410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	0.11626	0.011968	0.007574	0.0452676
321-N:1.4 304-MN:1.9 (Stainless Steel)	0.016612	0.007453	0.005599	0.009888
Monel 400	0.356263	0.034877	0.026729	0.13929

Table 5. Corrosion rates of metals coupons in Das Blend crude oil

Metal	**Corrosion Rate after 15 Days (cm^3 inch^{-1} day^{-1})**	**Corrosion Rate after 30 Days (cm^3 inch^{-1} day^{-1})**	**Corrosion Rate after 45 Days (cm^3 inch^{-1} day^{-1})**	**Average Corrosion Rate (cm^3 inch^{-1} day^{-1})**
Carbon Steel (High)	0.350249	0.224901	0.024738	0.1999627
Carbon Steel (Medium)	0.481055	0.140654	0.05911	0.2269396
Carbon Steel (Mild Steel)	0.162883	0.141093	0.100635	0.1348702
410-MN: 1.8 420-MN: 2.8 (Stainless Steel)	0.044146	0.034035	0.006149	0.0281102
410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	0.053701	0.034841	0.016363	0.0349681

321-MN:1.4 304-MN:1.9 (Stainless Steel)	0.022894	0.006503	0.002825	0.0107404
Monel 400	0.061554	0.037655	0.016067	0.0384254

As the conclusions of above results the average corrosion rates of metals with respect to both Murban and Das Blend crude oils have been shown in the Figure 4.

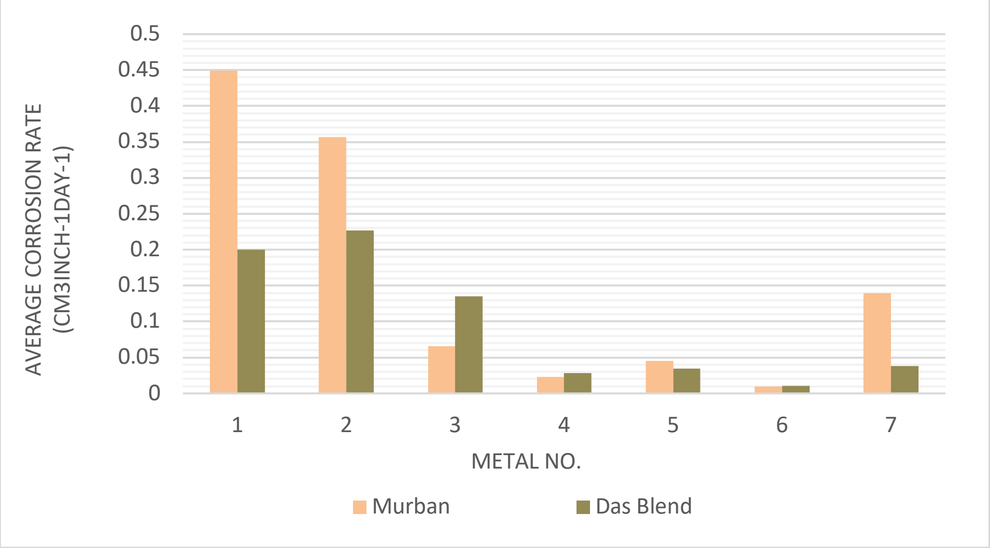

Figure 4. Average corrosion rates of metals

Based on the findings discussed above, it can be highlighted that stainless steels exhibit higher corrosion rates, Monel demonstrates moderate corrosion rates, and stainless steels show lower corrosion rates in relation to both crude oils. A comparison of corrosion rates among different grades of stainless steels reveals that 321-MN: 1.4 and 304-MN: 1.9 exhibit the lowest corrosion rates, with approximately 18% chromium and 8.65% nickel content. In light of the results obtained, it can be concluded that the addition of d-block elements to stainless steels to reduce corrosion rates enhances the effectiveness of the protective film consisting of nickel and chromium, particularly when the chromium content is at least 12% and there are sufficient levels of nickel present (Singh et al., 2006). In relation to the corrosive characteristics of crude oils and the findings on the corrosion rates of metals, it can be inferred that salts have a greater impact compared to organic acids. This is evidenced by the higher corrosion rates exhibited by four types of metals in Murban, whereas three other types displayed higher corrosion rates in Das Blend. Furthermore, it is improbable to anticipate a significant effect on metal corrosion from sulfur compounds due to temperature-related constraints

(Hassan et al., 2013). The corrosion rates of metals in Murban and Das Blend crude oils have been illustrated in Figure 5 and Figure 6 by analyzing the results mentioned above.

