Effects of Cycle and Fill Period Length on the Performance of a Single Sequencing Batch Reactor in the Treatment of Composite Tannery Wastewater.

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Abstract: A study was conducted to evaluate the feasibility of sequencing batch reactors (SBRs) for the treatment of tannery wastewater and to develop a simple design criteria under local conditions. A single laboratory-scale sequencing batch reactor was used to optimize cycle times and to evaluate the performance at the optimum cycle time. In addition the effect of period of fill time on the chemical oxygen demand (COD) removal efficiencies was studied. The average overall COD removal efficiencies obtained at the 6 and 8-hrs cycle times were 71 % and 85 %, respectively. At the cycle times of 12 and 24-hrs were 89% and 92%, respectively. The total nitrogen (TN) removal efficiencies were also increased from 33% to 49% as the cycle time increased from 6-hrs to 24-hrs. At the optimum cycle time (8-hrs), the reactor showed removal efficiencies of 85% for COD, 38% for total nitrogen, 99.9 % for sulfide and 100% for orthophosphate. The change in fill time was appeared to affect the removal of organic matter. The removal efficiencies decreased from 94 to 90 % and then decreased to 85 % with change of fill time from 180 to 83 and final to 48 and 39 minutes, respectively. In the steady-state condition, the removal efficiencies of COD and TN increased with increasing the cycle time. The removal efficiency of COD increased as the length of feed time increased. A single sequencing batch reactor was not efficient for the removal of nitrogenous compounds from tannery wastewater. This is an issue to be investigated in future work with multiple batch reactors in sequence.
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1. Introduction

Leather tannery effluents are a source of severe environmental impacts. Leather tanning is almost wholly a wet process from which a large volume of liquid waste is continuously generated. The wastewater generates from two main processes: beamhouse and tanning. In the beamhouse process highly loaded effluents are generated, containing organic matter, suspended solids (as a result of unhairing and skinning), sulphides (used in the unhairing) and chlorides (from the salt present in the hides). At this stage bactericides (to prevent degradation of the hide) and surfactants (to improve soaking) are also used. In the tanning process the main pollutant is chromium, while other compounds may be used in the retanning process, such as tannins (Bosnic *et al.,* 2000; Song *et al*., 2004). Due to the variety of chemicals added at different stages of processing of hides and skins, the wastewater has complex characteristics (UNIDO, 2000). In Ethiopia, there are more than 20 tanning industries, concentrated mainly in the southern central part of the country. The operation of tanneries in Ethiopia is causing severe environmental pollution due to the disposal of untreated effluent on land and in water bodies. The effluents, which are discharged without any pretreatment, contaminate surface as well as sub-surface water and soils (Khan *et al*., 1999). There is urgent need for the treatment of tannery effluent prior to their disposal. The continuous biological wastewater treatment systems in use for tannery wastewater are either inadequate or less cost (Ketchum, 1997; Ganesh *et al*., 2005). SBR technology has proved to be an effective system and a viable alternative to continuous flow systems when nutrient removal, in addition to carbon removal, is important (Farabegoli *et al.,* 2007; Ganesh *et al*., 2005; Lefebvre *et al*., 2005). Cyclic concentration gradients, to which biomass is exposed in a SBR reactor , permit selection and enrichment of particular microbial species more capable of carrying out biological processes in the presence of inhibiting substances and high load feed (Artan *et al*., 1996; Herzbrun, 1985). A Sequencing Batch Reactor (SBR) presents the following advantages: high degree of process flexibility in terms of cycle time and sequence; ability to incorporate aerobic, anoxic phases in a single reactor (if desired); sequencing batch closely resembles plug flow operation during the fill cycle; near ideal quiescent settling conditions and no separate clarifiers required (Goltara,2004). The purpose of this study was to investigate the suitability of sequencing batch reactor for tannery wastewater treatment.

