



Improvement of treatments process by oily wastewaters coagulation and flocculation applied at Haoud Berkaoui production center, Algeria

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ABSTRACT

Haoud Berkaoui is one of the most important oil-producing regions in Algeria. It covers three productive fields namely: Haoud Berkaoui, Guellala and Benkahla. Haoud Berkaoui's production center operations generate daily significant amounts of oily wastewater contaminated with hydrocarbons and suspended matters. At this field is set at each production center a deoiling station to ensure both hydrocarbons recovery and the production of reinjection water. These stations use three conventional techniques for oily wastewater treatment, namely: coagulation, flocculation and sedimentation. In order to improve the efficiency of the implemented process, three new coagulants were developed and tested. For settling period set at 30 min, dosage effects of coagulants ranging from 8 to 40 mL/L of treated oily wastewater were studied for turbidity elimination, suspended matter and hydrocarbons. The use of the coagulant composed of 4% sodium silicate and 1.4% sulfuric acid at a rate of 40 mL/L of treated oily wastewater gave the best results in terms of treatment by ensuring pH normalization, suspended matter; elimination of turbidity and reduction of hydrocarbons content by 81%.

Keywords: Haoud Berkaoui; Deoiling station; Oily wastewater; Coagulation; Flocculation; Sedimentation

1. Introduction

Oil industry is the most important development sector in Algeria's economy. It is based on three essential axes: prospecting, exploitation and secondary recovery. Haoud Berkaoui region is one of the main production areas of the Triassic province in Algeria. It includes three productive fields namely: Haoud Berkaoui, Guellala and Benkahla. Haoud Berkaoui center ensures: crude production, flared gas recovery, water injection and oily wastewater treatment.

These activities generate daily large quantities of oily water contaminated by hydrocarbons and suspended matter [1].

Oily water pollution is mainly manifested in the following aspects: (1) affecting drinking water and groundwater resources, endangering aquatic resources; (2) endangering human health; (3) atmospheric pollution; (4) affecting crop production; (5) destructing the natural landscape, and even probably because of coalescence of the oil burner safety issues that arise [2,3].

As part of the company's policy implementation about environmental protection, the Sonatrach Group establishes

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an ambitious program that aims to eliminate or reduce all significant environmental impacts related to its activities and services. And it is in accordance with the legislative texts, in particular the law n° 83-03 of 05/02/1983, related to the environment protection. The purpose of this act is to develop a national environmental protection policy that seeks the protection of natural resources by preventing and fighting, restructuration and natural resources development all forms of pollution and nuisance, also to improve environment and life quality.

As part of this policy application, a deoiling station is set for each production center at Haoud Berkaoui. These stations with a treatment capacity of 100 m³/h ensure both the hydrocarbons recovery and the reinjection water production. They use coagulation and flocculation as a process for treating oily wastewater derived from fields and various activities in production centers [1].

Coagulation and flocculation is a widely used process for the treatment of oily water. It is often recommended for the removal of suspended solids as well as unwanted or toxic substances; it is simple, efficient and consumes little energy [4–6]. It is known to be useful in environmental protection and human health [7]. These two phenomena are related and generally occur together [8,9].

The purpose of the coagulation–flocculation operation is to remove suspended solids and colloids by their destabilization then formation of flocs by absorption and aggregation, the flocs thus formed will be decanted and filtered afterwards [10–14]. The main factors influencing the coagulation process are pH, coagulant dose, agitation, temperature, turbidity and dissolved salts [15,16].

The objective of this study is to improve the treatment process used at the deoiling station of Haoud Berkaoui production center by developing three new coagulants and testing their effect on the treatment scheme implemented.

2. Materials and methods

2.1. Deoiling station of Haoud Berkaoui production center

Deoiling station at Houad Berkaoui center ensures hydrocarbons recovery and reinjection water production. Characteristics value threshold must be below 5 mg/L for the hydrocarbons content, 30 mg/L for suspended matter and a pH between 6.5 and 8.5. The technological scheme of oily wastewater treatment process applied at the deoiling station is presented in Fig. 1. It shows that hydrocarbons and suspended matter removal follows the described steps in the next paragraphs [1].

