The sedimentation of mixed cultures used in the treatment of effluents generated from terrestrial fuel distribution terminals

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ABSTRACT

This study evaluated the use of coagulants (ferric chloride and aluminium sulphate) and an anionic polyelectrolyte (polyacrylamide) in the settling of a mixed culture (C_1), which was used for the biodegradation of hydrocarbons present in effluent of fuel distribution terminals contaminated with diesel oil and gasoline. In preliminary investigations conducted in jar tests, the optimal concentrations of coagulant were obtained for further studies. After preliminary tests, biodegradation was evaluated in a central composite design (CCD) with varying concentrations of ferric chloride and polyelectrolyte. Ferric chloride and polyelectrolyte concentrations ranged from 77.9 to 422.12 mg/L and 0.0 to 3.2 mg/L, respectively. The responses monitored in CCD experiments included the sludge volume index (SVI), turbidity, and specific rate of oxygen uptake (SOUR), where values of 100 mL/g, 840 nephelometric turbidity unit (NTU) and 58 mg O_2/g h, respectively, were obtained. Subsequently, biodegradation was monitored in a sequencing batch reactor (SBR). The results indicated that within five cycles, total petroleum hydrocarbon (TPH) removal increased from $75 \pm 1.0\%$ to $79 \pm 0.5\%$, while the volatile suspended solids (VSS) increased from 1300 to 2500 mg/L.

1. Introduction

Coagulation/flocculation is a commonly used process in water and wastewater treatment in which compounds such as ferric chloride and/or polymers are added to wastewater to destabilise colloidal materials and cause small particles to agglomerate into large flocs that settle out of solution [1].

Chemical coagulation with aluminium sulphate, ferric chloride, ferric sulphate and lime has been studied extensively [2–5]. Recently, the use of synthetic polyelectrolytes as flocculants for the removal of suspended solids in wastewater treatment has grown rapidly [5–7]. Coagulation and flocculation, followed by sedimentation or flotation, may be used in various stages of sewage treatment prior to disinfection, from the processing of crude influent to post-treatment procedures. Another application of coagulation is the sedimentation of cultures used in the biodegradation of effluent. Research and practical applications have shown that coagulation will lower the pollution load and could generate an adequate water recovery [8–13]. Most systems use aerobic processes for the treatment of hydrocarbon residues in soil or groundwater because aerobic conditions remove more pollutants in less time. Although they are more effective, aerobic processes are more expensive than anaerobic processes [14].

Currently, treatment processes for waste oil consist primarily of aerobic methods, which are expensive due to the cost of oxygen. Vieira et al. [14–16] studied the biodegradation of an effluent contaminated with diesel oil and gasoline using intermittent aeration as an alternative to continuous aeration. Results revealed that intermittent aeration is an interesting and economic option for hydrocarbon removal. However, difficulties were observed in the flake formation of C_1 mixed cultures, resulting in decreased settling. Problems associated with flake formation could be due by vigorous agitation, which promote rapid homogenisation of diesel oil and gasoline in the effluent. Without proper stirring, phase separation occurred, preventing removal of total petroleum hydrocarbon (TPH). Alternatively, the addition of coagulants could be used to increase flake formation and the settling of microorganisms. An increase in flake formation caused the cultures to become mechanically resistant, facilitating the treatment of contaminated effluents in continuous systems. Thus, the object of this study was to evaluate the sedimentation of C_1 mixed cultures after the addition of coagulants (ferric chloride or aluminium sulphate) and polyelectrolytes (anionic polyacrylamide), by monitoring the sludge volume index (SVI), turbidity and specific oxygen uptake rate (SOUR) of the supernatant. Furthermore, cell growth behaviour and removal after each successive cycle was verified in a sequencing batch reactor (SBR).

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Nomenclature

CCD	central composite design
FC	ferric chloride concentration (mg/L)
NTU	nephelometric turbidity unit
PC	polyelectrolyte concentration (mg/L)
SBR	sequencing batch reactor
SOUR	specific rate of oxygen uptake (mg O_2/g h)
SVI	sludge volume index (mL/g)
TPH	total petroleum hydrocarbon
VSS	volatile suspended solids (mg/L)

2. Materials and methods

2.1. Contaminated effluent

Effluent samples were collected from a contaminated lagoon according to the procedures recommended by the Associação Brasileira de Normas Técnicas in document ABNT/NBR 9898 [17] titled "Preservation and sampling techniques of liquid effluents in receptor bodies", according to Vieira et al. [14]. After collection, samples were stored at 4 °C.