2. Materials and Methods

2.1. Experimental set-up

Laboratory-scale sequencing batch reactor with a working volume of 5L was used as shown in Fig. 1. It was complemented by an influent feeding tank (1), vertical stirrer (4) with stainless steel paddles and two peristaltic pumps (2, 6), one for feeding, the other for drawing off effluent and excess sludge. The reactor consisted of two exits: one for sludge withdrawal and the other for cleaning and emptying the reactor. Sludge was withdrawn directly from mixed liquor at the end of the aerobic phase. Influent feed and purified effluent decantation were performed from the top, by means of peristaltic pumps. Aeration was provided at the base of the reactor. Seeding sludge obtained from Ethiopian Tannery Wastewater Treatment Plant was acclimatized to prior to the start of the experiments.

The SBR was fed with a composite tannery effluents collecting from Modjo and Kolba tannery industries found in Modjo town (Ethiopia). The reactor was run at constant temperature of 20^0C which was controlled by water bath.

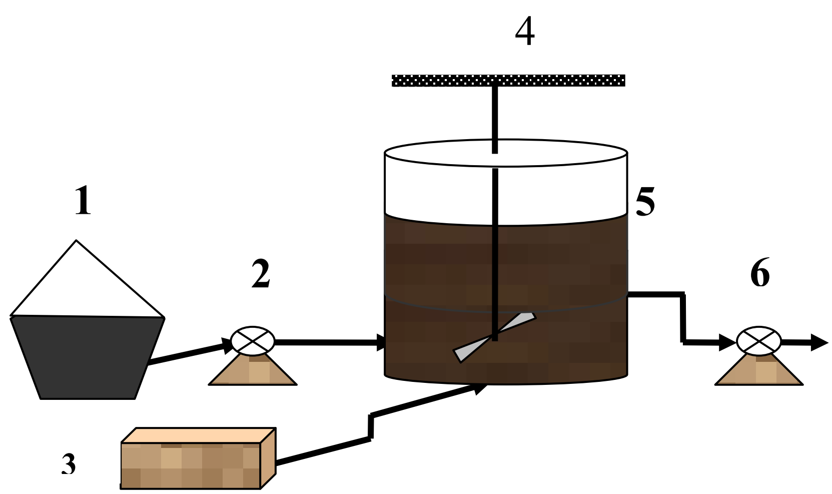


Figure 1. Schematic diagram of Sequencing Batch Reactor.

2.2. Operation of SBR system

The study was conducted over two different operational periods after the start up of the experiments: period 1, in order to optimize the cycle length, the SBR was operated with different aeration time to evaluate the effects on the removal efficiencies of COD and TN from the influents. The operating phases of the cycle were; 2-hrs for anoxic, 3-hrs, 5-hrs, 9-hrs and 21-hrs aeration, 1-hr for settling and discharge. In period 2, the reactor was operated for 8-hrs cycle to evaluate the reactor performance for the removal of COD, Sulfide, nitrogenous compounds, phosphorous and effects of feed times on COD removal (where, 7-hrs was devoted to the process phase and the remaining 1-hr for settle, draw and idle phase (T_S+T_D +T_I = 1-hr). The anoxic phase was 2-hrs and remaining 5-hrs was used in the aerobic phase. In both periods, the system was adjusted to a total sludge age of 15 days. A fill volume (V_F) of 1.25L was selected, leaving 3.75L for stationary volume (V_O) where $Vo/V_F = 3$.

2.3. Chemical Analysis

Chemical oxygen demand (COD), Total nitrogen (TN), Ammonium-Nitrogen (NH_4^+-N), Sulphides (S^{2-}), total phosphorous (TP) and orthophosphate (PO_4^{3-}) were measured colorimetrically using spectrophotometer (DR/2010 HACH, Loveland, USA) according to HACH instructions. Total chromium was also measured according to the methods described in standard methods (APHA, 1998).

3. Results and Discussion

3.1. Start-up and steady state performance

The laboratory-scale sequencing batch reactor was operated for a period of around 3 months to obtain steady state performance. Reactor performance was also assessed by evaluating COD and nitrogen removal efficiencies. As shown in the Fig.2, the system showed low removal efficiencies in the 8, 17, and 27th days' of acclimatization time. At this time, COD level was very high in the treated effluent, leading to average removal efficiencies as low as 50 %.

