Optimization of the Caustic Treatment of Jet-Fuel to Reduce the Consumption of Caustic Soda in an Oil Refinery

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Abstract: - Jet-Fuel desulfurization is one of the most important processes in the petroleum industries due to the high quality required for aviation fuels. In the pre-sent work, the desulfurization of Jet-Fuel with a content of sulfurous compounds was studied using a process assisted with sodium hydroxide. It was considered that the composition of the turbo fuel varies because different blends of crude oil are processed in the refinery under study. The effects of operating conditions, such as reaction time (15 to 30 min) and the amount of caustic soda (NaOH) in its solution (0.024 to 0.2 mol/L) were investigated. The objective functions (responses) were the NaOH consumption and the total cost of production while the acidity and the total sulfur content of Jet-A1 were considered as restrictions. The optimal conditions of the proposed model were obtained using the optimization with the fmincon function of the MATLAB software. These optimal conditions were found to vary according to the content of sulfur compounds present in the process feed. From the optimal points obtained, it was possible to determine the frequency of change of the NaOH solution that allows its maximum use.

Key-Words: - Desulfurization, jet fuel, neutralization reactions, objective function, optimization, sulfurous compounds.

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1 Introduction

Significant industrial developments, especially those using fossil fuels as the general source, intensely increase environmental pollution and corrosion problems. Exhaust gases of industrial towers consuming fossil fuels contain ample quantities of sulfur compounds that bring about many problems, [1]. Petroleum impurities mainly contain compounds such as H_2S, mercaptans, thiophene, or other sulfur compounds, [2]. Desulfurization, one of the fundamental fields in the prevention of environmental pollution, has been proposed in previous research, [3].

At the Refinery of Cienfuegos, Jet-Fuel desulfurization is carried out using sodium hydroxide (NaOH). The Jet-Fuel produced at this refinery contains mainly hydrogen sulfide (H_2S), light mercaptans (R-SH) and naphthenic acids (RCOOH). The reactions that occur in this process are, [4]:

$$H_2S + NaOH \rightarrow NaHS + H_2O \quad (1)$$

$$H_2S + 2NaOH \rightarrow Na_2S + 2H_2O \quad (2)$$

$$\mathrm{R-SH + NaOH \rightarrow R-SNa + H_2O} \quad (3)$$

$$\mathrm{RCOOH + NaOH \rightarrow RCOONa + H_2O} \quad (4)$$

One of the major operational problems in these systems is the possible formation of stable sodium naphthenate emulsions in the treated raw material. Acid extraction is enhanced by a flow of a high NaOH solution, but the possibility of NaOH eddies in the treated stream also increases. The formation of stable emulsions is more evident when using high concentrations of NaOH, [5]. Generally, when the feed fraction contains high concentrations of naphthenic acids, low concentrations of NaOH should be used to decrease emulsion formation. The fresh NaOH injection flow must be regulated according to the value of the acidity number in the feed, [6].

After the desulfurization treatment, the NaOH is termed spent caustic soda because it is the one that captures sulfur, phenolic, and naphthenic compounds, [7]. Spent soda is a waste classified as hazardous and it has a high pH, [8]. High values of reacted NaOH give the benefits of a lower consumption of fresh NaOH and lower volumes of spent soda to drain. As compensation, there may be problems with the properties of the treated product (acidity value higher than requested) and the tendency to emulsion formation increases, [9].

Today, in the Refinery of Cienfuegos, operational changes have been made to improve the jet fuel desulfurization process. However, large amounts of spent caustic soda are still generated having a high content of free NaOH and it is not treated in the waste plant. The objective of this paper is to optimize the jet fuel desulfurization process to minimize NaOH consumption at the Refinery of Cienfuegos.

2 Problem Formulation

A great variety of crude oils from different parts of the world such as Venezuela, Russia and Algeria enter the Refinery of Cienfuegos, including Mesa 30, Merey 16, Sahara, and Lagomar, blended in various proportions. This causes the amount of sulfur compounds to vary in the raw material of the process being analyzed. That is why seven different blends are used for the jet-fuel desulfurization optimization process. Table 1 characterizes these blends.