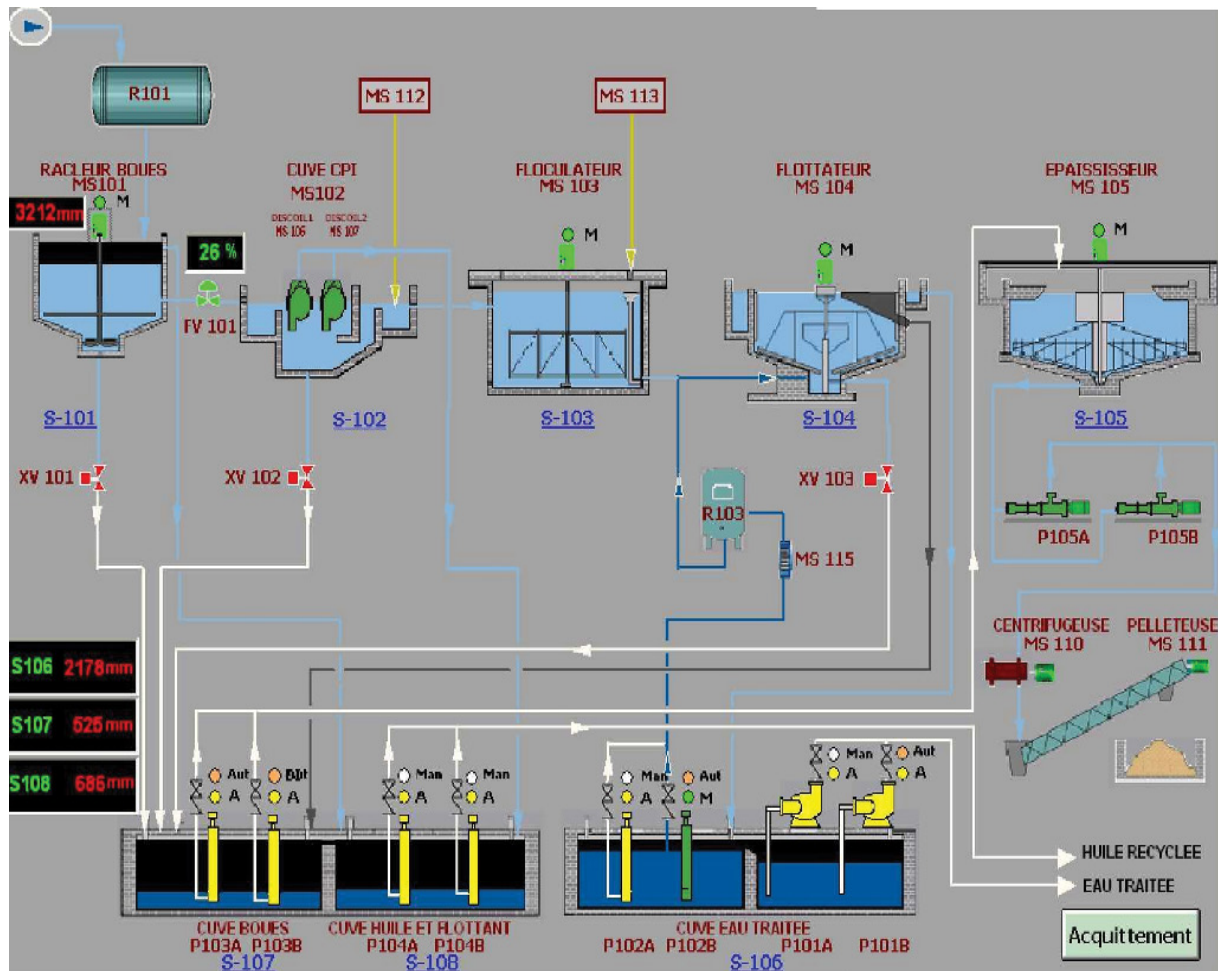


Fig. 1. Technological scheme of deoiling station [1].

A flash tank is placed upstream of the deoiling station in order to separate oily wastewater and gases. Gases are sent to the torch while the effluents collected at the bottom of the flash tank are sent to the buffer tank S-101. In this buffer tank, floating oil is recovered by a cream system then sent to the oil tank S-108 while suspended matter forming the sludge are gathered by a scraper MS 101 and sent to a sludge tank S-107 through an automatic valve. XV 101.

