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Optimization method for the treatment of Tehran petroleum refinery wastewater using activated sludge contact stabilization process

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ABSTRACT

This paper describes the results of side by side large scale testing of contact stabilization and extended aeration activated sludge processes on the petroleum refinery wastewater at Tehran (Iran) oil refinery by using continuous large scale experiments during the three month investigation period. The process optimization for performance evaluation was studied. Sampling results indicated that the contact stabilization process shows more effective performance than extended aeration process in removing chemical oxygen demand (COD) and biological oxygen demand (BOD), and also the mixed liquor volatile suspended solids were less in contact stabilization as compared to those in the extended aeration process. The excess sludge production was affected by the amount of sludge loading rate and aeration. The applied aeration to the mixed liquor and the sludge recycle rate were found to be critical parameters in the successful optimization of the contact stabilization process. The appropriate optimized operational conditions that allowed obtaining the best performance with COD removal efficiency of 96 and 95% were food to micro-organism ratio of 0.38 with sludge recycle rate of 110 and 77% sludge recycling in aeration of 0.27 L of air per liter of wastewater per minute, respectively. All in all, contact stabilization activated sludge process with high degree of assurance and less operation cost can be suggested for the treatment of wastewater originated from petroleum refineries.

Keywords: Petroleum refinery wastewater; Optimization; Contact stabilization; Sludge recycle rate; Aeration; F/M ratio

1. Introduction

Petrochemical industries and petroleum refineries generate large amounts of priority pollutants release a complex set of oxygen-demanding materials into the natural environment. The major pollutants found in these industries are petroleum hydrocarbons, specifically aliphatic hydrocarbons, arising from storage of crude oil, spills, wash downs, and vessel clean-outs from the processing operation which are mainly different from other industrial wastewaters [1]. Treatment of these complex wastes is a big problem and has become a challenging objective for protecting the environment. Reuse of treated wastewater for irrigation objectives is another goal. Usually refineries are

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among the highly water consuming centers which are often located near the large cities and the discharge of untreated wastewater from these centers into the surrounding areas results in polluting the surface and ground waters, in addition to the environmental problems pertaining to air and soil pollutions.

Since the treatment process is costly, it should be optimized in terms of effluent quality and the overall cost; also the treated water should be used in cooling towers of the refinery, or applied for the irrigation of green areas or crops. The treated water of Tehran petroleum refinery is used for recharging the ground water aquifer of the refinery's site. Therefore the effluent from the treatment plant should meet the WHO and EPA standards for reuse.

Biological methods for the treatment of oil refinery wastewater are mainly based on activated sludge, trickling filters and membrane processes [2,3]. Component elimination by biological oxidation processes is a core process in oil refinery wastewater treatment technology which is the subject of this investigation; by experimentally optimizing the process parameters which are mainly oxygen concentration and food to micro-organism (*F*/*M*) ratio in the contact stabilization activated sludge system.

Most of the literatures on the treatment of petroleum refinery wastewater were performed through a series of on-site treatment technologies, such as, American Petroleum Institute Separators [4,5], Tilted Plate Interceptor Separators, and Dissolved Air Floatation units [6–9]. These operations are followed by biological treatment in a suspended growth process. However, these processes are typically associated with numerous operational problems, which include: poor settleability of the sludge due to low *F*/*M* ratio; production of extra-cellular polymers consisting of lipids, proteins, and carbohydrates that adversely affect sludge settling; biological inhibition due to toxic compounds, which necessitates very long sludge retention time (SRT); long period of acclimation or start-up, and production of large amount of biological sludge [10–13].

2. Activated sludge process

Activated sludge process is an aerobic biological treatment with continuous flows that provide suspended growth of micro-organisms and oxidize the organic matters. Carbon dioxide, water, and new cells (sludge) are the end products of this process. Suspended aerobic micro-organisms mix with the raw wastewater of the refinery and a series of reaction accomplish in the aeration tank which results in the analysis of organic carbon and oxidation of ammonium. The excess sludge produced by this process. After a certain period of time in an aeration tank, wastewater enters into the secondary sedimentation tank where micro-organisms and the mixed liquor suspended solids (MLSS) separate from the treated wastewater. The main part of mixed liquor volatile suspended solids (MLVSS) (sludge) returns to aeration tank through return sludge line [14,15]. The rest of the sludge is transferred into the sludge establishment's basin. Adequate nitrogen and phosphorus should be provided to increase microbial growth in the oil refinery wastewater treatment plant.