At the Refinery of Cienfuegos, the caustic treatment plant for the jet-fuel fraction is designed to process 73 m^3/h. The jet-fuel fraction (feed) reaches the caustic treatment where sulfur compounds are neutralized. This stream is mixed with a sodium hydroxide solution through a mixing valve before entering the vessel where the treatment occurs. The treated stream exits the top of the vessel towards the water wash stage. Chemically, the vessel where the caustic treatment occurs is not particularly sensitive to changes in either temperature or operating pressure. The operating temperature of this unit depends on the temperature of the feed fraction, which must be kept close to 40 °C. The temperature of the feed fraction, the concentration of fresh NaOH, and the degree of consumption of the soda are the most important parameters. These must be regularly monitored to avoid operational problems.

It is necessary to determine the reaction rates for each of the reactions involved. The initial and final NaOH concentrations and the NaOH depletion time are known. These values are obtained from the operational control of the studied processes. With the results obtained from the operational control, the simulation, and the general expressions of reaction speed, the mathematical models that define the reaction speed of NaOH are obtained. The differential method was used, which is based on the real rates of the reactions and measures the slopes of the concentration-time curves. In these systems, bimolecular irreversible reactions are observed, with different initial concentrations of the reactants. [10], state that the equilibrium constants of reactions (Eq. 1) and (Eq. 2) are K1 = 9.0×10^6 L/gmol and K2 = 0.12 L/gmol respectively, at 25°C and infinite dilution. Since the value of K2 is small, it may be assumed that reaction (Eq. 2) does not occur at all and the only reaction taking place in the liquid is reaction (Eq. 1). Considering a batch reactor, the kinetic equations of the reactions that occur in desulfurization are as follows:

$$-r_{NaOH_1} = e^{0.006/T} C_{H_2S}^{0.33} C_{NaOH}^{3.82} \quad (5)$$

$$-r_{(NaOH)_2} = 0.99 e^{0.08/T} C_{R-SH}^{0.238} C_{NaOH}^{3.44} \quad (6)$$

$$-r_{(NaOH)_3} = 0.99 e^{0.16/T} C_{RCOOH}^{0.226} C_{NaOH}^{1.158} \quad (7)$$

Polynomial regression was applied and the equations show a deviation of less than 5%, the adjustments being satisfactory. The global order of the reactions of each process (n_1=4.15, n_2=3.678 and n_3=1.384) confirms the reaction rate and the affinity of NaOH with each sulfur compound present. It is confirmed that high concentrations of NaOH favor the reaction rate with naphthenic acids.

Table 1. Characterizes these blends

Blends	Mesa 30 (%v)	Merey 16 (%v)	Sahara (%v)	Lagomar (%v)	C_{H_2S}(mol/L)	C_{R-SH}(mol/L)	C_{RCOOH}(mol/L)
M1	73.14	-	20.86	-	0.0149	6.51 x 10^{-05}	1.18 x 10^{-14}
M2	19	5	76	-	0.0055	2.58 x 10^{-05}	1.04 x 10^{-14}
M3	-	30.24	69.76	-	0.0104	6.47 x 10^{-05}	1.04 x 10^{-14}
M4	60.86	-	-	39.14	0.0247	0	1.18 x 10^{-14}
M5	100	-	-	-	0.0211	1.05 x 10^{-05}	1.26 x 10^{-14}
M6	-	-	-	100	0.0293	0	1.08 x 10^{-14}
M7	23.06	-	23.06	53.88	0.0207	0	1.09 x 10^{-14}

2.1 Optimization Method

[11], established that multiobjective optimization implies a series of objective functions that are going to be optimized. As in the nonobjective optimization problem, the multiobjective optimization problem usually has many constraints that must satisfy any feasible solution (including the optimal solution). The term "optimize" means to find a solution that provides values, for all objective functions and that is acceptable to the designer. However, if the same values minimize or maximize all the objectives simultaneously, the multiobjective optimization problem can be considered as monobjective. In this investigation, there are 2 objective functions with various constraints. It is desired to optimize as a function of two variables (NaOH concentration and time) and both objective functions must be minimized. The two variables affect the objective functions in the same direction. This means that there is a multi-objective optimization that can be considered a single objective. MATLAB® R2015a software was used to optimize the process; it contains a set of tools to optimize all kinds of equations. Fmincon function was used as the target certain functions are nonlinear functions and the system has equality constraints and inequality.