Effluent leaving the buffer tank is channeled to the C.P.I MS102 tank or the supernatant oil is sent by a rotary disk oil separator MS106 and MS107 to the oil tank S-108. Sludge is discharged through the automatic valve XV102 to the sludge tank S-107 and the water charged with suspended solids is channeled to the flocculation tank S-103 for a possible treatment which consists to add a coagulant at the inlet and a flocculant at the outlet. These two chemical agents are provided by chemical dosing units MS112 and MS113.

Water entering to the flotation tank S-104 by gravity mixes with air saturated water. Floccs scraped off water surface and the sludge formed are also fed to the sludge tank. While treated water is sent to a treated water tank S-106 consisting of two compartments, each comprising two pumps. The pumps P101A/B represses treated water to outside, when the pumps P102A/B recirculates a part of the treated water to an air saturation tank R-103 for possible mixing with the fluid exiting from flocculation tank S-103.

The sludge collected in the S-107 tank is pumped by the P103A/B pumps to a sludge thickener S-105 in which the scraper MS105 ensures the sludge concentration homogeneity. After that, it will be sent by P105A/B pumps to the centrifuge MS110 for dehydration, then outside the hangar where they are removed by mechanical shovel and buried in impermeable trenches.

2.2. Sampling of oily wastewater

Taking a water sample is an operation to which greater care must be taken; it conditions the analytical results and the interpretation that will be given [17]. The oily wastewater was sampled at the entrance of the deoiling station. The oily wastewater sampling operation was carried out quickly to avoid any change in its quality and characteristics. It was poured into rinsed and dried glasses bottles; if necessary, propylene bottles can also be used. The bottles were stored at standard temperature and sent to the laboratory for the studied parameters analysis.

2.3. Analysis of oily wastewater

Oily wastewater quality parameters analyzed in this study are: pH measured using a HACH pH meter, suspended

matter (SM), turbidity and hydrocarbons content (HC) evaluated by means of a DR/2000 spectrophotometer under different spectral conditions as shown in Table 1 [17].

2.4. Preparation of coagulants

In this study, three sodium silicate-based coagulants were prepared according to the operating conditions presented in Table 2.

2.5. Coagulation tests

In order to choose the coagulant and the optimal treatment dose, the coagulation and flocculation tests were carried out according to the jar test protocol using an ISCO brand flocculator. It is composed of six beakers of one liter capacity and six mechanical agitators comprising 1.5 cm × 8.5 cm blades which rotate from 20 to 250 rpm. The coagulation and flocculation treatment steps applied to oily wastewater are as follows:

- A rapid stirring phase at 80 rpm for 15 min, after adding increasing doses (4–20 mL) of each of the 03 coagulants solutions previously prepared to 500 mL of oily wastewater;
- A slow stirring phase at 30 rpm for 10 min, that followed the addition of 1 mL of kurifix (floculant);
- A decantation phase for at least 30 min [18,19].

A volume of 25 mL from each beaker is taken in order to measure the following parameters: pH, suspended matter (SM), turbidity and hydrocarbons content (HC). Percentage removal of each parameter is calculated by using Eq. (1):

$$C(\%) = \frac{C_i - C_f}{C_i} \times 100 \quad (1)$$

where C_i and C_f correspond to initial and final contents of studied parameter.

3. Results and discussions

3.1. Characterization of oily wastewater

Oily wastewater treated at the deoiling station is characterized by a slightly acidic pH, high turbidity and high levels of suspended matter and hydrocarbons content, as shown in Table 3.

Comparison of obtained results with standards of industrial liquid effluents discharges applied, in Algeria and at

Table 1
Spectral conditions for analysis with a DR/2000 spectrophotometer

Parameters	Wavelength (nm)
Suspended matter	810
Turbidity	750
Hydrocarbons	410

Table 2
Operating conditions for the preparation of coagulants

Designation of the coagulant	Preparation water	Sodium silicate (mass %)	Sulfuric acid (mass %)
Coagulant (C ₁)	Service water	4	1.2
Coagulant (C ₂)	Service water	4	1.4
Coagulant (C ₃)	Service water	4	2

Haoud Berkaoui production center, shows that apart pH, which is close to standards, parameters analyzed present values largely exceeding these standards, which reflects significant pollution of oily wastewater treated at the deoiling station. In 2016, an analysis carried out by Sellami et al. [19], in the same deoiling station gave similar results.