The optimum process of activated sludge is obtained by changing some factors. These major factors include the amount of organic loading, sludge residence time or mean cell residence time (MCRT), hydraulic detention time, dissolved oxygen (DO) concentration, and aeration method (surface or diffused air aeration).

Organic loading is obtained through the *F*/*M* or daily applied biological oxygen demand (BOD_5) in kilogram. The *F*/*M* can create conditions that favor the predominance of filamentous organisms affecting the settling properties of the sludge, causing brown foam in the aeration tank and deterioration in effluent quality [16,17]. Sludge age is effective on the degree of treatment and excess sludge production. Hydraulic detention time is obtained from the size of aeration tank and evaluated according to *F*/*M*, SRT, and MLSS values. SRT can affect the floc structure and the settling properties of the sludge [18–20]. The amount of required oxygen is obtained from the total BOD, and the percentage of internal breathing of micro-organism [21]. Low levels of DO can affect the sludge settling properties and the metabolic activity of micro-organisms, generating an incomplete removal of substrate, which is reflected in the poor effluent quality [22].

The treatment method and design parameters could be changed depending on the type of wastewater to be treated. Basically one or more major parameters are required to be changed, to accomplish optimization. The change in organic loading, location and type of aeration, the percentage of returned sludge (RS), and form of aeration tank are some examples [23].

In contact stabilization activated sludge process, raw wastewater enters into the contact tank and then mixes with bacteria. In the contact tank, soluble and insoluble organic materials absorbed and adsorbed by the bacteria and in the presence of oxygen, oxidation of organic matters, and cell growth take place prior to enter into the secondary sedimentation tank. The settled sludge from secondary sedimentation tank enters into the stabilization tank where digestion of the rest of organic material takes place. The stabilized sludge among with the fresh bacteria overflow into the contact tank and mixes with the raw incoming wastewater. Since these bacteria have consumed the restored food in their cell, they rapidly absorb the organic matters of untreated influent. Therefore the detention time decreases and the treatment efficiency increases.

In comparison to contact stabilization, the extended aeration activated sludge process requires more hydraulic residence time and cell residence time for the bacteria in order to digest the organic matters.

3. Materials and methods

3.1. Pilot plant description

The process studied was a biological treatment pilot plant which consists of a continuous flow reactor. The treatment involves three distinct zones: contact, stabilization, and settling tank as shown in Fig. 1. The contact-stabilization pilot plant reactor is constructed from Plexiglas rectangular shape and has an effective volume of 450 L for the first section (contact), 200 L for the second section (stabilization), and 200 L for the last section (settling basin) as depicted schematically by Fig. 1. Air was supplied by the air compressor through porous plate diffuser, lying on the bottom of the reactors. The air-flow in the reactor ensures a mixing intensity that simulates the mixing characteristic in activated sludge process. By considering the small scale of pilot plant and in order to control and reduce the intense shocks of H_2S and oil molecules in the incoming wastewater, an equalization basin with an effective volume of 200 L with a unit of diffuser and air flotation was installed before the pilot plant.

The reactor was maintained at a constant hydraulic retention time of 1.5 h for contact tank and 4.5 h for stabilization tank, also the retention time for settling tank maintained at 2 h.

The sludge was removed continuously by the installing a turbidimeter that controls the sludge level in the sedimentation tank. The sludge was recycled from the settler by means of mono pumps at recirculation ratio 0.5–1.5 and then was returned into stabilization tank to be treated for 4.5 h and overflows to contact tank where incoming raw wastewater mixed with stabilized mixture.

Different recirculation ratios results in different *F*/*M* ratio to operate the contact stabilization method. The minimum time of one MCRT between subsequent test series was needed to ensure a steady state condition.

In parallel, the activated sludge extended aeration pilot plant reactor is constructed with same size and material as those of contact stabilization pilot plant with 450 L volume of aeration tank and 200 L volume of settling tank as depicts schematically by Fig. 2. In order to permit the occurrence of short-circuiting in aeration basin, a wooden hinder was installed in front of aeration inlet. The operational conditions of extended aeration pilot plant were same as those applied to contact stabilization pilot plant system.