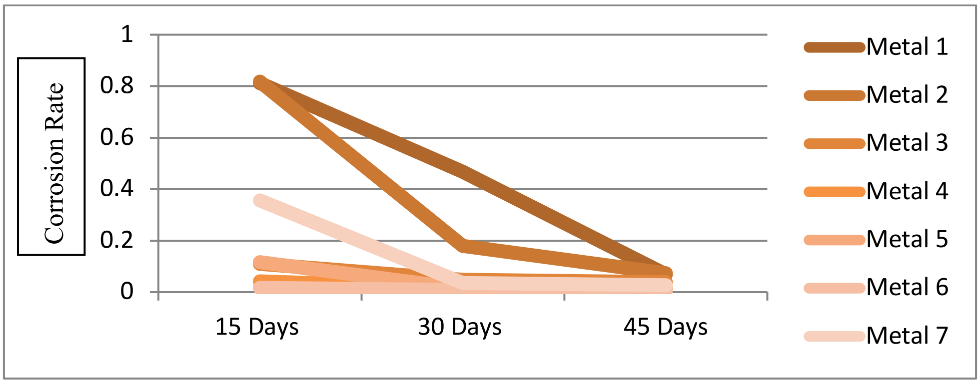

Figure 5. Variations of the corrosion rates of metals with the exposure time in Murban

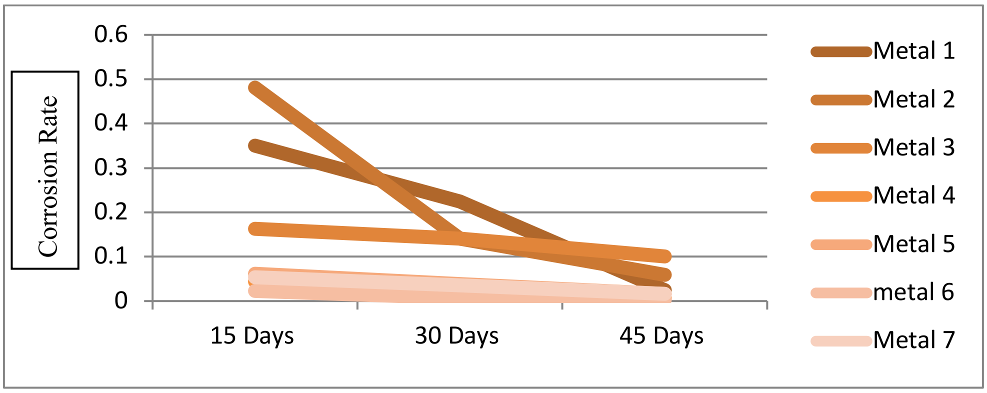

Figure 6. Variations of the corrosion rates of metals with the exposure time in Das Blend

Based on the two aforementioned variations, it is feasible to detect and highlight certain analogous patterns in the corrosion curves over time, wherein the diminishing corrosion rates over time when exposed to crude oils are inversely proportional to the exposure duration. This phenomenon is elucidated in the weight loss method, attributed to the self-protective properties of previously formed corrosion compounds against continued corrosion at the same rate (Afaf et al., 2007). The microscopic examination of the corroded metal surfaces revealed a variety of compounds, some of which were identified as distinct from others, as illustrated in Figure 7.