3 Problem Solution

To perform the optimization, the objective functions and the process constraints were defined.

3.1 Objective Functions

The objective functions are determined by the consumption of NaOH and the total cost of production. In both cases, to achieve the minimum values it is necessary to obtain the optimal NaOH concentration and reaction time.

NaOH consumption

It is necessary to know the optimal concentration of NaOH that allows an adequate quality of the final product, [12]. This function is defined by the reaction rate of NaOH against sulfurous compounds. The kinetic expressions of each reaction are used for that purpose. By design, the process should work with a concentration of 3.3% wt of fresh NaOH. Due to practical experiences, the concentration of soda was initially reduced between 1 % wt and 2 % wt, finally to 0.8 % wt, considerably reducing stable emulsions and satisfactorily complying with the acidity of the finished product. The NaOH solution is considered depleted when it has a concentration of 0.1% wt. According to the literature, vessel volumes are frequently sized to provide a 15 to 30 minutes hold time, [12]. Taking into account that the volume of solution is 25 m3 and that the molar mass of NaOH is 40 kg/kmol:

$$Consumo_{NaOH} = 100t\left[e^{0.006/T}C_{H_2S}^{0.33}C_{NaOH}^{3.82} + 0.99e^{0.08/T}C_{R-SH}^{0.238}C_{NaOH}^{3.44} + 0.99e^{0.16/T}C_{RCOOH}^{0.226}C_{NaOH}^{1.158}\right] \text{(kg)} \quad (8)$$

Constraints: $0.024\,\frac{\text{mol}}{\text{L}} \leq \text{C}_{\text{NaOH}} \leq 0.20\,\frac{\text{mol}}{\text{L}}$;
$15 \text{ min} \leq \text{t(time)} \leq 30\text{min}$

Total cost of production (TCP)

To define the total cost of production function, the procedure proposed by [13], was used (Table 2). According to data available at the refinery, it reached a direct estimate of the fixed capital investment (FCI) of FCI = 24,493,024.8 CUP (24 CUP = 1 USD) and the total capital invested TCI = 26,942,325.6 CUP. Being the price of water of 37.2 CUP/m^3 and of NaOH of 19.1 CUP/kg, [14], the total cost of production is defined as follows:

$$\text{TCP} = \text{Manufacturing cost (MC)} + \text{General Expenses (GE)} \quad (9)$$

$$\text{MC} = \text{direct production cost (DC)} + \text{fixed charges(FC)} + \text{Indirect costs (IC)} \quad (10)$$

$$\text{GE} = \text{Administrative costs} + \text{Distribution and selling costs} + \text{Research and development costs} \quad (11)$$

For the calculation of general expenses (GE), only administrative expenses were considered (0.04 TCP), in this case the other aspects have no impact. Being V_{H_2O} is the volume of water (m^3) used to prepare the NaOH solution used in the desulfurization of jet-fuel and $m_{NaOH}$the mass of NaOH (kg) used in said solution.

$$TCP_{anual} = 12{,}534{,}665.63 + 109.41V_{H_2O} + 56.18m_{NaOH} \tag{12}$$

The refinery operates 330 days in a year, the volume of the solution in the drum is 25 m^3. Therefore, the volume of water and the amount of NaOH consumed in a year is defined as follows:

$$V_{H_2O} = 11{,}880{,}000\left[e^{\left(0{,}006/T\right)}C_{H_2S}^{0.33}C_{NaOH}^{2.82} + 0.99e^{\left(0.08/T\right)}C_{R-SH}^{0.238}C_{NaOH}^{2.44} + 0.99e^{\left(0.16/T\right)}C_{RCOOH}^{0.226}C_{NaOH}^{0.158}\right](\mathrm{m}^3) \tag{13}$$

$$\mathrm{m_{NaOH}} = 19{,}008{,}000\left[e^{\left(0{,}006/T\right)}C_{H_2S}^{0.33}C_{NaOH}^{3.82} + 0.99e^{\left(0.08/T\right)}C_{R-SH}^{0.238}C_{NaOH}^{3.44} + 0.99e^{\left(0.16/T\right)}C_{RCOOH}^{0.226}C_{NaOH}^{1.158}\right](\mathrm{kg}) \tag{14}$$