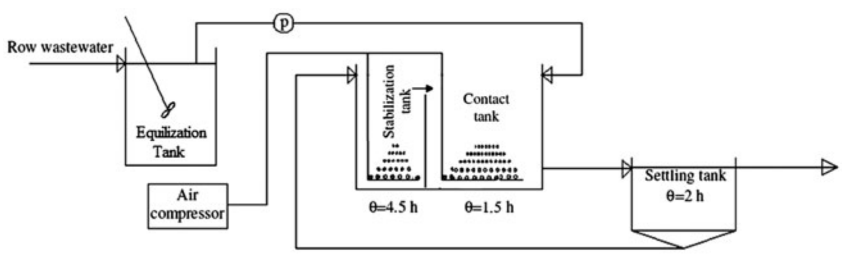

Fig. 1. The experimental setup of contact stabilization pilot plant.

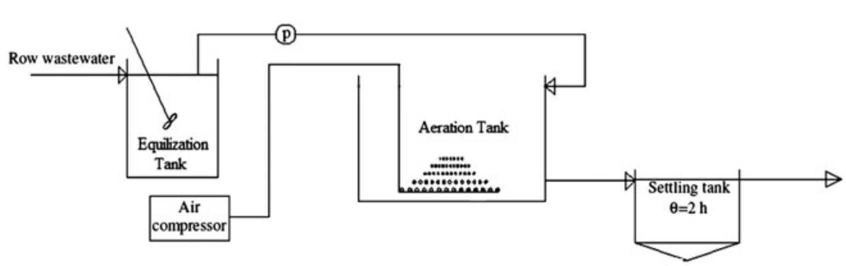

Fig. 2. The experimental setup of extended aeration pilot plant.

Table 1
Treatment plant wastewater influent quality parameters

Parameter	COD	BOD_5	TSS	Oil	NH_3–N	PO_4^{-3}–P	H_2S	pH
Maximum	5,060	360	580	0.8	48	10	40	9
Minimum	340	90	30	*	10	0.6	0.7	7.4
Mean	641	198	198	0.22	26	1.55	5.5	8.4

Note: *Less than a typical detection limit.

Table 2
Effluent parameters from contact stabilization pilot plant

Parameter	COD	TSS	Oil	NH_3–N	PO_4^{-3}–P	H_2S	pH
Maximum			*	20	1.4	1	7.8
Minimum	22	8	*	8	*	*	7.2
Mean			*	15.6	0.4	0.3	7.6

Note: *Less than a typical detection limit.

The reactors were inoculated with activated sludge taken from the Tehran petroleum refinery's wastewater treatment plant.

3.2. Monitoring, sampling, and testing

Inline and offline variables of the process were monitored in all experiments. Inline variables included flow rates (influent, internal recycle, and external recycle), pH, temperature, DO, MLSS, MLVSS, PO_4^{3-}–P, NH_3–N, total suspended solids (TSS), chemical oxygen demand (COD), BOD, total dissolved solids, H_2S, and free oil. Sampling was accomplished from inline and offline in-flow wastewater, in certain hours and during the aeration and settling times. COD, BOD, TSS, and VSS and other parameters in the reactors have been measured in the Environmental Laboratory according to the Standard Methods for Examination of water and wastewater [24].The results of sampling and measurement of the parameters for input and output wastewater are presented in Tables 1 and 2. In parallel, another pilot plant based on the activated sludge extended aeration process was established and the treatment efficiency of that with regard to the same input wastewater was studied.

4. Results and discussion

Successful results were obtained in the optimization of oil refinery wastewater treatment method. Table 3 shows the comparison between the effluent quality parameters from contact stabilization and extended aeration activated sludge processes in two parallel pilot plants studied. In general, the quality parameters of the effluent from contact stabilization process pilot plant are better than those of the extended aeration process. The relationship between BOD and COD of influent wastewater at the beginning of the experiment was developed. Fig. 3 shows that BOD and COD are highly correlated with correlation coefficient of 0.993 that means there are not much chemical inorganic compound present in the oil refinery wastewater. In addition, the inhibitors were not found to depress the bacterial growth in the process of treatment.