3.2. Effect of composition and dosage of coagulants

In the process of coagulation and flocculation, coagulant's dose is one of the most important factors considered in the coagulant performances study. Insufficient or over-dose coagulants will bring poor treatment performance. Therefore, it is important to define the optimal coagulants dosage to minimize dosage cost, sludge formation and also to obtain the optimum treatment performance [20–23]. The following paragraphs show the effect of coagulant doses on the treatment process.

3.2.1. Coagulant C₁

Results relating to coagulant C₁ dose's effect studied on the treatment process are presented in the following figures: variation of pH is represented in Fig. 2, suspended matter in Fig. 3, Fig. 4 illustrates turbidities evolution and Fig. 5 reveals hydrocarbons content variation.

Treatment of oily wastewater with increasing coagulant C₁ doses is from 4 to 20 mL per 500 mL of treated oily wastewater. This allows pH increase and a decrease in the rest of parameters analyzed. Different doses of coagulant C₁ generate pH increase from 4.46 to 6.69, suspended matter rate reduction from 151.34 to 65.8 mg/L, turbidity reduction from 484.90 to 0 NTU and reduction of hydrocarbons content from 514.30 to 118.15 ppm.

Table 3
Characteristics of oily wastewater treated at the deoiling station

Parameters	Content	Algerian Liquid Discharge Standard	Standards applied at Haoud Berkaoui
pH	6.14	6.5–8.5	6.5–8.5
Suspended matter (SM) (mg/L)	658	35	≤30
Turbidity (NTU)	746	–	30
Hydrocarbons content (HC) (ppm)	695	10	≤5

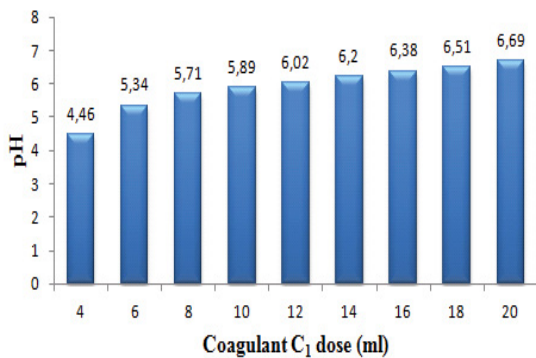


Fig. 2. Effect of coagulant C₁ dose on the pH.

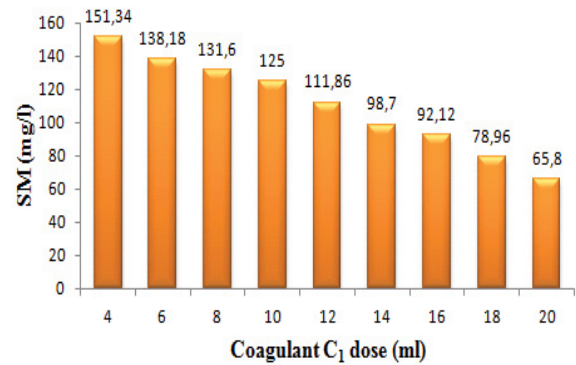


Fig. 3. Effect of coagulant C₁ dose on the suspended matter (SM).

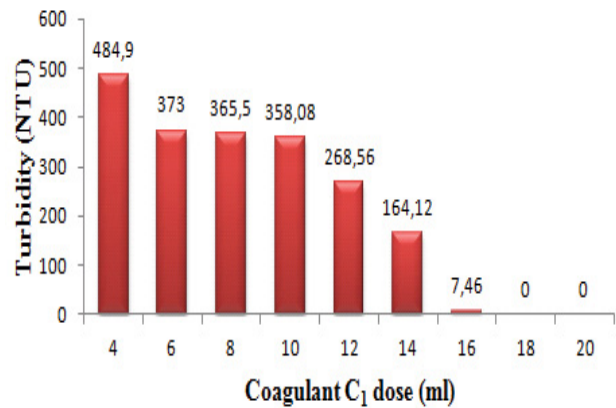


Fig. 4. Effect of coagulant C₁ dose on the turbidity.