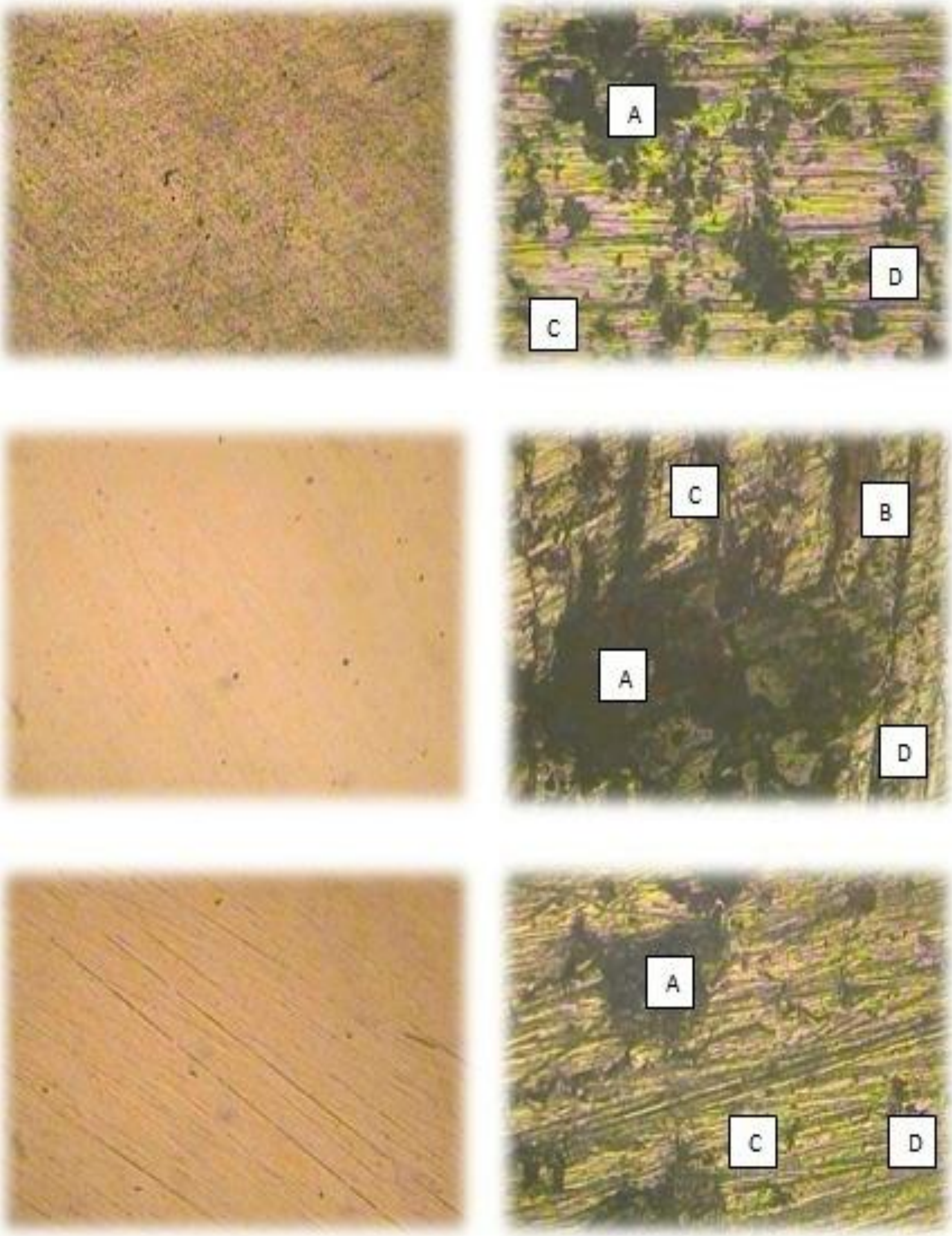

Figure 7. The corroded metal surfaces

Among different observations some of specific corrosion compounds were highlighted by referring the visible appearances of such corrosion compounds foremost of the color as explained in the Table 6 (Muller, 1982).

A- Ferrous Sulfide/ Copper Sulfide (FeS/ CuS)
B- Ferrous Oxide (Fe_2O_3)
C- Corrosion Cracks
D- Pitting Corrosion

Table 6. The 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, property of powder	Unable to specify

The formations of ferrous sulfide (FeS) and, less frequently, ferrous oxides (Fe2O3), corrosion cracks, and asymmetric distributions of cavities on most metal surfaces have been highlighted based on observations and conclusions. These phenomena align with the theoretical explanations of corrosion processes with corrosive properties as described in existing research. Additionally, specific observations revealed the presence of a black-colored compound on certain Monel metal surfaces, which proved challenging to distinguish from ferrous sulfide (FeS) through visible inspections alone. Consequently, it is recommended that compositional analysis of such corrosion compounds be conducted using advanced analytical techniques such as X-ray diffraction (XRD) to yield more precise results and analysis for future studies. In the study, an analysis was conducted on the transformation of deteriorated ferrous and copper metals into crude oils during immersion in crude oils using atomic absorption spectroscopy (AAS). The obtained results were found to be indicative of the corrosion rates of the respective metals, as detailed in Table 7.

Table 7. Decayed metallic concentrations 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	-	-
	Das Blend	-	-
410-MN: 1.8 420- MN: 2.8 (Stainless Steel)	Murban	-	-
	Das Blend	-	-
410-MN: 1.7 420-MN: 1.7 (Stainless Steel)	Murban	-	-
	Das Blend	-	-

321-MN:1.4 304-MN:1.9 **(Stainless Steel)**	Murban	-	-
	Das Blend	-	-
Monel 400	Murban	-	10.47
	Das Blend	-	9.49

The concluded interpretations of the above results have been shown in the Figure 8 and Figure 9.