Table 3
Treatment efficiency comparison between contact stabilization and extended aeration activated sludge processes in pilot plant studies

Parameter	Influent	Effluent	
		Extended aeration	Contact stabilization
pH	7.4	7.2	7.2
NH_3–N (mg/L)	48	8	6
PO_4^{-3}–P (mg/L)	3	*	*
H_2S (mg/L)	40	2	1
BOD_5 (mg/L)	360	20	15
COD (mg/L)	1,060	70	22
TSS (mg/L)	580	14	8

Note: *Less than a typical detection limit.

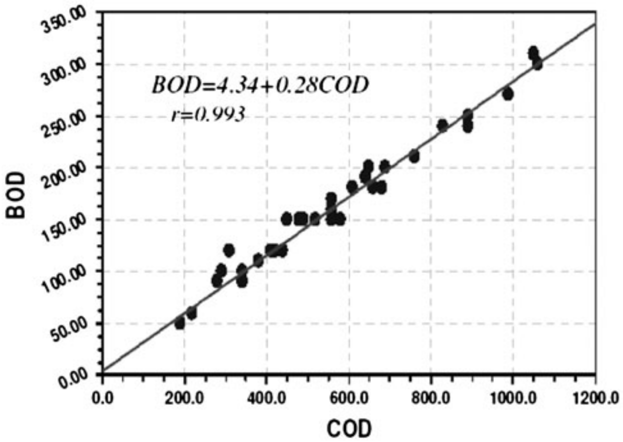

Fig. 3. The relationship between influent BOD and COD concentrations.

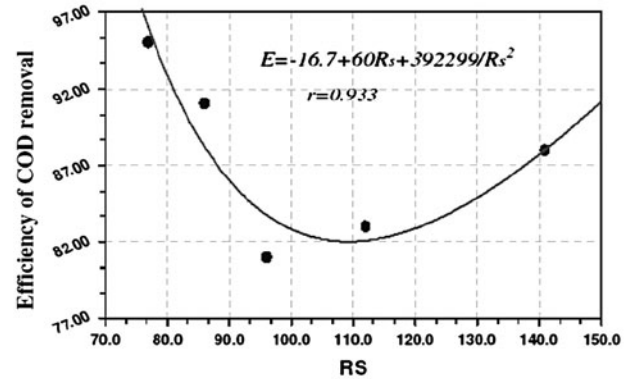

Fig. 4. Treatment efficiency of contact stabilization system in different RS percentages with aeration of 0.27 L of applied air per liter of wastewater.

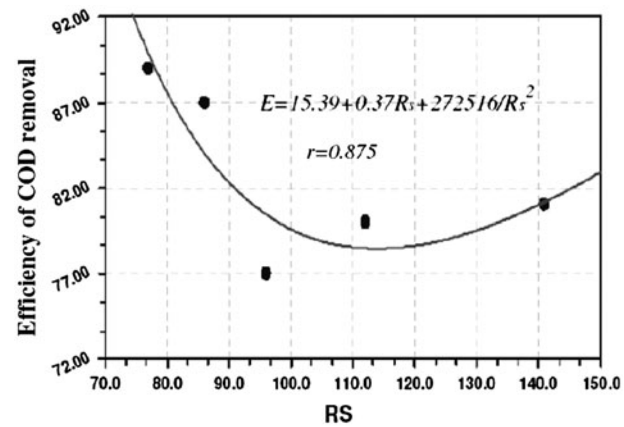

Fig. 5. Treatment efficiency of contact stabilization system in different RS percentages with aeration of 0.16 L of applied air per liter of wastewater.

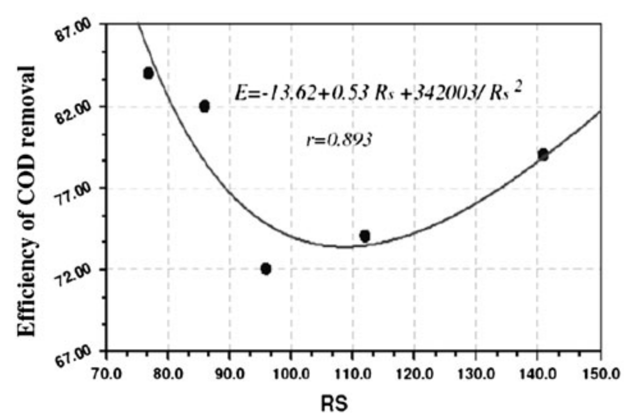

Fig. 6. Treatment efficiency of contact stabilization system in different RS percentages with aeration of 0.11 L of applied air per liter of wastewater.