However,after 38th day of acclimatization time, however, there was drastic reduction of COD in the effluent. During this phase of operation, COD removal efficiency of the reactor was progressively increased from 39% removal efficiency to at the 8th day 85% removal efficiency at the 38th day. After 38th day, there was a marked increase in COD removal and it steadily increased (92-94%) and remained more or less constant thereafter (Fig.2). It took 38 days after inoculation with acclimatized wastewater to reach stabilized steady state performance. Rapid start up in this case may be attributed to the ability of inoculum and the operating condition adopted. Start up of sequencing batch reactor can be satisfactorily achieved in a very short time if adequate inoculums are available (Rittmann and Whitemen, 1994).

The effluent TN concentration was high throughout the experiment days, although there was significant reduction after the 56 day of acclimatization time. The removal rate progressively increased from 8% in 8th day removal to 49 % in 57th day removal. There after the removal rate was constant.

3.2. Optimization of cycle length

Following the steady state operation of the system, the SBR was operated with different aeration times in order to determine optimum total cycle time. The SBR operation was designed for 6, 8, 12 and 24 hrs total cycle time with the aeration times of 3, 5, 9 and 22-hrs, respectively. The average organic loading rate (OLR) was 2.3kg COD m^{-3} d^{-1} and total nitrogen loading rate was 0.239 kg TN m^{-3} d^{-1}.

The concentration of COD decreases sharply with an increase in the cycle time from 6-hrs to 24-hrs (Table 1). In the 6-hrs cycle time, the concentration of COD in the final effluent was 892 ± 177.2 mg/l. In this cycle, the removal efficiency obtained was 71 % and mass of COD removed per day, with four different feeding per 24 hours, was the 8705.2mg/L (Fig. 3). In the 8-hrs cycle, the COD concentration was further reduced to 458 ± 62.8mg/l and the removal efficiency and COD mass removal rate per day (with three different feeding per 24 hours) were 85% and 7830.9mg/l, respectively (Fig. 3). When cycle time increased to 12-hrs, the concentration of COD was reduced to 331.7 ± 24.6 mg/l and the observe removal efficiencies and COD mass removal rate (for two different feeding) were 89% and 5473.2mg/L, respectively (Fig. 3). However, the COD concentration decreased to 234.2 ± 17.8 mg/l as the cycle time increased to 24-hrs. The removal efficiency and mass removal rate (for single feeding per 24-hrs) were of 92% and 2834.1mg/l, respectively (Fig. 3).