2.2. Microorganisms

The C_1 mixed culture employed in this study was identical to the culture used by Vieira et al. [14]. The microorganisms present in this culture were previously isolated and identified [14].

2.3. Biodegradation of the effluent

2.3.1. Reactor

A reactor specifically configured for this study was constructed of acrylic plastic and sealed with rubber stoppers. The 5 L reactor contained six ports connected to a mechanical agitator, thermometer, dissolved oxygen probe, gas outlet, perforated stainless steel sample collector, and a pH meter. The configuration of reactor is identical to the system used by Vieira et al. [14] and is displayed in Fig. 1.

2.3.2. Operating conditions

After an adaptation period, the culture was inoculated into effluent contaminated with diesel and gasoline, as described by Vieira et al. [14,16].

The conditions optimised by Vieira et al. [14,16] were adopted in this study and included the following: 0.509 g/L of nitrogen, 0.068 g/L of phosphorous, 1.32 g/L of inoculum, and 2 g/L of autolysed residual brewery yeast (ARBY). Intermittent aeration consisted of air injections at a flow rate of 150 L/h for 5 min every 33 h agitation at 110 rpm. The air flow rate was measured using a rotameter and was controlled by an in-line valve. The experiments were conducted for 3 days.

2.4. Coagulants

Aluminium sulphate ($Al_2(SO_4)_3 \cdot 14–18H_2O$) and ferric chloride ($FeCl_3 \cdot 6H_2O$) were selected as coagulants. Anionic polyacrylamide was used as a polyelectrolyte.

2.5. Preliminary tests of coagulation

After 3 days of effluent biodegradation in the presence of C_1 in a batch reactor, the resultant solution (effluent and culture) was used in preliminary coagulation tests. In these experiments, the concentrations of ferric chloride and aluminium sulphate were varied from 100 to 1000 mg/L to determine the optimal coagulant and concentration range.

Jar test experiments were performed with a jar test apparatus equipped with six 250 mL beakers. Different combinations of aluminium sulphate (100, 200, 300, 400, 600, 800, and 1000 mg/L) and ferric chloride (100, 200, 300, 400, 600, 800, and 1000 mg/L) were tested to determine the optimal range of coagulant concentrations. The sludge was stirred for 2 min at 80 rpm, followed by; slow mixing for 15 min at 30 rpm. Flocks were allowed to settle for 30 min. After settling, the turbidity and SVI of the supernatant were determined.
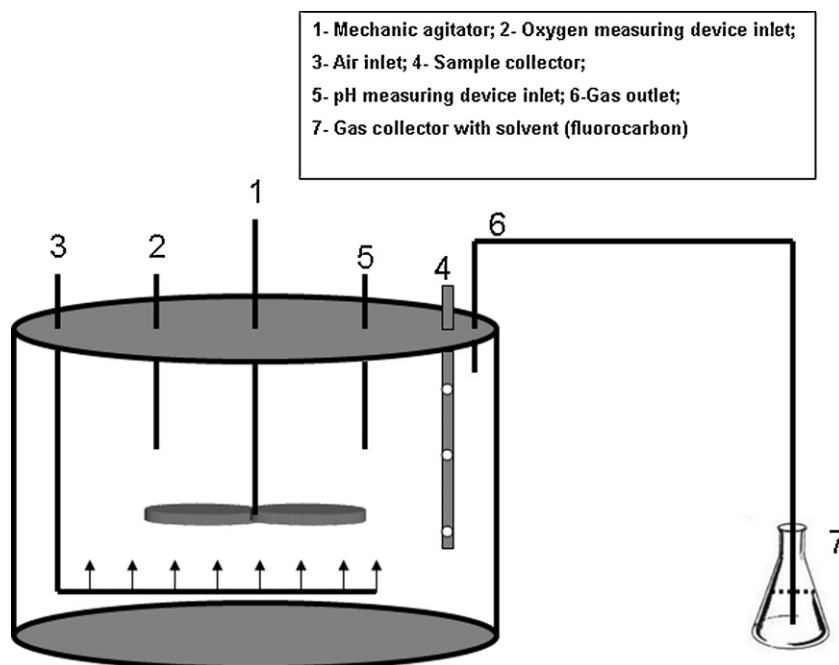

1- Mechanic agitator; 2- Oxygen measuring device inlet;
3- Air inlet; 4- Sample collector;
5- pH measuring device inlet; 6-Gas outlet;
7- Gas collector with solvent (fluorocarbon)

Fig. 1. Reactor scheme.

