

Article The Effect of Corrosion on Crude Oil Distillation Plants

Timur Chis *^(D), Ancaelena Eliza Sterpu ^(D) and Olga Valerica Săpunaru

Chemical and Chemical Engineering Department, Ovidius University Constanta, Mamaia Blv 224, 900450 Constanta, Romania; asterpu@univ-ovidius.ro (A.E.S.); olgutasapunaru@yahoo.com (O.V.S.) * Correspondence: tchis@univ-ovidius.ro; Tel.: +40-723-670-107

Abstract: The analysis of the corrosion phenomenon of the installations of the largest refinery in Romania confirmed the existence of its conditions, in the atmospheric distillation plants of crude oil, in the heat exchangers, and in the petrol and diesel hydrofining plants. However, the slightest effect of this phenomenon was found in the dome, the plates, and the exhaust pipes at the top of the distillation column, as well as in the reflux vessel of the atmospheric distillation plant. The main cause of the corrosion phenomenon and the increase in the corrosion areas of the material of the crude oil refining installations is the presence of hydrochloric acid, obtained after incorrect desalination (hydrolysis of calcium and magnesium chloride). To prevent this phenomenon, in laboratory conditions, we tested an antacid inhibitor (alkyl phenol) and a residual product (undistilled polyamine), with the role of supplementing the commercial inhibitor with a cheap product. The effectiveness of these additives was evaluated by introducing specimens made from the material taken from the refining column and from the reflux vessel into a solution of hydrochloric acid of various concentrations (5–15%). This solution was treated with the two corrosion inhibitors, the analysis of their effectiveness being performed at concentrations ranging between 0.05 and 0.2%. In addition, in this article, we deduced the equations of variation of the corrosion rate, depending on the working temperature of the refinery.

Keywords: refinery columns; distillation column corrosion; corrosion inhibitor; hydrochloric acid; undistilled polyamine; corrosion rate

1. Introduction

In recent years, the study of corrosion of atmospheric crude oil distillation plants has been discussed more in articles published in journals. This is primarily due to the aging process of petrochemical plants (in the last 10 years, the existing facilities have been modernized due to changes in the quality requirements of the finished products and especially due to the worsening environmental conditions) [1].

Thus, the corrosion of the distillation columns has been the subject of several studies, an important and triggering element of this phenomenon being the modification of the properties of refined fluids (pH, water content, inorganic salt content, etc.) [2–4]. Corrosion has also been detected in crude oil distillation plants in Romania, which makes the phenomenon global and affects petrochemical plants [5–7].

The crude oil processing plants are subjected to the corrosion process, the phenomenon being the most common in the atmospheric distillation plant [8]. Corrosion was also found in the gasoline and diesel hydrofining plant and in the heat exchangers related to the crude oil processing area [9–12]. The analysis of the integrity of the distillation column and the reflux vessel from the largest refinery in Romania led to the repair and replacement of the first four plates (related to light products), the column dome, the aerator of the installation, and the product flue pipe, as well as components from the reflux vessel (heating pipe and some portions of the vessel walls) [13–15].

The corrosion phenomenon is mainly due to the hydrolysis of calcium and magnesium chloride, present in the crude oil water (following an incomplete desalination).



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Copyright: © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). The temperature at the top of the column is also a factor that can activate the corrosive phenomenon, especially when the demand for gasoline is high and the installation works at a temperature at which the passivation phenomenon occurs [16–20].

Prevention of the corrosive effect is currently performed by injecting anticorrosive additives (to form a protective film inside the distillation column and to neutralize the acid effect of crude oil), monitoring the pH and chlorides in the reflux vessel, and monitoring inorganic chlorides in crude oil and the one treated in desalination [21–24].