From the economic point of view, the unit production cost (TCP_U) is the annual production cost divided by the annual production volumes. This will depend on variables that decide the process and its productivity, such as reaction time and NaOH concentration, which allows obtaining an expression as follows, [12]:

$$TCP_U = \frac{TCP_{anual}}{Output} \tag{15}$$

And

$$Output = \frac{V_{jet-A1}}{t} = \frac{396000}{t} \tag{16}$$

Substituting Eq. (12) – Eq.(14) and Eq. (16) in Eq (15):

$$TCP_U = 31.65t\ 3{,}282.35t\left[e^{\left(0{,}006/T\right)}C_{H_2S}^{0.33}C_{NaOH}^{2.82} + 0.99e^{\left(0.08/T\right)}C_{R-SH}^{0.238}C_{NaOH}^{2.44}0.99e^{\left(0.16/T\right)}C_{RCOOH}^{0.226}C_{NaOH}^{0.158}\right] + \left[e^{\left(0{,}006/T\right)}C_{H_2S}^{0.33}C_{NaOH}^{3.82} + 0.99e^{\left(0.08/T\right)}C_{R-SH}^{0.238}C_{NaOH}^{3.44} + 0.99e^{\left(0.16/T\right)}C_{RCOOH}^{0.226}C_{NaOH}^{1.158}\right](\mathrm{CUP}) \tag{17}$$

Components		Composition	Cost (CUP)
Direct Costs ($\boldsymbol{DC = 0.57TCP + 1{,}469{,}581.49 + 37.2V_{H_2O} + 19.1m_{NaOH}}$)			
Raw Material		20 % TCP	
Labor		10% TCP	
Supervision		15 % TCP	
Requirements		1 % TCP	
Maintenance and repair		6 % FCI	1,469,581.49
Supply	Water	37.2$\boldsymbol{V_{H_2O}}$	
	NaOH	19.1$\boldsymbol{m_{NaOH}}$	
	Electricity	10 % TCP	
Laboratory expenses		1 % TCP	
Fixed Charges ($\boldsymbol{FC = 2{,}792{,}204.83}$)			
Depreciation		10 % FCI	2,449,302.48
Taxes		1 % FCI	244,930.25
Insurance		0.4 % FCI	97,972.10
Indirect costs ($\boldsymbol{IC = 0.05TCP}$)			
Other costs		5 % TCP	

3.2 Constraint Function

For future marketing of Jet-A1 fuel as a product of high added value is essential that this meets the quality standards of the market. In this case, only the acidity and total sulfur content will be considered.

Jet-A1 Acidity

The final acidity of Jet-A1 only depends on the reaction of NaOH with naphthenic acid (RCOOH). According to quality standards the final product should have an acid content of 0.011 mgKOH/gJet-A1, then,

$$A = \left[C_{RCOOH} - 0.99e^{0.16/T}C_{RCOOH}^{0.226}C_{NaOH}^{1.158}\right] < 0.011\ \left(mgKOH/g_{Jet-A1}\right) \tag{18}$$

Jet-A1 total Sulfur (S_T)

For total sulfur, only the reactions of NaOH with H_2S and mercaptans (R-SH) are considered. According to quality standards, the final product should have a total sulfur of 0.3 ppm, then,

$$S_T = \frac{10^6}{m_{Jet-A1}}\left[V_{turbo}\left(C_{H_2S} - 34.1te^{\left(0.006/T\right)}C_{H_2S}^{0.33}C_{NaOH}^{3.82} + C_{RSH} - 61.38te^{\left(0.08/T\right)}C_{R-SH}^{0.238}C_{NaOH}^{3.44}\right)\right] \leq 0.3\ (ppm) \tag{19}$$

3.3 Optimization Results

The models obtained are fractional polynomials with two variables to optimize that represent an industrial process.