2.6. Central composite design (CCD)

After culture growth in the effluent, varying concentrations of ferric chloride and polyelectrolyte were evaluated by monitoring the settling of C_1 cultures.

A central composite design (CCD) was employed including two variables ($k = 2$), three replicates at the central point (CP = 3), four experiments at the axial point ($2 \times k = 4$), and an orthogonal α of 1.147, resulting in 11 experiments. The experiments were repeated in triplicate to obtain average values.

CCD experiments were conducted in jar tests. After the addition of coagulant, the system was agitated at 80 rpm for 2 min. The polyelectrolyte was added and the sludge was stirred for a period of 2 min at 80 rpm, followed by mild agitation.

In this study, the independent variables were X_1 (ferric chloride concentration) and X_2 (polyelectrolyte concentration), while the dependent variables included SVI, turbidity, and SOUR.

In preliminary tests, an actual ferric chloride concentration of 300 mg/L was observed in the upper level (+1) of the CCD, while a concentration of 100 mg/L was observed in the lower level (−1). According to the literature [18], the polyacrylamide concentration in the upper level (+1) should be 3 mg/L. Alternatively, a polyacrylamide concentration of 0.0 mg/L was maintained in the lower level (−α) de 0.0 mg/L, to verify the response to ferric chloride in the absence of polyacrylamide.

2.7. Operation of sequencing batch reactor (SBR)

The aforementioned reactor, displayed in Fig. 1, was used in SBR experiments. Conditions optimised by Vieira et al. [14] were adopted, which included the following: 0.509 g/L of nitrogen, 0.068 g/L of phosphorous, 1.32 g/L of inoculum, and 2 g/L of autolysed residual brewery yeast (ARBY). Intermittent aeration consisted of air injections at a flow rate of 150 L/h for 5 min every 33 h. The air flow rate was measured with a rotameter and was controlled by an in-line valve. The system was maintained at $29 \pm 2\,°C$ and was agitated continuously at 110 rpm. At beginning of the process, the pH was adjusted to 7.0.

Previously optimised concentrations of $FeCl_3 \cdot 6H_2O$ and polyelectrolyte were used in the SBR.

The total cycle period was 73 h, consisting of 15 min of filling, 72 h of reaction with intermittent aeration, 30 min of settling, and 15 min of withdrawal. At the beginning of each cycle, immediately after withdrawal (earlier sequence), a pre-defined feed volume was added to the system. At the beginning and end of each cycle, VSS, TPH removal and SOUR were analysed.

2.8. Analytical techniques

The concentration of TPH was analysed according to the methodology described by Vieira et al. [15]. The volatile, total and fixed suspended solids, along with the SVI and SOUR were determined according to Standard Methods for the Examination of water and wastewater [19].

Turbidity was measured with a turbidity meter (model 2100N, Hach).

3. Results and discussion

3.1. Preliminary tests of coagulation with ferric chloride and aluminium sulphate

The SVI and turbidity obtained in preliminary tests are shown in Fig. 2. An increase in the concentration of ferric chloride resulted in an increase in the SVI and a decrease in turbidity. Therefore, it was necessary to optimise the turbidity to obtain an appropriate SVI. A ferric chloride concentration of 300 mg/L resulted in an SVI of 100 mL/g. According to Ahmad et al. [13], an SVI of 100 mL/g or less indicates a sludge with good settling properties. Thus, SVI values near of 100 mL/g are desired to assure that the sludges produced in the coagulation–flocculation process have sufficient settling characteristics. Aluminium sulphate concentrations between 400 and 800 mg/L provided an SVI of 80–120 mL/g.

The contaminated effluent had an initial turbidity of 2200 NTU. For comparison, the brute effluent was centrifuged at $18,500 \times g$ (corresponding to 12,500 rpm) for 20 min, yielding, supernatant with a turbidity of 750 NTU. This value was used as a reference for future experiments.

As shown in Fig. 2, the turbidity of the supernatant from jar tests with ferric chloride was lower than experiments with aluminium sulphate, which indicated that ferric chloride was a more efficient coagulant than aluminium sulphate. Specifically, an aluminium sulphate and ferric chloride concentration resulted in a turbidity of 1600 and 1100 NTU, respectively.

An aluminium sulphate concentration of 400 mg/L caused the turbidity to decrease to 1350 NTU, while a ferric chloride concentration of 400 mg/L resulted in a turbidity of 950 NTU. At this coagulant concentration, the addition of a polyelectrolyte can provide a greater reduction in turbidity without promoting an excessive increase in the SVI.