The mechanical integrity of the distillation plant is achieved [25]:

- Preventively, by taking specimens from the column material and determining in the laboratory the rate of corrosion of the metal;
- Actively, by performing non-destructive measurement (in points) of wall thickness, determining the corrosion rate, and entering the repair of the installation (with high economic consequences due to the reduction in the current repair period, sometimes from 5 years to 2 years).

In the distillation column, it was observed that the corrosive phenomenon is due to the presence of HCl and water in the high area of the distillation column and HCl, H_2S , and water in the reflux vessel (Figures 1 and 2).



Figure 1. Pores in the pipes at the top of the distillation column (photo by Timur Chis).



Figure 2. Corrosion in the reflux vessel (photo by Timur Chis).

Crude oil contains significant amounts of salts (NaCl, MgCl₂, or CaCl₂). In principle, NaCl does not hydrolyze under normal operating conditions, this salt being a low contributor to the corrosion process.

It was also found that HCl is not corrosive if the temperature in the distillation is higher than the dew point temperature of the water. However, there is a danger of the formation of alkaline salts (above the dew point temperature) and the formation of hydrochloric acid.

2. Materials and Methods

In order to determine the corrosion rate and the effectiveness efficiency of two inhibitory solutions, products from the upper plates of the atmospheric distillation column (gasoline separation zone) and from the reflux zone were taken from a refining plant.

The laboratory analysis of these products determined the components S^{2-} , Cl^- , Fe^{3+} , and Cu^{2+} in correlation with the pH of the solution.

Sample metals were also taken from the plates and pipes affected by corrosion, which were cleaned to metallic luster and measured (weight and volume).

The metal samples were placed in the products taken and kept for 20 h at a temperature of 25 $^{\circ}$ C.

Subsequently, they were measured again, determining the corrosion rate and the corrosive penetration.

At the same time, the metal samples were introduced in a solution of hydrochloric acid of various concentrations (5–15%).

This solution was treated with the two corrosion inhibitors, with a concentration ranging from 0.05 to 0.2%.

The immersion time of the specimens was 20 h at a temperature of 25 $^{\circ}$ C, 6 h at a temperature of 40 $^{\circ}$ C, and 2 h at a temperature of 70 $^{\circ}$ C.

After completion of the test, the final corrosion rate was determined.

The determination of the water content of the products taken from the atmospheric distillation column (the first four plates) and from the reflux vessel was carried out according to the standard ASTM D6304-20-Standard Test Method for Determination of Water in Petroleum Products, Lubricating Oils, and Additives by Coulometric Karl Fischer Titration [26].

The determination of chlorides in samples taken from the atmospheric distillation column was performed by potentiometric titration according to the ASTM 4929 ASTM D4929-19a Standard Test Method for Determination of Organic Chloride Content in Crude Oil [27] and X-ray fluorescence spectrometry.

Sulfur was determined according to the ASTM D1266-18 Standard Test Method for Sulfur in Petroleum Products (Lamp Method) [28].

pH was determined by the potentiometric method according to the ASTM D7946-19 Standard Test Method for Initial pH (i-pH) -Value of Petroleum Products [29].

Fe³⁺ was determined by the atomic adsorption method according to the ASTM D5863-00a (2016) Standard Test Methods for Determination of Nickel, Vanadium, Iron, and Sodium in Crude Oils and Residual Fuels by Flame Atomic Absorption Spectrometry [30].

Each test was repeated 3 times, determining the measurement error and the statistical deviation of the data obtained. The error was up to 0.1%.

3. Results

Hydrochloric acid (HCl) in combination with NH₃ forms NH₄Cl in solid form in distillation tower overhead systems [25].

The hydrolysis equations that take place in the atmospheric distillation column are presented in Table 1. In Tables 2 and 3, we have presented the results obtained from the analysis of the water taken from the top of the distillation column and the reflux vessel.

The corrosion rate was determined by repeated weighing of the tested samples and the penetration index by calculation (Table 4).