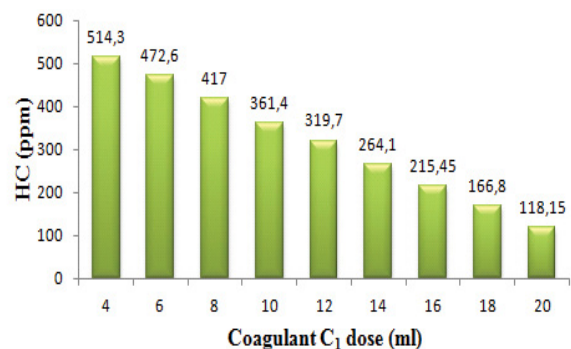


Fig. 5. Effect of coagulant C₁ dose on the hydrocarbons content (HC).

A coagulant C_1 dose of 20 mL per 500 mL of treated oily wastewater, that is, 40 mL/L ensures pH normalization, turbidity elimination, suspended matter reduction by 90% and decrease hydrocarbons content by 83%.

3.2.2. Coagulant C_2

Coagulant C_2 added dose's effects on treated oily wastewater are illustrated in the following figures: variation of the pH is represented in Fig. 6, suspended matter in Fig. 7, Fig. 8 illustrates turbidity evolution and Fig. 9 reveals variation of the hydrocarbons content.

With C_2 coagulant, we found that, apart pH, studied parameter contents decreased progressively with the increase of added doses. Different coagulants C_2 doses ensure pH increase from 5.03 to 8.5, reduction in suspended matter rate from 144.76 to 30 mg/L, important turbidities diminution from 373 to 0 NTU and reduction in hydrocarbons content from 465.65 to 132.05 ppm.

In this case, a coagulant C_2 dose of 20 mL per 500 mL of treated oily wastewater, that is, 40 mL/L guarantee pH and suspended matter normalization, turbidity elimination and hydrocarbons content reduction by 81%.

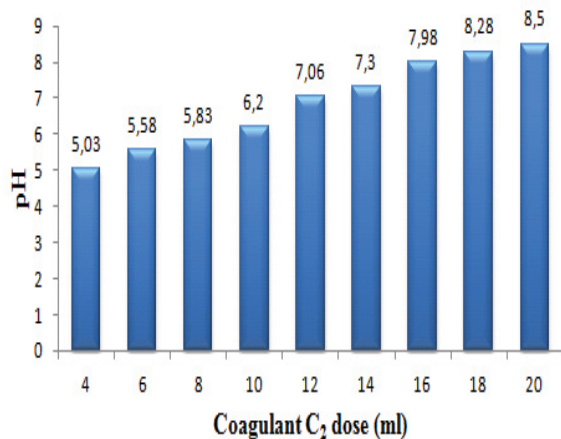


Fig. 6. Effect of coagulant C_2 dose on the pH.

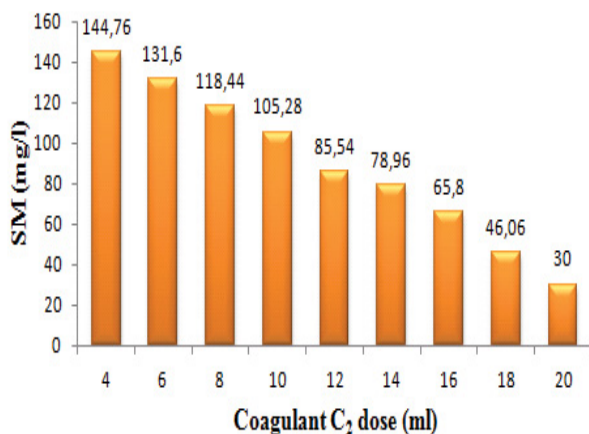


Fig. 7. Effect coagulant C_2 dose on the suspended matter (SM).

3.2.3. Coagulant C_3

Results relating to coagulant C_3 doses effect on the treatment process are presented in the following figures: pH variation is represented in Fig. 10, suspended matter in Fig. 11, Fig. 12 illustrates turbidity evolution and Fig. 13 reveals hydrocarbons content variation. They reflect same comments observed when using previous coagulants in terms of changes in parameters analyzed with the dose.