Amuda and Amoo [1] analysed ferric chloride concentrations of 100, 300, 500, 750 and 1000 mg/L in jar test experiments and found an optimal ferric chloride concentration of 300 mg/L. According to
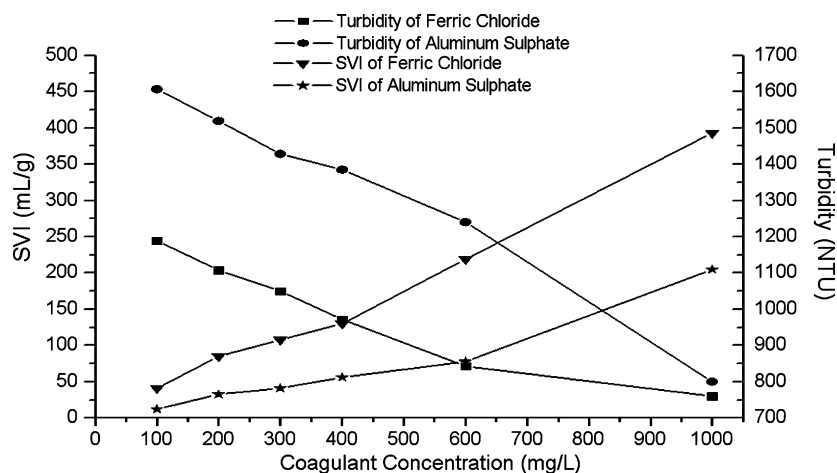

Fig. 2. SVI and turbidity response as a function of coagulant concentration.

Table 1
Results of SVI, turbidity and SOUR under different experimental conditions according to the central composite design matrix.

Exp.	FC (X_1) (mg/L)	PC (X_2) (mg/L)	SVI (mL/g)	Turbidity (NTU)	SOUR (mg O_2/g VSS por h)
1	100.0 (−1)	0.2 (−1)	34.7 ± 3.0	1180.0 ± 5.0	42.1 ± 3.0
2	100.0 (−1)	3.0 (1)	55.5 ± 2.0	1040.0 ± 2.0	45.5 ± 1.5
3	400.0 (1)	0.2 (−1)	208.3 ± 2.0	920.0 ± 3.0	40.7 ± 2.0
4	400.0 (1)	3.0 (1)	236.1 ± 1.8	744.1 ± 4.0	41.9 ± 1.0
5	77.9 (−α)	1.6 (0)	37.8 ± 2.0	1166.4 ± 2.0	44.3 ± 3.0
6	422.1 (α)	1.6 (0)	208.3 ± 3.0	850.0 ± 5.0	48.9 ± 2.0
7	250.0 (0)	0.0 (−α)	120.0 ± 3.0	980.0 ± 7.0	46.6 ± 1.0
8	250.0 (0)	3.2 (α)	166.7 ± 4.0	804.4 ± 6.0	44.8 ± 2.0
9	250.0 (0)	1.6 (0)	110.4 ± 1.0	820.0 ± 3.0	59.2 ± 1.0
10	250.0 (0)	1.6 (0)	100.7 ± 1.5	821.0 ± 3.0	58.3 ± 2.0
11	250.0 (0)	1.6 (0)	104.2 ± 2.0	822.0 ± 4.0	60.3 ± 1.5

FC: ferric chloride concentration; PC: polyelectrolyte concentration; SVI: sludge volume index; SOUR: specific oxygen uptake rate; X_1: codified variable for ferric chloride; X_2: codified variable for polyelectrolyte; pH = 7.0.

the results obtained in the preliminary tests and presented in the article of Amuda and Amoo [1], it was chosen a concentration range for ferric chloride of 77.9–422.1 mg/L in the CCD applied for the optimisation.

Kim et al. [20] reported that sludge with an SVI between 80 and 120 mL/g possessed good settling characteristics. According to the literature [5,13,21,22], an SVI value below 100 mL/g is considered a good settling sludge. Thus, in this study it was adopted as reference values of SVI of 80–120 mL/g in order to optimise the ferric chloride and polyelectrolyte concentrations in relation to the SVI responses, the turbidity and the SOUR.

3.2. Optimisation of ferric chloride and polyelectrolyte concentrations

Table 1 shows the average SVI, turbidity, and SOUR obtained with the C_1 mixed culture.