Tables 5–7 present the values determined after the introduction of the specimens taken from the distillation column and the reflux vessel into hydrochloric acid solution and then into the HCl solution treated with a corrosion inhibitor, such as alkyl phenol and a residual product (undistilled polyamine).

Reaction	Approximate Starting Temperature °C (°F)	Approximate Degree of Hydrolysis at 340 °C (650 °F)
$\begin{array}{l} NaCl+H_2O{\rightarrow}NaOH+HCl\\ CaCl_2+H_2O{\rightarrow}Ca(OH)_2+2HCl\\ MgCl_2+2H_2O{\rightarrow}Mg(OH)_2+2HCl \end{array}$	≥500 (≥930) 210 (410) 120 (248)	2% 10% 90%

 Table 1. Hydrolysis reactions in the atmospheric distillation column of crude oil.

Table 2. The results obtained from the analysis of the products at the top of the distillation column.

pH	Fe ³⁺ , ppm	Cu ²⁺ , ppm	S ^{2–} , ppm	Cl ⁻ , g/1000 mL
4,3	3.3	0	0	0.0366
4.5	8.2	0	0	0.0567
5.1	30.5	0.015	0	0.0494
6.4	0	trace	0	0.0298
7.3	0	0.0014	92.0	0.0308
8.4	0	0	47.5	0.0535
9.2	0	0	0	0.0714

Table 3. Results obtained from the analysis of the product samples taken from the reflux vessel of the distillation column.

pH	Fe ³⁺ , ppm	Cu ²⁺ , ppm	S ^{2–} , ppm	Cl-, g/1000 mL
4.3	5.3	0	0	0.0855
4.5	7.4	0	0	0.1473
5.1	34.3	0	0	0.1435
6.4	0	0	0	0.0822
7.3	0	0.0025	80.95	0.0354
8.4	0	0	12.88	0.1199
9.2	0	0	0	0.2148

Table 4. Corrosion rate determined on samples collected from the top and bottom of the distillation column.

Material Column	Collection Area	Corrosion Rates, g/m ² h	Penetration Corrosion Rates, mm/an
OL37 steel	Reflux vessel	0.18890	0.2022
OL37 steel	Reflux vessel pipe	0.18455	0.1934
OL37 steel	Gasoline plate	0.24908	0.2564
OL37 steel	Vapor exhaust pipe	0.26598	0.2804
Brass	Reflux vessel	0.12994	0.1474
Brass	Reflux vessel pipe	0.15043	0.1494
Brass	Gasoline plate	0.58467	0.5939
Brass	Vapor exhaust pipe	0.43057	0.4536

Test Temperatures, °C	Antacid Additive Content (% Alkyl Phenol, % Undistilled Polyamide)	Corrosion Rates, g/m ² h, Alkyl Phenol Additive	Corrosion Efficacy Reduces, %, Alkyl Phenol Additive	Corrosion Rates, g/m ² h, Undistilled Polyamide Additive	Corrosion Efficacy Reduces, %, Undistilled Polyamide Additive
25	0.00	0.3000	-	0.3000	-
25	0.05	0.0182	93.93	0.0193	93.57
25	0.10	0.0133	95.57	0.0154	94.87
25	0.20	0.0080	97.33	0.0120	96.00
40	0.00	1.7000	-	1.7000	-
40	0.05	0.1900	88.82	0.1600	90.59
40	0.10	0.1500	91.18	0.1400	91.76
40	0.20	0.1100	93.53	0.0900	94.71
70	0.00	16.000	-	16.000	-
70	0.05	1.4500	90.94	1.3300	91.69
70	0.10	0.9900	93.81	0.9400	94.13
70	0.20	0.8500	94.69	0.7300	95.44

Table 5. Corrosion rate determined on samples collected from the material of the distillation column (OL37), introduced in a bath with HCl in proportion of 5%.

Table 6. Corrosion rate determined on samples collected from the material of the distillation column (OL37), introduced in a bath with HCl in proportion of 10%.