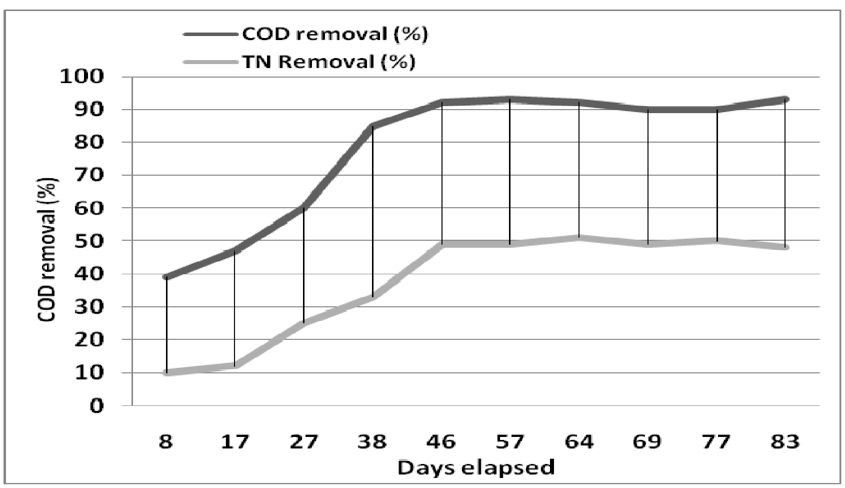
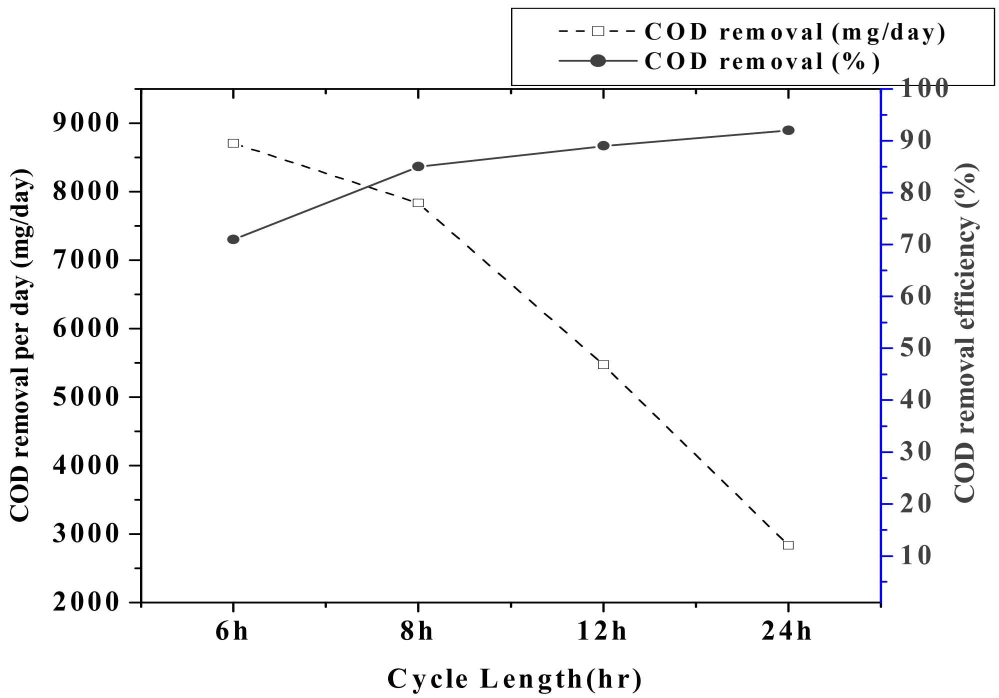


Figure 2. COD and TN removal efficiencies of the SBR

Table 1. Influence of cycle length on COD and TN removal efficiencies

Parameter	Influent	Effluent in the different cycle time			
		6-h	8-h	12-h	24-h
COD, mg/l	3068.3± 548.7	892± 177.2	458± 62.8	331.7± 24.6	234.2± 17.8
TN, mg/l	319± 39.6	212.6± 39.6	195± 30.6	175±34	163±28



*The mass of COD removed/day= no. of cycle × the mass of COD removed/cycle.

Figure 3. Influence of cycle length on COD removal efficiency and mass of COD removed per day

Table 2: The treatment performance of SBR at 8-h cycle

Parameter	Influent	End of anoxic	Effluent	%, Removal
COD, (mg/l)	3850 ±182	1730 ± 121	520 ± 64	85
TN, (mg/l)	340 ± 12.7	255.7 ± 6.7	210.8 ± 9.5	38
NH_4^+-N (mg/l)	152 ± 9.5	207 ± 9.5	134 ± 11.1	35
Sulfide (mg/l)	274.8±47.4	2.19 ± 1.0	0.15 ± 0.06	99.9
Orthophosphate (mg/l)	12.4 ± 3.4	1.13 ± 0.2	0.00 ± 0.00	100
Total Chromium(mg/l)	10.2 ± 0.31	-	1.319 ± 0.5	87.0
pH	11.7 ± 0.9	8.17 ± 0.18	8.1± 0.12	
EC , μ s/cm	18250± 2202	-	17114±2929	-
TDS (mg/L)	7935 ±1089	-	7677±1176	-