During the operation of contact stabilization pilot plant, different ranges of applied air to the influent wastewater in the contact tank reactor at a constant temperature (20 + 0.1℃) were examined and five different percentages of RS were applied to the stabilization tank reactor. Before starting the measurement of the mentioned quality parameters at each aeration period and return sludge percentages, the pilot plant was operated at a study state for at least one MCRT. Also, the aeration period of time in the stabilization tank reactor in different percentages of RS changed in a manner that when RS considered equal to 77%, the detention time for aeration was equal to 3.4 h. Furthermore, when RS was 84% the detention time for aeration was 3.1 h, in RS of 96%, it was 2.8 h, in the RS of 112% it was 2.4 h and finally, in RS of 141% the detention time for aeration was 1.9 h. Figs. 4–6 show the treatment efficiency of contact stabilization reactor for different RS percentages and aeration periods.

Fig. 3 also indicates that application of 0.27 L of air per each liter of wastewater per minute results in 110% RS with 82% treatment efficiency with regard to COD removal.

Similarly, Figs. 5 and 6 show that as the application of air decreases to 0.16 L of air per liter of wastewater per minute for 110% RS, the treatment efficiency decreases to 78.5%.

Therefore, from Figs. 4–6 it can be inferred that the optimum aeration for the most COD removal is 0.27 L of air per liter of wastewater per minute with 80% RS. The increase of the oxygen concentration in the bulk liquid mixture leads to a deep diffusion of oxygen which subsequently causes an enlargement of the aerobic volume inside the floc. As a result, the hydrolyzed micro-organisms in the floc degraded. The influence of different sludge loading rates by changing the RS percentage on the efficiency of effluent COD were studied by many researchers, but they did not consider the effect of different DO concentration in the aeration tank.

The effect DO on the efficiency of COD removal was also studied. As it shown in Fig. 7, for DO concentration of about 4 mg/L the COD removal efficiency was achieved about 45%, but as the DO increases the COD removal percentage decreases. This phenomenon was due to the effect of different sludge loading rates on excess sludge production and followed by the influence of high DO concentration on the rising of sludge in the secondary settling tank, known as sludge bulking. The reduction of excess sludge production can be achieved by increasing the DO from 2 to 4 mg/L at sludge loading rate of about 0.4 mg BOD_5/mg MLVSS (*F*/*M* = 0.4) and 27% aeration.

The effect of RS percentage on the effluent COD concentration can be described as following:

(a) In aeration of 120 L/min (0.27 L of air for each liter of wastewater per minute), the least output in RS was 109.2% with 82% of deletion, as shown in Fig. 4.
(b) In aeration of 70 L/min (0.16 L of air for each liter of wastewater in each minute), the least output in RS was 113.8% with 78.5% of deletion, as shown in Fig. 5.
(c) In aeration of 50 L/min (0.11 L air for each liter of wastewater in each minute), the least output in RS was 113.7% with 72.5% of deletion, as shown in Fig. 6.

Figs. 8–10 show the effect of various *F*/*M* ratios on the effluent COD concentration in the term of treatment efficiency.

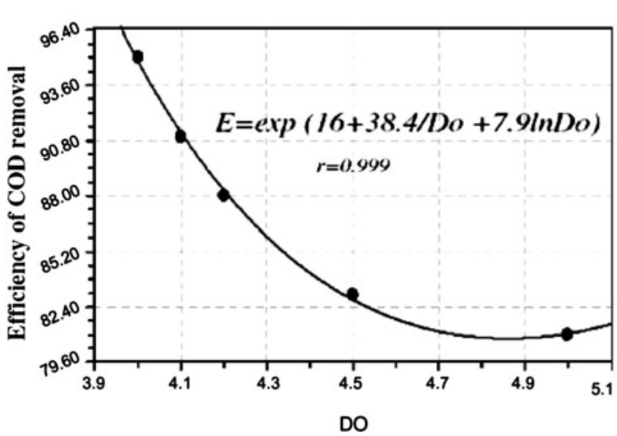

Fig. 7. Effects of different DO concentrations on the COD removal efficiency.