Test Temperatures, °C	Antacid Additive Content (% Alkyl Phenol, % Undistilled Polyamide)	Corrosion Rates, g/m ² h, Alkyl Phenol Additive	Corrosion Efficacy Reduces, %, Alkyl Phenol Additive	Corrosion Rates, g/m ² h, Undistilled Polyamide Additive	Corrosion Efficacy Reduces, %, Undistilled Polyamide Additive
25	0.00	0.7000	-	0.700	-
25	0.05	0.0510	92.71	0.061	91.29
25	0.10	0.0418	94.03	0.053	92.43
25	0.20	0.0215	96.93	0.042	94.00
40	0.00	2.9800	-	2.980	-
40	0.05	0.1510	94.93	0.320	89.26
40	0.10	0.1672	94.39	0.210	92.95
40	0.20	0.1530	94.87	0.156	94.77
70	0.00	36.000	-	36.00	-
70	0.05	5.9300	83.53	6.400	82.22
70	0.10	3.9310	89.08	2.700	92.50
70	0.20	2.9450	91.82	2.530	92.97

Table 7. Corrosion rate determined on samples collected from the material of the distillation column (OL37), introduced in a bath with HCl in proportion of 15%.

Test Temperatures, °C	Antacid Additive Content (% Alkyl Phenol, % Undistilled Polyamide)	Corrosion Rates, g/m ² h, Alkyl Phenol Additive	Corrosion Efficacy Reduces, %, Alkyl Phenol Additive	Corrosion Rates, g/m ² h, Undistilled Polyamide Additive	Corrosion Efficacy Reduces, %, Undistilled Polyamide Additive
25	0.00	1.500	-	1.500	-
25	0.05	0.130	91.33	0.092	93.87
25	0.10	0.082	94.53	0.072	95.20
25	0.20	0.071	95.27	0.053	96.47
40	0.00	10.22	-	10.22	-
40	0.05	0.630	93.84	0.820	91.98
40	0.10	0.501	95.10	0.640	93.74
40	0.20	0.220	97.85	0.530	94.81
70	0.00	68.20	-	68.20	-
70	0.05	14.20	79.18	15.30	77.57
70	0.10	9.20	86.51	9.500	86.07
70	0.20	6.90	89.88	6.450	90.54

The analysis of the samples collected from the top of the distillation column and from the reflux vessel indicates the presence of ion Fe^{3+} in the products that have an acidic pH; they confirm the beginning of the corrosion phenomenon.

Sulfur ion S^{2-} was detected in the alkaline zone of the solution.

The presence of Cu in the harvested products indicates the corrosive role of these solutions on the installations containing this material (vaporization furnace) and which were subsequently transported to the trays and the reflux vessel in the distillation plant.

The equations used for the numerical modeling of the corrosion process of the installations were determined according to the analysis temperature, the HCl content of the solution, and the amount of antacid additive used (Tables 8–10).

Table 8. Corrosion rate equations (y) as a function of temperature, HCl content of the solution (5%), and the amount of antacid additive used (x, %).