This indicated that the COD mass removal rate per day was highest in the 6-hrs and lowest in 24-hrs cycle time. On the other hand, the removal efficiencies were lowest in 6-hrs and highest in 24-hrs cycle time. The 6-hrs cycle time showed residual COD of roughly 30 % from the influent, which needs for further treatment. The mass removal rates per day and removal efficiencies were higher in the 8-hrs cycle. The average removal efficiency of 8-hrs cycle time was significantly greater than 6-hrs cycle time and not significantly different from 12-hrs cycle time (p=0.139). In addition, its' COD removal rate per day was significantly different from 24-h cycle time (p=0.025). The high effluent COD (350–400mg/l) was indicative of the presence of bio-recalcitrant chemicals in tannery wastewater (Carucci *et al*., 1999; Di Iaconi *et al*., 2002) such as the melamine and phenol–naphthalene based synthetic tanning agents (Arun kumar *et al.,* 2004). Although the 12-hrs and 24-hrs cycle times have good effluent quality, they have had low mass COD removal rate per day. However, the 8-hrs cycle time could be taken as the best cycle time, as cost, time and effluent quality are taken into considered in the treatment process.

There was also a variation in TN removal efficiency with cycle time. The average values of TN in the effluent at the end of 6-hrs, 8-hrs, 12-hrs and 24-hrs cycle were: 212.6 ± 39.6, 195 ± 30.6, 175 ± 34, 163 ± 28mg/l, respectively (Table 2). The lowest TN removal efficiencies (33%) was obtained at cycle time of 6-hrs while the removal efficiencies at 8, 12 and 24-hrs cycle times were 39, 44 and 49%, respectively. The removal efficiency of 6-hrs is much less significantly different than that of 8, 12 and 24-hrs cycle time ($p< 0.001$). The 24-hrs cycle has greater significant removal efficiency than 8-hrs ($p< 0.001$) while the mass removal rate of 8-hrs was significantly greater than 24-hrs ($p< 0.001$). Generally the COD and TN removal efficiencies were increased with increasing cycle times. This indicated that the removal efficiencies increased with increasing cycle time. This might be due to increasing contact time of the substrate to micro-organism. Similar observation was also made by Moosavi *et al.* (2005).

3.3. Process Performance at Optimal cycle time (8hrs)

The average characteristic of the influent, the supernatant at the end of the anoxic phase and effluents are given in the Table 2. The influents were characterized by high alkalinity content with a resulting pH value of above 10.0 due to the chemicals used in leather processing. Influent COD and total N concentrations were 3850 ±182 and 340 ± 12.7 mg/l, respectively, whereas influent ammonium-N- 152 ± 9.5 mg/l. Sulphide and Tot-Cr concentrations were 274.8±47.4 and 10.2 mg/l, respectively.

The COD concentration in the supernatants at the end of anoxic phase was 1730 ± 121mg/l and the concentration in the treated effluents was 580 ± 64 mg/l. The overall removal efficiency of the system was 85 % (Table 2). The resulted obtained in study were compared with other studies; Farabegoli *et al.* (2007) found removal efficiency of 67% from treatment of tannery wastewater in SBR using 6-hrs cycle time. Carucci *et al.* (1999) found 84% COD removal from tannery wastewater in SBR using 8-hrs cycle. The removal efficiencies obtained in 24-hrs cycle time (93%) was also in line with the reported COD removal efficiency of 90% by Murta *et al.* (2006) from tannery wastewater in full scale SBR using 24-hrs cycle time).

Total nitrogen and ammonium-N in the supernatants at the end of anoxic phase were 255.7 ± 6.7mg/l and 207 ± 9.5mg/l, respectively. Total nitrogen in the anoxic phase was decreased from influents concentration while ammonium-N increased from152 ± 9.5 to 207 ± 9.5mg/l (Table 2). This increment of ammonium was resulted from the hydrolysis of protein into amino acid by hydrolytic micro-organisms followed by ammonification of amino acid by consortia of acidogenic bacteria. However, the removal efficiencies found for TN and $NH4^{+}$-N were low. This might be associated with inhibition of nitrification by excessive COD loading. This can be attributed to the depletion of dissolved oxygen caused by heterotrophic organisms which utilized the organic matter present in the wastewater (Barnes and Bliss, 1983). This phenomenon was postulated to be due to low levels of dissolved oxygen due to autcompetition by heterotrophs, even though dissolved oxygen meters values indicated sufficient levels of oxygen were present in the bulk solution (Hanaki *et al*., 1990; Van *et al.,* 1993).