The values of SVI varied between 34.7 ± 3.0 mL/g (experiment 1) and 236.1 ± 1.8 mL/g (experiment 4). The turbidity ranged from 744.1 ± 4.0 NTU (experiment 4) to 1180.0 ± 5.0 NTU (experiment 1). The SOUR varied from 40.7 mg O_2/g VSS h (experiment 3) to 60.3 mg O_2/g VSS h (experiment 11).

In experiment 1, where the ferric chloride concentration (FC) and polyelectrolyte concentration (PC) were low, lower SVI and higher turbidity were obtained, indicating that lower amounts of coagulant and polyelectrolyte were not sufficient. An SVI of 100 mL/g, a turbidity of 820 NTU and a SOUR of 59 mg O_2/g VSS h were achieved with the addition of 250 mg/L of ferric chloride and 1.6 mg/L of polyelectrolyte. Moreover, the best results were obtained at the experiments localized at the central point (experiments 9–11).

The behaviour of SVI, turbidity and SOUR was evaluated with multiple regression analysis. Variables with a significance levels (p) greater than 10% were considered insignificant and were eliminated from the analysis.

3.2.1. Analysis of SVI results

A correlation coefficient of 98% was obtained after the elimination of the insignificant parameters, indicating that experimental results (Table 1) concurred with model predictions.

Eq. (1) describes the adjusted model after the elimination of insignificant parameters.

$$\text{SVI (mL/g)} = 107.7 + 82.9X_1 + 15.4X_2 + 22.6X_2^2 \tag{1}$$

According to Eq. (1), the SVI was influenced by X_1 and X_2. A response surface plot and contour curves were constructed with SVI data (Fig. 3) to determine the optimal conditions producing the maximum SVI.

According to the contour curves (Fig. 3), the FC and PC that produced an SVI near of 100 mL/g were 160–250 and 0–3.2 mg/L, respectively.

3.2.2. Analysis of turbidity results

After the elimination of insignificant parameters, a correlation coefficient of 99% was obtained. These results indicated that experimental and predicted values were in agreement. Eq. (2) represents the adjusted model, after elimination of insignificant parameters.

$$\text{Turbidity (NTU)} = 829.7 - 138.5X_1 + 121.0X_1^2 - 78.0X_2 + 32.9X_2^2 \tag{2}$$

Eq. (2) indicates that lower concentrations of FC and PC provided the highest values of turbidity. The effects of FC and PC concentration on turbidity are shown in Fig. 4. Analysis of the contour curves indicated that lowers values of turbidity were obtained with a FC and PC of 280–380 and 2.4–3.2 mg/L, respectively.

3.2.3. Analysis of SOUR results

After the elimination of insignificant parameters, a correlation coefficient of 93% was obtained. These results indicated that experimental and predicted values were in agreement. Eq. (3) represents the adjusted model, after elimination of insignificant parameters.

$$\text{SOUR (mg } O_2\text{/g VSS h)} = 58.7 - 8.1X_1^2 - 8.8X_2^2 \tag{3}$$

Analysis of the contour curves (Fig. 5) indicated that an optimal SOUR was achieved with a FC and PC of 190–300 and 1.0–2.0 mg/L.

A maximum in the contour curves (250 mg/L of ferric chloride and 1.6 mg/L of polyelectrolyte) was obtained after canonical analysis of the response surface (Software Maple 9.5). After substituting the maximum values of PC and FC into Eq. (3), an optimised SOUR of 58.7 mg O_2/g VSS h was obtained.

3.3. Optimisation of multiple responses

Using the algorithm in Maple 9.5, Eqs. (1) and (2), were optimised with the recommended SVI (80–120 mL/g) to obtain a ferric chloride and polyelectrolyte concentration of 223 and 1.92 mg/L, respectively. At these concentrations, the experimental results included a turbidity of 844.83 ± 10 NTU, an SVI of 98.0 ± 2 mL/g, and a SOUR of 55.8 ± 5 mg O_2/g VSS h.

Because polyacrylamide is expensive a lower concentration of polyelectrolyte within the optimised region of the contour plot was evaluated by monitoring the SVI, turbidity, and SOUR under economical conditions.