Diverse coagulant C_3 doses generate pH increase from 4.35 to 12.34, reduction in suspended matter rate from 85.54 to 71.06 mg/L, turbidity diminution from 92.50 to 52.22 NTU and hydrocarbons content reduction from 118 to 48.56 ppm.

Although 20 mL coagulant C_3 dose per 500 mL of treated oily wastewater, that is, 40 mL/L ensures a reduction in hydrocarbons content rate about 93%, it produces basic, cloudy water loaded with suspended matter. Therefore, 2% sulfuric acid quantity added during this coagulants preparation did not ensure the expected effectiveness.

3.3. Selection of the best coagulant and optimal treatment dose

Through the analysis relating to each coagulant, it could be seen that for each preparation, treatment effectiveness

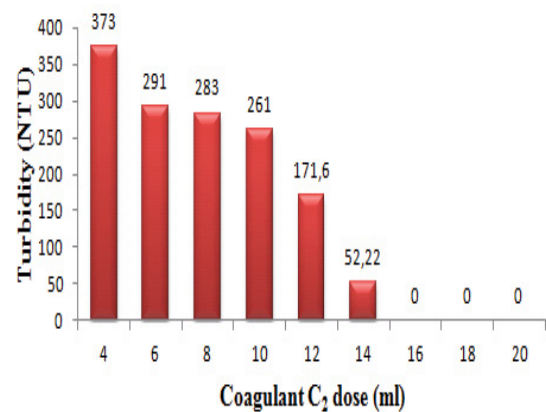


Fig. 8. Effect of coagulant C_2 dose on the turbidity.

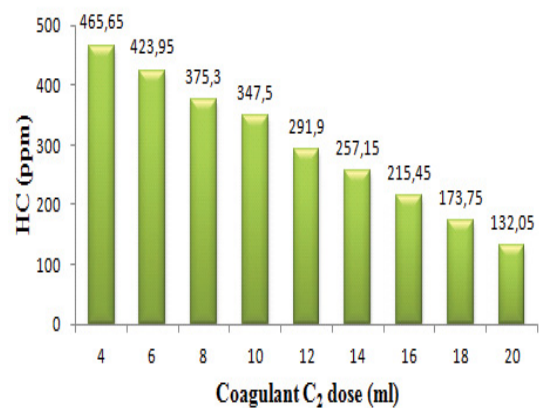


Fig. 9. Effect coagulant C_2 dose on the hydrocarbons content (HC).

increases directly with coagulant dose. Thus, the optimum dose for each prepared coagulant is 40 mL/L of oily wastewater treated.

As shown in Fig. 14, the coagulant C₂ showed the best treatment efficiency. It ensures normalization of two parameters (pH at suspended matter), turbidity elimination and a significant reduction in hydrocarbons content level reaching 81%.

pH values measured during coagulant C₂ addition vary from 5.03 to 8.5 which correspond to the upper limit of rejections norms when using the optimal dose. In 2015, at the same deoiling station, Sellami et al. [24] have studied pH variation according to the activated silicates volume for two cases. Firstly, without adding ascorbic acid, and secondly with addition of this chelating agent. In the first case, all measured pH values were between 4.83 and 4.72, which mean that addition of activated silicates slightly decreases pH solution, even in absence of ascorbic acid. This slight acidity allows eventual use of other treatment products which were effective in this pH environment. In presence of ascorbic acid, pH solution increases slightly from 4.83 to 5. These values are far from standards required for this wastewater type.

The study also shows that removal of suspended matter is 87.95% in absence of ascorbic acid and reaches 93.43% in case of its presence for the same quantity of silicates [24]. The

last value is close to our results where the reduction rate of suspended matter is 95.44 %, thus ensuring this parameter normalization. The results obtained using coagulant C₂ are also better in comparison with recent study results realized in 2020 by Mohamed et al. [25]. These authors prepared five new poly inorganic coagulants, namely poly hydrolyzed aluminum chloride, poly ferric chloride poly aluminum hydroxyl sulfate, hydrolyzed poly aluminum ferric chloride and hydrolyzed poly aluminum ferric chloride with silicate. The investigation of these coagulants in wastewater treatment shows that the highest suspended matter removal percentages reached 92%.