On the other hand, the concentration of orthophosphate was decreased from 12.4 ± 1.9 to 1.13 ± 0.2 mg/l at the end of anoxic phase and further reduced to 0.00 ± 0.00mg/l. The observed removal efficiency was 100 % (Table 2). The study obtained in this study is higher than other studies; obaja *et al* (2003) reported 97.3% removal from poultry wastewater in SBR while Lefebvre *et el* (2005) reported 93% removal from tannery wastewater in SBR.

The concentration of sulfide was drastically decreased from the value of 274.8 ± 6.3mg/l to 2.19 ± 1.0 mg/l at the end of anoxic phase and it further reduced to 0.15 ± 0.06. In the anoxic phase, sulfide concentration was reduced by 99.2 %. This showed that sulfide removal was highly occurred in the anoxic phase. The overall sulfide removal efficiency was 99.9 % (Table 2) which is comparable to the reported values (99.6%) by Chung *et al.,* (1997) using biofilters and Che et al., (2005) reported 100% sulfide removal using *Thiobacillus novellus* in biofilter under mix trophic condition.

The level of total chromium in the final effluent was decreased from 10.2 ± 0.31 to 1.319 ± 0.5 mg/l and its' removal efficiency was 87.0 % (Table 2). This removal of chromium might be occurred due to the precipitation of chromium as chromium hydroxide and/or adsorption on the sludge (David *et al*., 1994).

It was interesting to note that the effluent values of COD, Sulfide, orthophosphate and chromium that were observed in the final effluents were in line with the permissible limits for discharge of tannery effluents into surface water bodies adopted in many countries compiled by the United Nations Industrial Organization (UNIDO, 2000).

3.4. The Effects of feeding length on COD removal

A set of four feeding length (i.e. 180, 83, 48 and 39 minutes) was tested for COD removal efficiencies with average volumetric loading rate of 3.1kg/m3/day. As Table 3 shows, in feed I operation, the concentration of COD was 1185.3 ± 10.7 mg/l in anoxic phase and it was reduced to 223 ± 35.1 mg/l in the treated effluents. In feed II operation, the COD were 1413 ± 32.8 mg/l and 420± 24.3 mg/l in the anoxic and the oxic phase, respectively. In feed III operation, the COD were 1830 ± 25.16 mg/l and 603 ± 34.8 mg/l, in the anoxic and the oxic phases, respectively. In the final feed operation COD level were 1873± 35.3 mg/l in the anoxic and 595 ±8.8 mg/l oxic phases. This indicates that the concentration of COD decreased with increasing feed length in the final effluents.

Table 3. Influence of fill time on COD removal in the anoxic and oxic phases.

Feed	Fill time (minute)	Influent (mg/l)	End of anoxic (mg/l)	Effluent (mg/l)
I (F_1)	180	4183.3 ± 25.166	1185.3 ± 18.6	223 ± 61.1
II (F_2)	83	4203.3 ± 20.8	1413 ± 56.9	420 ± 42
III (F_3)	48	4123.3 ± 51.3	1830 ± 43.6	603 ± 60
IV (F_4)	39	4140.3 ± 4.5	1873 ± 61.1	595 ± 15.3

The average removal efficiencies in the anoxic and aerobic phases were: 75 and 81% for feed I, 66 and 73% for feed II, 57 and 67% for feed III and 55 and 68% for feed IV while the overall removal efficiencies obtained were 94, 90, 85 and 85%, respectively (Fig.4).