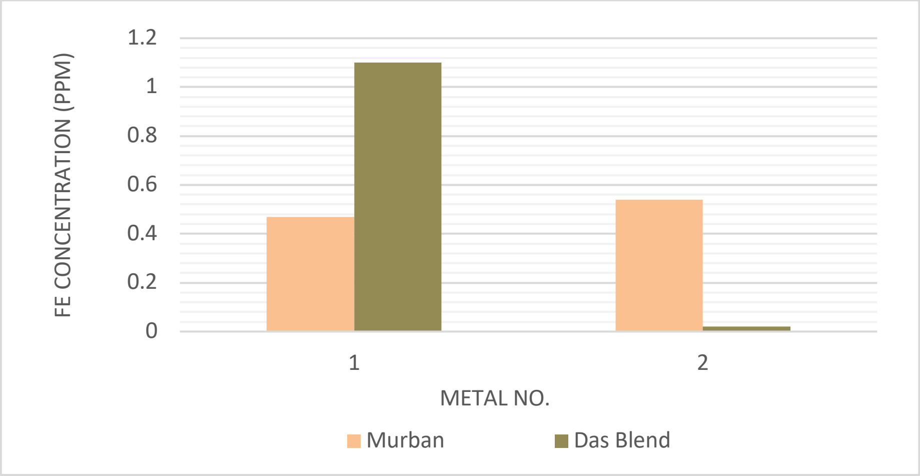

Figure 8. Decayed ferrous concentrations into crude oils from metals

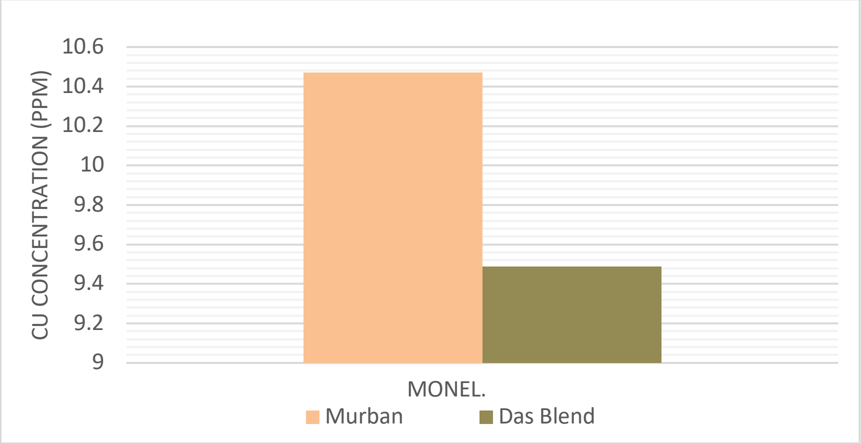

Figure 9. Decayed copper concentrations into crude oils from metals

According to the aforementioned analyses, there are noticeable deteriorations of iron in carbon steels which exhibit the highest rates of corrosion when exposed to crude oils. Similarly, there are relatively higher deteriorations of copper in Monel metal, resulting in intermediate corrosion rates when exposed to crude oils. In contrast, there were no observed deteriorations in the metallic concentration of stainless steels, which showed the lowest corrosion rates among the materials tested.

The process of metal deterioration can be elucidated through the mechanism of electron repulsion. Following the formation of corrosive compounds on the metal surfaces, these compounds tend to detach from the surfaces due to the repulsive and attractive forces between the electrons and protons in the compounds and the metals (Hashemi and Smith, 2006). Consequently, metal deterioration may occur concomitantly with corrosion.

According to the analysis of the corrosion effect on the hardness of metals by the Vicker's hardness tester the obtained results have been interpreted in the Figure 10 and Figure 11.