Table 2. Estimation of production costs

Table 3. Results obtained in the optimization of the desulfurization of the turbo-fuel

Blends	Initial Sulfur (kg)	C_{H_2S} (mol/L)	C_{RSH} (mol/L)	C_{RCOOH} (mol/L)	C_{NaOH} (mol/L)	t (min)
M1	609.48	0.0149	6.51 x 10-5	1.18 x 10-14	0.157	15
M2	142.44	0.0055	2.58 x 10-5	1.04 x 10-14	0.0563	15
M3	193.35	0.0104	6.47 x 10-5	1.04 x 10-14	0.126	15
M4	1006.75	0.0247		1.18 x 10-14	0.2	15
M5	787.80	0.0211	1.05 x 10-5	1.26 x 10-14	0.18	15
M6	1188.37	0.0293		1.08 x 10-14	0.2	15
M7	463.29	0.0207		1.09 x 10-14	0.2	15

Table 4. Results of the evaluation of optimal points

Blends	NaOH consumed (kg)	Rate of change (day)	TCP_U (CUP)	A(mgKOH/$g_{Jet\text{-}A1}$)	S_T (ppm)	t (min)
M1	609.48	0.0149	6.51×10^{-5}	1.18×10^{-14}	0.157	15
M2	142.44	0.0055	2.58×10^{-5}	1.04×10^{-14}	0.0563	15
M3	193.35	0.0104	6.47×10^{-5}	1.04×10^{-14}	0.126	15
M4	1006.75	0.0247		1.18×10^{-14}	0.2	15
M5	787.80	0.0211	1.05×10^{-5}	1.26×10^{-14}	0.18	15
M6	1188.37	0.0293		1.08×10^{-14}	0.2	15
M7	463.29	0.0207		1.09×10^{-14}	0.2	15

The objective functions must be minimized for what they were added, turning the multiobjective problem into a monobjective problem. The software calculation time was approximately 0.5 s.

In addition, it has the advantage that it can be used for other crude oil mixtures where the concentrations of naphthenic acids and total sulfurs vary and it will always have the standard quality standards. However, it is only contemplated for a caustic treatment process, that is, for concentrations greater than 1.0×10^{-4} mol/L and 1.5×10^{-14} mol/L of mercaptans and naphthenic acids respectively, this process does not apply and therefore neither does the model.

Table 3 shows the result of the optimization. The optimal concentration of NaOH and the optimal reaction time are obtained for each blend studied.It is observed that the blends that need the highest content are those with the highest content of naphthenic acids and mercaptans. The high concentrations of NaOH favor the extraction of H_2S and mercaptans but cause the formation of emulsions when reacting with naphthenic acids. It is established that the operating temperature of the studied process is 40 °C, reducing the formation of emulsions that lead to the entrainment of NaOH. On the other hand, increasing the temperature increases the thermal energy of the reaction. This alters the interaction between sodium hydroxide and naphthenic acid, thus decreasing the degree of extraction. Due to this, the compromise that exists between the NaOH concentration and the operating temperature must be taken into account.

From the points obtained, compliance with the quality parameters can be estimated using the functions defined above. Table 4 shows the result of the evaluation of the optimal points. By evaluating the optimal points in Eq. (8), the amount of NaOH consumed in desulfurization can be determined. From this value, it is calculated how often must change the NaOH solution. Using Eq. (17), the minimum production cost in the desulfurization of the jet-fuel of each mixture is calculated. To determine the acidity Eq. (18) was used and the total sulfur content of Eq. (19) was used.

All the blends comply with the established quality parameters. When the solution reaches, the minimum allowed concentration must be changed. It is observed that the NaOH solution can be changed for a longer period than is currently happening in the refinery studied. The NaOH is not completely reacted and a part of it is being sent to waste. If any crude oil blend does not meet the quality parameters for Jet-A1, it is recommended to recirculate the product for a second cleaning step.

4 Conclusion

This study investigated the jet-fuel desulfurization process and the factors affecting NaOH consumption based on operational control data. It was confirmed that high concentrations of NaOH favor the rate of reaction with naphthenic acids. The compromise between temperature and NaOH concentration must always be taken into account to avoid the formation of emulsions. Defined mathematical models for optimization can be considered predictive tools for technologists of the studied processes. Therefore, it is possible to extend its implementation in the analysis of blends used by the refinery in the future.

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