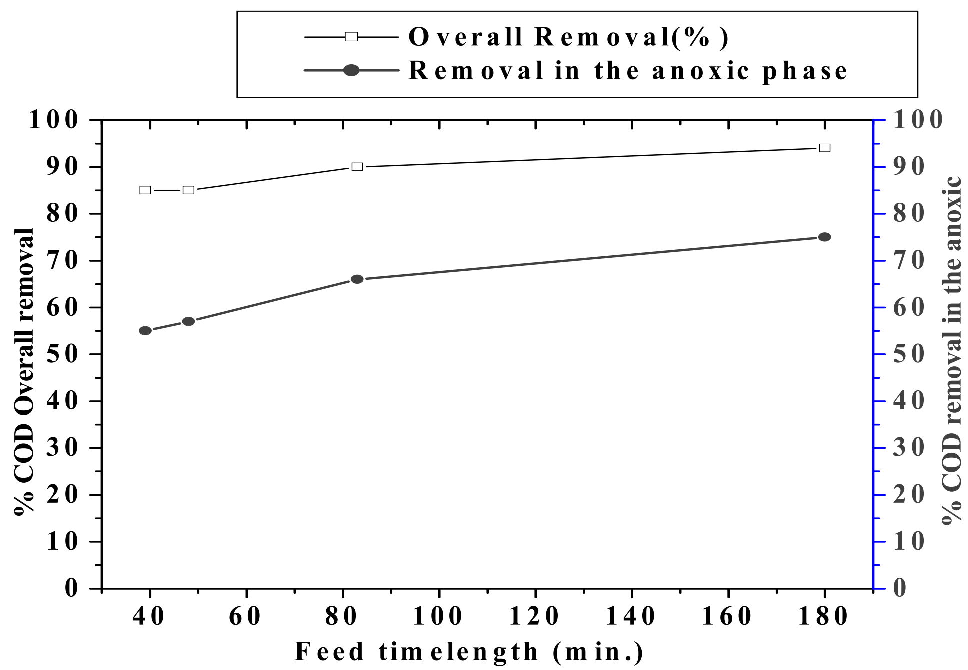


Figure 4. Overall and anoxic COD removal with increasing fill times

The overall COD removal of F_1 (94%) was significantly greater than F_2 (90%) ($p<0.001$) while, the overall COD removal efficiency of F_3 (85%) was significantly less than the removal efficiency of both F_1 and F_2 ($p<0.001$). This study indicate that the COD removal efficiency in SBR treatment technique increase with increasing the feeding time ($p<0.001$). In the anoxic phase, the removal efficiencies were increased (55 - 75 %) with increasing feed time from 39 minutes to 180 minutes. In the anoxic phase length of feed has direct relation with COD removal efficiency (Kennedy *et al.*, 1991). Microorganisms exposed to high OLRs during the fill cycle (i.e. short fill time) may behave differently than microorganisms exposed to similar influent wastewater concentration but operated at longer fill time. The increase in feed time would cause a reduction in total volatile acids (TVA). This reduces the availability of substrate to microorganism and cause less accumulation of these acids. This in turn increases reactor performance (Kennedy and Lentz, 2000). However, the removal efficiency of COD decreased with decreasing fill time. This might be at a high organic load, the overall growth rate of acidogenic bacteria proceeds faster (10-fold) than that of methanogenic bacteria and inhibitory products such as volatile fatty acids and H_2 accumulate in the reactor, which slows slow down the entire process (Ronnachai *et. al.,* 2007).

5. Conclusion

The study showed that sequencing batch reactor is an efficient tool for COD, sulfide, chromium and phosphorus removal from composite tannery wastewater. At the optimum cycle, COD, Sulfide, Chromium and phosphorus removal yields are around 85, 99.9,87 and 100%, respectively. The time cycle and the duration of fill time within the time cycle must be designed properly, in order to optimize the removal of efficiencies. The choice of hydraulic residence times and cellular retention times will depend on this optimization. Effect of period of filling was varied from 180 to 39 minutes and a very good COD removal was observed with increase of period of fill time (94% reduction in 180 minutes of fill period and decrease in fill period to 39 minutes result in deterioration of the treatment process and poor COD reduction (85% for 39 and 48 minute fill period). A single sequencing batch reactor was not efficient for the removal of nitrogenous compounds from tannery strong wastewater. This is an issue to be investigated in future work with multiple batch reactors in sequence.

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