Thus, 250 mg/L of ferric chloride (a higher concentration of FC, as previously discussed) and 1.4 mg/L of polyelectrolyte (a lower
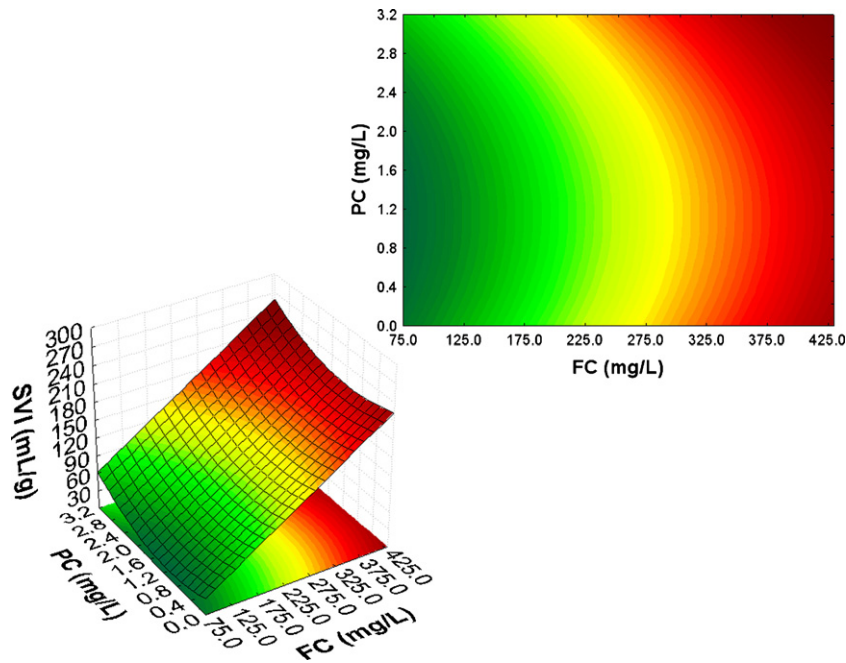
Fig. 3. Response surface and contour curve for SVI response as a function of FC: ferric chloride concentration (X_1) and PC: polyelectrolyte concentration (X_2).

concentration of PC) were analysed. At these concentrations, the experimental responses included an SVI of 105 ± 3 mL/g, a turbidity of 838 ± 4 NTU, and a SOUR of 57.77 ± 3 mg O_2/g VSS h. The experimental results at the central point (experiments 9–11), the optimised conditions obtained from Eqs. (1) and (2) (223 mg/L of FC and 1.92 mg/L of PC) and the selected conditions (250 mg/L of FC and 1.4 mg/L of PC) indicated that optimised conditions were reproducible.

Optimised values of SVI, turbidity and SOUR were obtained under different conditions. Fig. 6 displays the contour lines for the region of the contour plot where the SVI was approximately 100 mL/g, the turbidity was between 810 and 890 NTU, and the

SOUR was between 52 and 55 mg O_2/g VSS h. As shown in Fig. 6, the values obtained for SOUR did not vary significantly; thus, the region was assessed in detail to obtain the optimal SVI and turbidity, as a function of ferric chloride and polyacrylamide concentration.

Various proportions of ferric chloride and polyelectrolyte were evaluated to obtain satisfactory values of SVI, turbidity and SOUR under economically favourable conditions, which are dependent on the cost of coagulant and polyelectrolyte. These results indicated that a joint evaluation of FC and PC produced experimental conditions that lead to reduced process costs, without compromising performance.
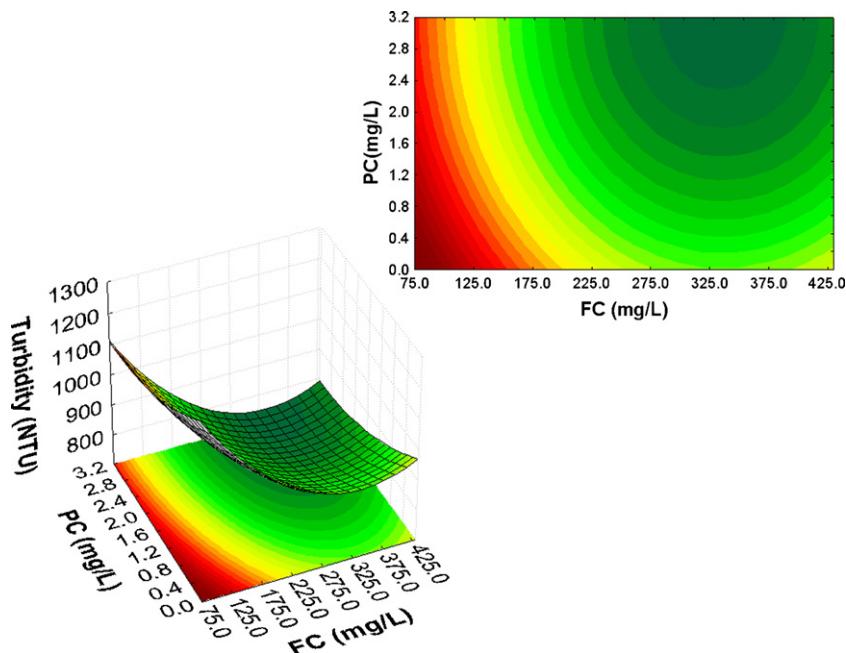