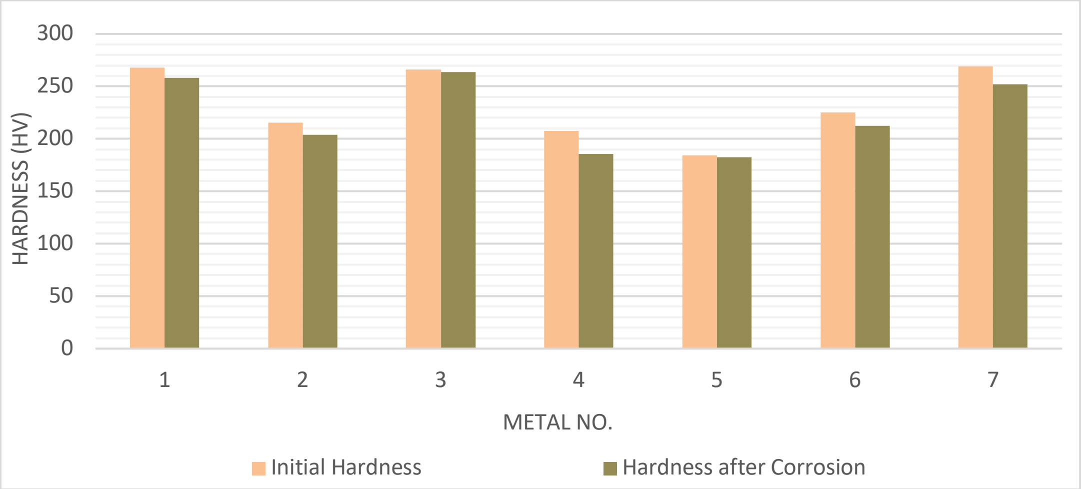

Figure 10. Variations of the initial hardness of metals in Murban

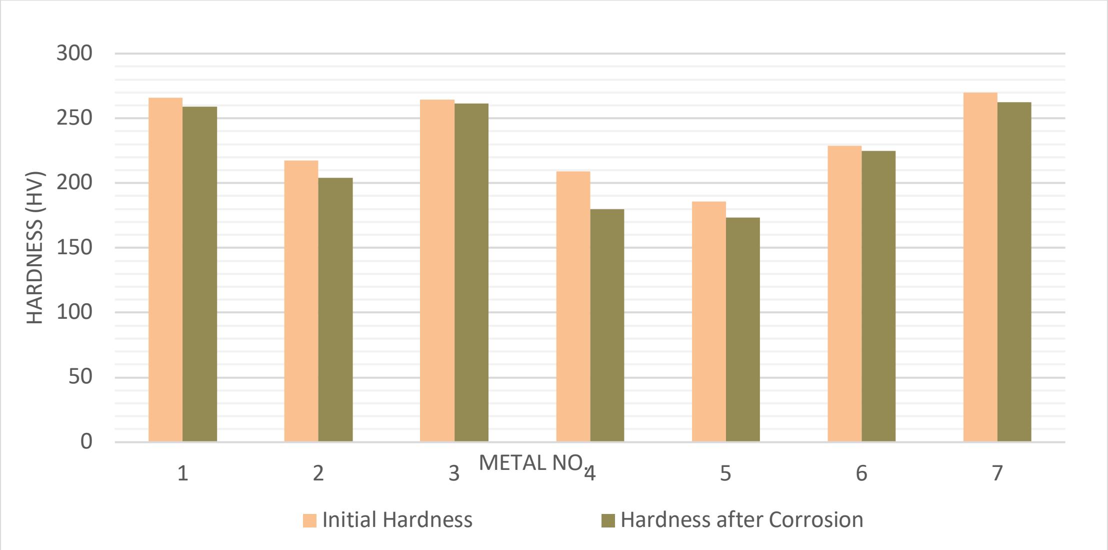

Figure 11. Variations of the initial hardness of metals in Das Blend

The two variations demonstrated a slight decrease in the initial hardness of most metals following exposure to corrosion caused by different types of crude oil. Despite these observations, establishing any correlation between corrosion rates and reductions in initial hardness of metals proved challenging. The reductions in initial hardness can be attributed to two recognized reasons, as outlined (Bolton et al., 1994). Uncertain conditions are created on the surfaces of metals as a result of the interplay between repulsive and attractive forces among the consecutive electrons and protons within these compounds. The diversity of metal surfaces can vary due to the formation of various metal oxide layers, leading to differences in hardness among corrosion compounds.

- The formed uncertain conditions on the metal surfaces due to the repulsive and attractive forces between the successive electrons and protons of such compounds
- Heterogeneity of the metal surfaces after formations of the different metal surfaces on the metals surfaces and such corrosion compounds may have different hardness

4. Conclusion

The overall findings of the existing research indicate that stainless steels with chemical compositions containing at least 12% chromium and a sufficient amount of nickel demonstrate lower corrosion rates compared to other materials. Salts have been observed to have a relatively higher corrosive impact on metallic corrosion when compared to other corrosive compounds, particularly sulfur compounds which show less aggressiveness at lower temperatures. Corrosion compounds such as FeS/CuS, Fe2O3, corrosion cracks, and irregular pitting are commonly formed on metal surfaces. Additionally, the decay of copper and ferrous metals with higher corrosion rates has been noted in relevant crude oil samples. Furthermore, there is a slight reduction in the initial hardness of metal coupons due to the effects of corrosion.

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