Test Temperature, °C	Equation Type	Corrosion Rate Regression Equation (y) Depending on the Amount of Additive Used (x,% Alkyl Phenol)	R ²	Corrosion Rate Regression Equation (y) Depending on the Amount of Additive Used (% Undistilled Polyamide)	R ²
25	Exponential	$y = 0.1083e^{-15.62x}$	0.8548	$y = 0.1052e^{-13.55x}$	0.8163
25	Linear	y = -1.1825x + 0.1883	0.4953	y = -1.1626x + 0.1884	0.4871
25	Polynomial (grad 2)	$y = 17.825x^2 - 4.8749x + 0.2775$	0.8995	$y = 17.815x^2 - 4.8527x + 0.2775$	0.8978
25	Polynomial (grad 3)	$y = -275.4x^3 + 96.69x^2 - 9.782x + 0.3$	1	$y = -275.33x^3 + 96.66x^2 - 9.7587x + 0.3$	1
40	Exponential	$y = 0.7529e^{-11.71x}$	0.7996	$y = 0.7216e^{-12.49x}$	0.809
40	Linear	y = -6.4743x + 1.104	0.508	y = -6.5314x + 1.094	0.5041
40	Polynomial (grad 2)	$y = 95.364x^2 - 26.228x + 1.5808$	0.9038	$y = 96.273x^2 - 26.474x + 1.5754$	0.8974
40	Polynomial (grad 3)	$y = -1456.7x^3 + 512.5x^2 - 52.183x + 1.7$	1	$y = -1523.3x^3 + 532.5x^2 - 53.617x + 1.7$	1
70	Exponential	$y = 6.3134e^{-12.57x}$	0.8086	$y = 6.2226e^{-13.23x}$	0.8201
70	Linear	y = -61.549x + 10.208	0.4969	y = -61.989x + 10.174	0.4976
70	Polynomial (grad 2)	$y = 932.09x^2 - 254.62x + 14.868$	0.9062	$y = 934.64x^2 - 255.59x + 14.847$	0.9038
70	Polynomial (grad 3)	$y = -13830x^3 + 4892.5x^2 - 501.05x + 16$	1	$y = -14,090x^3 + 4969.5x^2 - 506.65x + 16$	1

Table 9. Corrosion rate equations (y) as a function of temperature, HCl content of the solution (10%), and the amount of antacid additive used (x,%).

Test Temperature, °C	Equation Type	Corrosion Rate Regression Equation (y) Depending on the Amount of Additive Used (x,% Alkyl Phenol)	R ²	Corrosion Rate Regression Equation (y) Depending on the Amount of Additive Used (% Undistilled Polyamide)	R ²
25	Exponential	$0.2804e^{-15.03x}$	0.8508	$0.2763e^{-11.76x}$	0.7857
25	Linear	y = -2.753x + 0.4445	0.5039	y = -2.6583x + 0.4466	0.4906
25	Polynomial (grad 2)	$y = 40.628x^2 - 11.169x + 0.6476$	0.898	$y = 40.382x^2 - 11.023x + 0.6485$	0.8972
25	Polynomial (grad 3)	$y = -640.43x^3 + 224.02x^2 - 22.58x + 0.7$	1	$y = -629.33x^3 + 220.6x^2 - 22.237x + 0.7$	1
40	Exponential	$0.9201e^{-11.8x}$	0.7661	$y = 1.2954e^{-12.86x}$	0.826
40	Linear	y = -11.296x + 1.8512	0.467	y = -11.546x + 1.9268	0.5124
40	Polynomial (grad 2)	$y = 178.94x^2 - 48.363x + 2.7459$	0.888	$y = 169.82x^2 - 46.723x + 2.7759$	0.9105
40	Polynomial (grad 3)	$y = -2860.7x^3 + 998.15x^2 - 99.336x + 2.98$	1	$y = -2494.7x^3 + 884.2x^2 - 91.173x + 2.98$	1
70	Exponential	$18.539e^{-11.05x}$	0.8144	$18.277e^{-12.18x}$	0.8502
70	Linear	y = -136.77x + 24.169	0.5386	y = -140.42x + 24.194	0.5505
70	Polynomial (grad 2)	$y = 1923.2x^2 - 535.16x + 33.785$	0.9211	$y = 1976.8x^2 - 549.9x + 34.078$	0.9424
70	Polynomial (grad 3)	$y = -27,067x^3 + 9674.2x^2 - 1017.4x + 36$	1	$y = -23,490x^3 + 8703.5x^2 - 968.45x + 36$	1