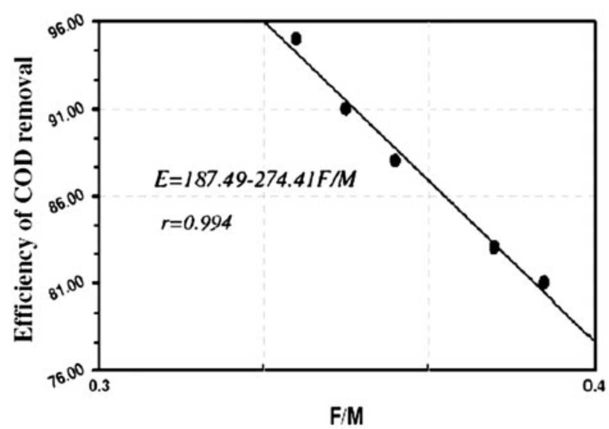

Fig. 8. Effect of different *F*/*M* ratios on COD removal efficiency in aeration of 0.27 L of applied air per liter of wastewater.

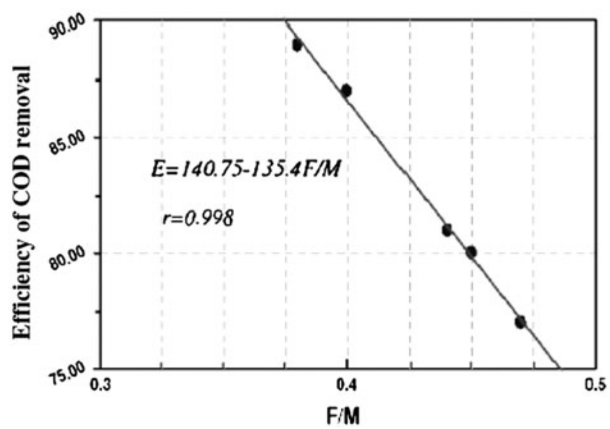

Fig. 9. Effect of different *F*/*M* ratios on COD removal efficiency in aeration of 0.16 L of applied air per liter of wastewater.

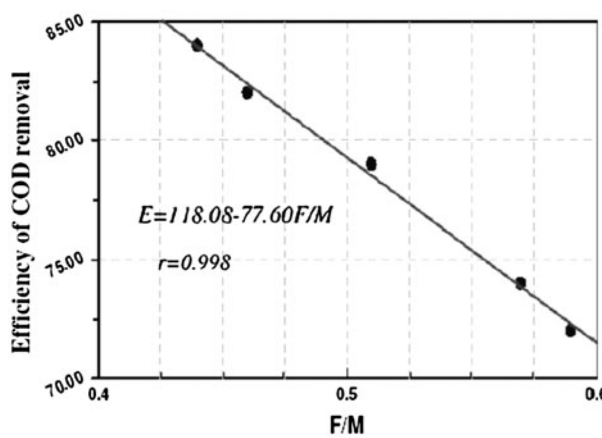

Fig. 10. Effect of different *F*/*M* ratios on COD removal efficiency in aeration of 0.11 L of applied air per liter of wastewater.

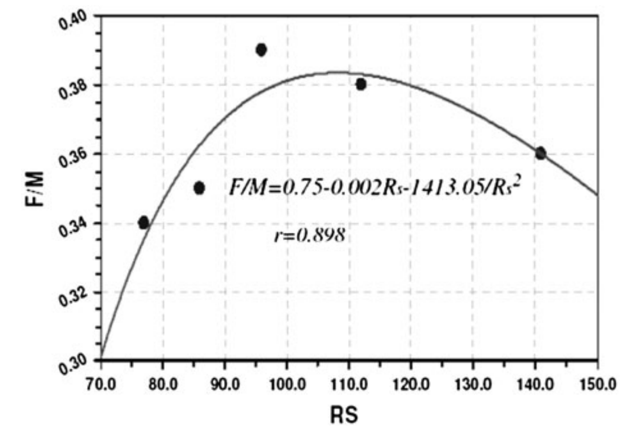

Fig. 11. Effect of Different RS percentages on *F*/*M* Ratio in aeration of 0.27 L of applied air per liter of wastewater.