Turbidity values explain the presence of suspended particles and colloids in water. This study is a real challenge, indeed a coagulant dose of 16 mL per 500 mL of treated oily wastewater is sufficient to completely eliminate the water turbidity. According to Sellami et al. [24] research, the absence of chelating agent, the lower value obtained for the turbidity is 15 NTU. Despite the absence of a complexing acid, the excess of activated silicates in water makes it colloidal. In the presence of ascorbic acid as sequestering agent, so turbidity value decreases to 09 NTU.

Petroleum hydrocarbons contained in oily wastewater represent a significant challenge for conventional treatment plants, which often do not adequately remove these pollutants. To limit pollution by petroleum products and improve discharge water quality, more efficient wastewater treatment

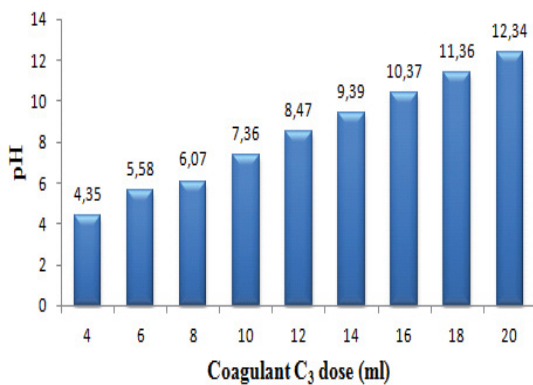


Fig. 10. Effect of coagulant C₃ dose on the pH.

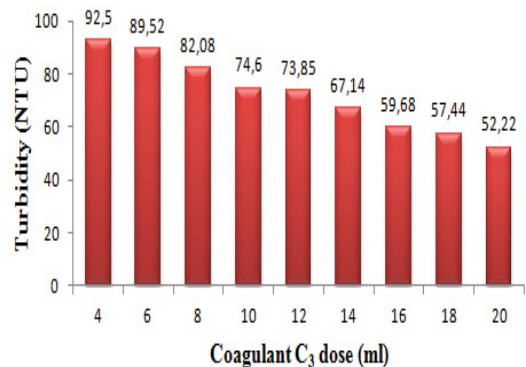


Fig. 12. Effect of coagulant C₃ dose on the turbidity.

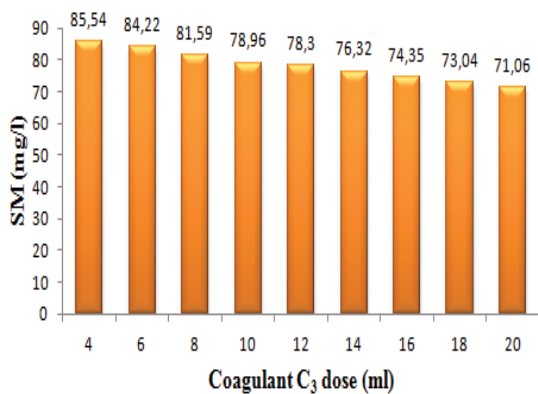


Fig. 11. Effect of coagulant C₃ dose on the suspended matter (SM).

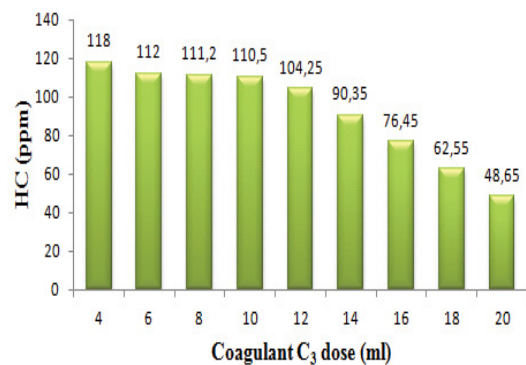


Fig. 13. Effect of coagulant C₃ dose on the hydrocarbons content (HC).