Test Temperature, °C	Equation Type	Corrosion Rate Regression Equation (y) Depending on the Amount of Additive Used (x,% Alkyl Phenol)	R ²	Corrosion Rate Regression Equation (y) Depending on the Amount of Additive Used (% Undistilled Polyamide)	R ²
25	Exponential	$y = 0.5804e^{-13.16x}$	0.8203	$y = 0.5221e^{-14.14x}$	0.8273
25	Linear	y = -5.8109x + 0.9542	0.4978	y = -5.844x + 0.9406	0.4885
25	Polynomial (grad 2)	$y = 87.991x^2 - 24.038x + 1.3942$	0.9077	$y = 89.282x^2 - 24.338x + 1.387$	0.898
25	Polynomial (grad 3)	$y = -1293.7x^3 + 458.45x^2 - 47.088x + 1.5$	1	$y = -1381x^3 + 484.75x^2 - 48.945x + 1.5$	1
40	Exponential	$y = 3.9527e^{-16.69x}$	0.8723	$y = 3.8688e^{-12.48x}$	0.8003
40	Linear	y = -40.542x + 6.4402	0.5017	y = -39.194x + 6.482	0.4903
40	Polynomial (grad 2)	$y = 601.02x^2 - 165.04x + 9.4453$	0.8977	$y = 598.09x^2 - 163.08x + 9.4725$	0.9003
40	Polynomial (grad 3)	$y = -9468.7x^3 + 3312.5x^2 - 333.75x + 10.22$	1	$y = -9136.7x^3 + 3214.5x^2 - 325.88x + 10.22$	1
70	Exponential	$y = 38.563e^{-10.24x}$	0.8184	$y = 40.53e^{-10.69x}$	0.8436
70	Linear	y = -256.4x + 47.06	0.5618	y = -260.43x + 47.65	0.583
70	Polynomial (grad 2)	$y = 3468.2x^2 - 974.81x + 64.401$	0.9311	$y = 3385.9x^2 - 961.8x + 64.58$	0.937
70	Polynomial (grad 3)	$y = -46,433x^3 + 16,765x^2 - 1802.2x + 68.2$	1	$y = -44,250x^3 + 16,057x^2 - 1750.2x + 68.2$	1

Table 10. Corrosion rate equations (y) as a function of temperature, HCl content of the solution (15%), and the amount of antacid additive used (x, %).

The analysis of the corrosion rate on the collected metal samples indicates the strong destructive-corrosive effect of hydrochloric acid in the high area of the distillation column (high-temperature zone).

In addition, the material OL37 (a normal steel) behaves quite well in corrosion, compared to brass, which at high temperatures develops in contact with oil products a corrosion rate 60% higher.

The corrosive effect can be reduced by introducing commercial antacid additives (based on alkyl phenol, having an efficacy of 91–95% at temperatures of 20 °C, 92–97% at temperatures of 40 °C, and 79–97% at temperatures of 70 °C (Figure 3).



Figure 3. Effect of an additive on corrosion reduction.

This viscous inhibitor forms a protective film over the installations, ensuring the reduction of the corrosive effect of hydrochloric acid.

We also used a chemical waste (undistilled polyamide), which provides increased protection at high temperatures.

Polynomial grade 3 equations best describe the corrosive phenomenon without computational errors, compared to exponential equations, which have errors of up to 21%.

4. Conclusions

The presence of hydrochloric acid in atmospheric distillation plants creates the possibility of triggering the corrosion process of the plants, the use of antacid additives being necessary.

The analysis of the specimens taken from the atmospheric distillation installations indicates their risk of corrosion, the presence of the Fe³⁺ ion in the products taken from the installation being observed in the analyses performed.

Finally, we recommend the following procedures to combat or reduce corrosion processes:

- Selection of the most resistant metal material for the corrosive environment in certain working conditions;
- b. Design of machinery or installations with regard to mechanical strength and corrosion resistance;
- c. Making welds and joints by ensuring compatibility between different metal materials;
- d. Injection of corrosion inhibitors.

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