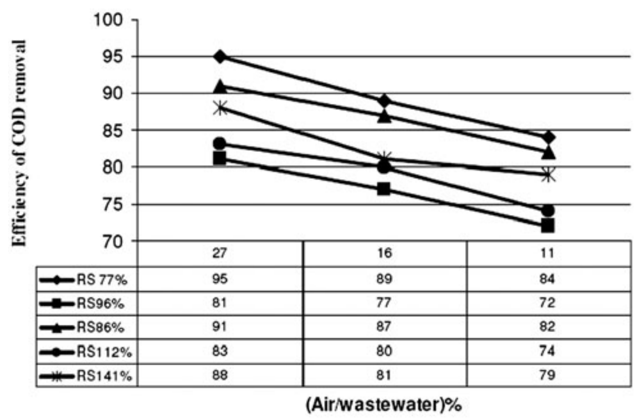

Fig. 12. Effect of different RS percentages and influent air/wastewater ratio on cod removal efficiency.

These figures indicate that as *F*/*M* ratio increases the COD removal efficiency decreases due to excess sludge production. Also, the rising of sludge in the settling tank causes to increase total solids and consequently increase the COD concentration in the effluent. The point of optimization for *F*/*M* ratio is about 0.38 with COD removal efficiency of about 96%.

Moreover, Fig. 10 shows the effect of RS percentage on *F*/*M* ratio. It implied that the optimization point for RS is about 110% for *F*/*M* ratio of 0.38 which corresponds to maximum effluent COD removal efficiency.

The COD removal efficiency in relation to the change of applied air/wastewater ratio was studied for RS of about 96%. The amount of applied air/wastewater ratio of about 0.91 is the optimum point for COD removal as shown in Fig. 11.

The overall results of this study were presented in Fig. 12. As a result of mentioned figure, the maximum COD removal efficiency takes place for aeration of 0.27 L of air per liter of wastewater per minute and RS of 77% which could lead to about 25% reduction in the amount of solids (sludge) in the stabilization tank reactor.

5. Conclusion

The pilot plant experimental results show that the contact stabilization activated sludge process is the optimum method for the treatment of wastewater produced from Tehran petroleum refinery. The present research shows that reduction of effluent BOD and COD concentrations to the standard level can be achieved by raising the concentration of DO in the mixed liquor of aeration tank from 2 to 4 mg/L and RS to 88% with 180 min of aeration time and *F*/*M* ratio of about 0.4. However, we can optimize the COD removal efficiency by raising the percentage of RS up to 138% with aeration time of 120 min. In this condition, the excess sludge production in aeration reactor and probably of the bulking phenomena in secondary settling tank could take place. Besides, according to the fact that the amount of SRT in contact stabilization system is considerably higher than those in extended aeration system and also other activated sludge systems, on one hand and by considering the point that the concentrated activated sludge from the secondary clarifier flows to the stabilization basin, where the majority of produced sludge digest on the other hand, it has been ascertained that competition between floc-forming and filamentous micro-organism is strongly affected by the organic concentration during mixing of influent and return activated sludge. Therefore, most of the MLVSS concentration is utilized and the

Table 4
Optimized functions between parameters

#	Function	*r*
1	$BOD = 4.34 + 0.28\,COD$	0.993
2	$\mathbf{E = -16.7 + 60\,Rs + 392{,}299/Rs^2}$	0.933
3	$E = 15.39 + 0.37Rs + 272{,}516/Rs^2$	0.875
4	$E = -13.62 + 0.53\,Rs + 342{,}003/Rs^2$	0.893
5	$\mathbf{E = 187.49 - 274.41F/M}$	0.994
6	$E = 140.75–135.4F/M$	0.998
7	$E = 118.08 - 77.60F/M$	0.998
8	$\mathbf{F/M = 0.75 - 0.002Rs - 1413.05/Rs^2}$	0.898
9	$F/M = 0.96 - 0.003Rs - 2111.31/Rs^2$	0.897
10	$F/M = 1.63 - 0.006Rs - 4338.15/Rs^2$	0.887
11	$E = \exp\,(16 - 38.4/Do + 7.9\ln Do)$	0.999

amount of excess sludge is considerably digested and decreased. In other words, the MLVSS concentration is directly in proportional to the produced sludge. As it is shown in Table 4, another finding of present research is the ratio of BOD to COD which leads to the equation BOD = 0.28 COD + 4.34 and the relationship between the RS and *F*/*M* ratio ($y = a + bx + c/x^2$) which is complementary to the relationship between the RS and the percentage of pollution detention.

All in all, the treatment efficiency of petroleum refinery wastewater can be substantially increased with available resource while avoiding wasting energy on aeration or recirculation.

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