Fig. 4. Response surface and contour curve for turbidity response as a function of FC: ferric chloride concentration (X_1) and PC: polyelectrolyte concentration (X_2).
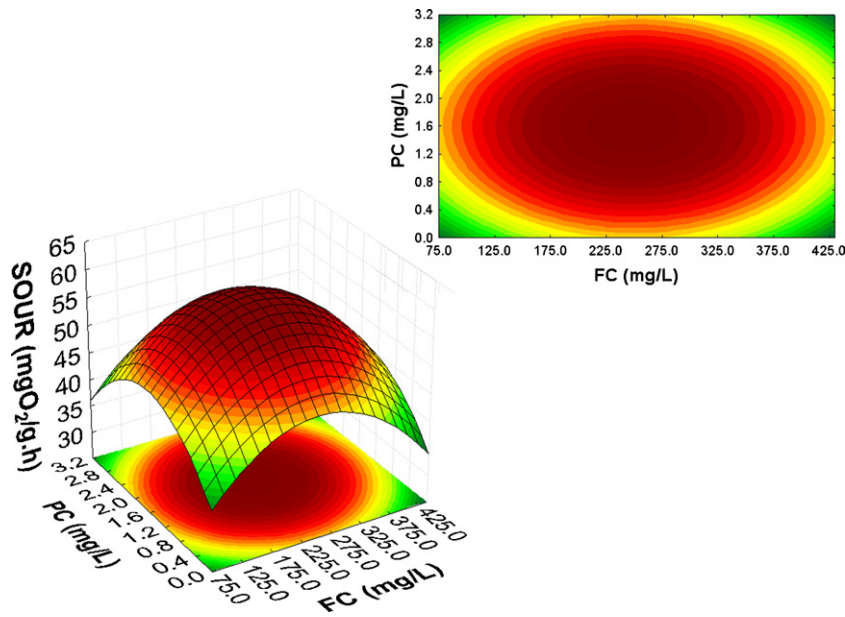
Fig. 5. Response surface and contour curve for SOUR response as a function of FC: ferric chloride concentration (X_1) and PC: polyelectrolyte concentration (X_2).
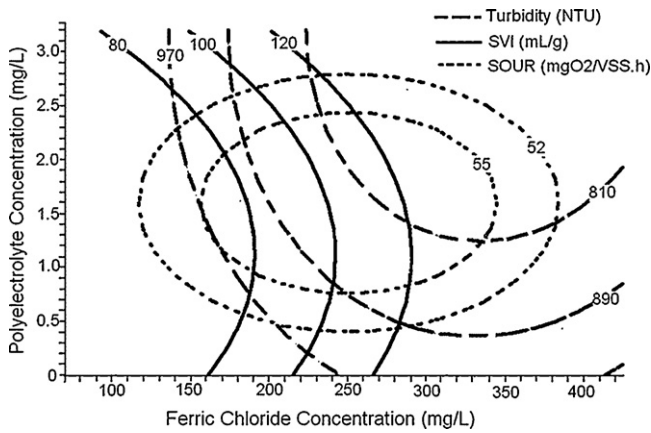

Fig. 6. Contour curve for SVI, turbidity and SOUR responses as a function of ferric chloride and polyelectrolyte concentrations.

3.4. Results of the SBR

To investigate microbial activity, the concentration of ferric chloride (250 mg/L) and polyelectrolyte (1.4 mg/L) that produced optimal results in the CCD were used in five cycles of a SBR.

A ferric chloride concentration of 250 and 1.4 mg/L of polyacrylamide was used in SBR biodegradation. The experiments were monitored over five cycles of operation. The results of TPH removal after each cycle are shown in Fig. 7. After each 3-day process with the addition of coagulant and polyelectrolyte, an increase in the removal of TPH was observed. This suggests that the joint use of ferric chloride and polyacrylamide in optimals concentrations promoted greater settling of microorganisms, which may have caused an increase in TPH removal.