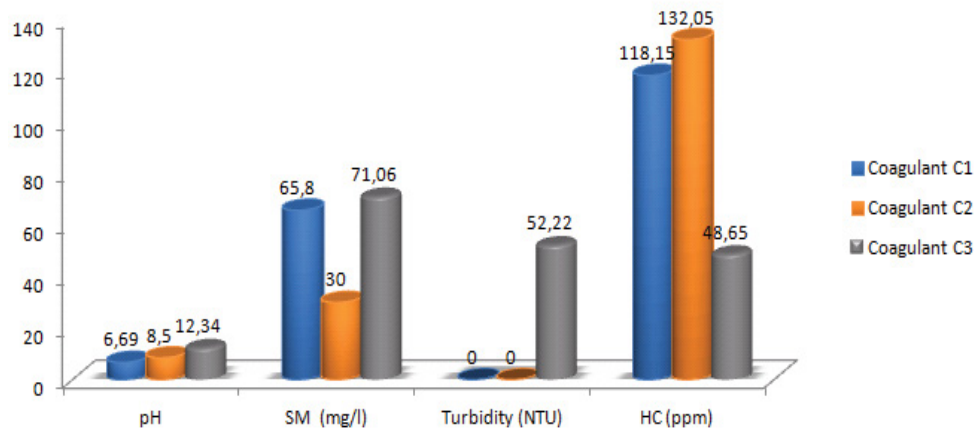


Fig. 14. Comparison of the treatment effectiveness of the three coagulants at optimal dose.

technologies are continually being researched, developed and implemented. Governments, companies, and researcher's groups around the world devote considerable efforts and resources to remove oil pollutants from water [26,27].

At present, coagulation–flocculation as an indispensable oily wastewater treatment technology receives much attention because it is very well established, economical, practical and relatively efficient. Given strict emission standards and the refractory nature of oily wastewater, the combination process with coagulation/flocculation, such as electrocoagulation, coagulation-membrane filtration hybrid process, and coagulation/flocculation–flotation can present better application potential [28–31].

In the current study we were able to remove significant levels of hydrocarbons ranging from a minimum of 81% when using the coagulant C_2 and could reach 93% when using the coagulant C_3 composed of 4% sodium silicate and 2% of sulfuric acid. In 2016, Sellami et al. [19] achieve a reduction rate of 97.32% using a coagulant composed of 4% sodium silicate and 2% ascorbic acid. However, the use of coagulant C_3 coagulant is not recommended, despite it ensures the best efficiency in the treatment of hydrocarbons content. In fact, doses greater than or equal to 40 mL/L produce water with basic pH, cloudy and loaded with suspended matter.

4. Conclusion

The petroleum industry is a water-intensive sector that produces both wastewater from extraction and refining processes. The release of these large wastewater quantities in nature without any treatment induces harmful effects on humans and environment by contaminating the soil and consequently the groundwater which constitutes the only source of drinking water supply for people of the region. At the same time, the reinjection of these waters into the oil wells will lead to an increase in oil pressure, clogging of the wells and formation of scale in the discharge wells.

Permanent evolution of the quality of wastewater produced at Haoud Berkaoui production center, requires periodic determination of treated water quality and operating conditions optimization of treatment process based on coagulation and flocculation.

The analysis of oily wastewater at the entrance of deoiling station gives a slightly acidic pH, high turbidity and high rate of suspended matter. Hydrocarbons content exceeds the standards applied in Algeria and more than the stricter standards applied in Haoud Berkaoui production center. Thus, the improvement of the coagulation and flocculation process put in place consists to modify coagulants composition by preparing three new coagulants which we have named C_1 , C_2 , C_3 composed of 4% sodium silicate while varying the activation rate with sulfuric acid by, respectively, 1.2%, 1.4% and 2%.

As a result, it appears that coagulant C_1 ensures normalization of pH, elimination of turbidity, reduction of suspended matter by 90% and reduction of hydrocarbons content by 83%, while coagulant C_2 gives the best results in terms of treatment by ensuring pH normalization and suspended matter, turbidity elimination and hydrocarbons content reduction by 81%. Coagulant C_3 is not recommended although it ensures the best efficiency in the hydrocarbons treatment, about 93% because it produces basic, cloudy water loaded with suspended matter.

Furthermore, this research demonstrates also correlation between coagulants dose and treatment effectiveness. It pointed out that the three coagulants optimal dose is 40 mL/L of oily wastewater.

At the end of this research, we can say that under the current operating conditions of Haoud Berkaoui deoiling station, it is recommended to opt for coagulant C_2 based on 4% sodium silicate and 1.4% sulfuric acid at a rate of 40 mL per oily wastewater liter giving the best results for the majority of studied parameters.

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