In studies on the biodegradation of effluent from fuel distribution terminals, Vieira et al. [14] observed difficulties in the settling of C_1 mixed cultures. In the aforementioned study, a TPH removal of 76% was achieved after 3 days of operation. The results of this investigations and previous studies indicated that the presence of ferric chloride and polyelectrolytes in the effluent promote an increase in TPH removal. In this study, the results obtained in five cycles of SBR suggested that the increase in TPH removal may
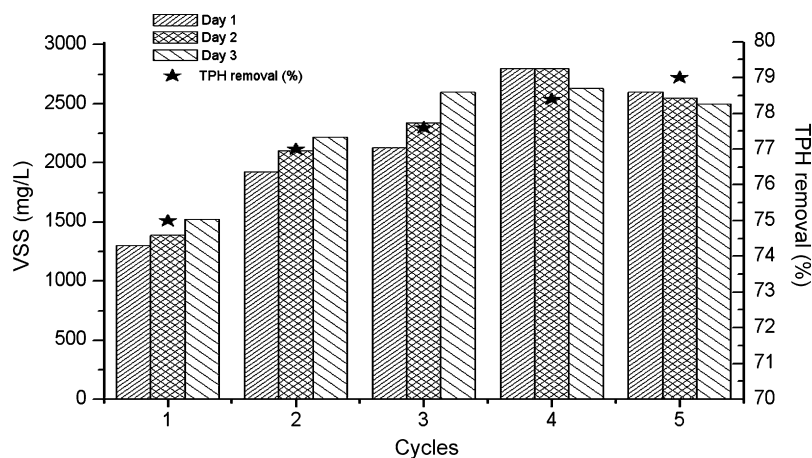

Fig. 7. TPH removal and VSS concentration as a function of cycles in the SBR.

Table 2
Values of the SOUR at the end of the each cycle in the SBR.

	Effluent	1° cycle	2° cycle	3° cycle	4° cycle
SOUR (mg O_2/g VSS por h)	58.9 ± 1.0	56.4 ± 1.1	54.6 ± 0.9	51.9 ± 0.8	48.1 ± 1.0

be related to an increase in settling during each cycle. As shown in Fig. 7, the initial concentration of VSS in the reactor (without addition of ferric chloride and polyelectrolyte) was 1500 mg/L, which increased to 2500 mg/L after five cycles in the presence of coagulant and polyelectrolyte. The observed increase in VSS and TPH removal indicates that microorganisms present in reactor adapt and multiply in each cycle.

According to Lee et al. [23], the chemical oxygen demand (COD) increases slightly with an increase in sludge retention time (SRT). COD values of 90.7%, 91.1%, 91.9%, 92.1% and 92.2% were obtained from cycles of 1 to 5 of SBR, respectively. This may reflect an increase in the concentration of microorganisms that are available to adsorb and oxidise organic compounds.

Table 2 shows the values of SOUR in SBR. In each cycle, a reduction in the SOUR was observed. Strong agitation was necessary to promote rapid homogenisation of diesel oil and gasoline in effluent, resulting in the destruction of flakes during agitation. However, the reduction of SOUR did not compromise the removal of TPH or the growth of biomass, as shown in Fig. 7.

The increase in hydrocarbon removal and cell growth in each cycle may be related to changes in microbial metabolism, while the observed reduction in SOUR may be related an increase in viscosity after each cycle, which may have impeded the transfer of O_2.

4. Conclusion

Ferric chloride was a superior coagulant for the sedimentation of C_1 culture than aluminium sulphate. By applying a CCD with varying concentrations of ferric chloride and polyelectrolyte, optimal conditions were determined and process costs were reduced. The selected operating condition included 250 mg/L of ferric chloride and 1.4 mg/L of polyelectrolyte.

The results under optimised conditions were included a SOUR of 57.77 ± 3 mg O_2/g VSS h, a SVI of 105 ± 3 mL/g and a turbidity of 838 ± 4 NTU. The addition of ferric chloride and polyelectrolyte to each cycle, in a sequencing batch reactor, did not hinder the removal of TPH (increased from 75 ± 1.0% to 79 ± 0.5%), but caused an increase in the concentration of VSS (1300–2500 mg/L) and a reduction in the SOUR (58.9 ± 1.0–48.1 ± 1.0 mg O_2/g VSS h). The reduction in SOUR may be related to an increase in medium viscosity. The optimisation of FC and PC produced experimental conditions that lead to reduced process costs, without compromising performance.

Acknowledgements

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