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Research Article

STUDY ON ROBUSTNESS OF AEROBIC MEMBRANCE BIOREACTOR IN THE TREATMENT OF PHARMACEUTICAL INDUSTRIAL EFFLUENT

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ABSTRACT

Industrial development along with increase in population leads to environmental pollution and water scarcity. Hence there arise a need for wastewater treatment and water reuse. Membrane bioreactor is an advanced wastewater treatment technology which is a combination of activated sludge process with micro – and ultra filtration. The present study was carried out to investigate the robustness of MBR in treating pharmaceutical wastewater under different hydraulic shock loads. The study was carried out in a lab scale aerobic submerged membrane bioreactor. The membrane bioreactor was installed and operated at three different HRTs (8 h, 6 h and 4 h) with the flow rates of 0.75 L/h, 1 L /h and 1.5 L / h. Reactor was run for the duration of 8 days at each hydraulic retention time (HRT) and the effluent characteristics were studied to find out optimum HRT. The hydraulic shock loads was imposed to the MBR gradually by increasing the influent flow rate in stepwise manner. Each shock was applied for duration of 24 h, after 24 h the flow rate was brought back to steady flow. The effect of each hydraulic shock load was assessed by comparing the effluent quality, sludge characteristics before and after shock load. Removal of organics in the MBR was around 86 - 88 % efficiency, TSS removal efficiency of 87%, TKN and Phosphate removal efficiency of 84 % and 31 % respectively was reported in the steady state condition (HRT 8 h). COD, BOD, TSS, removal efficiencies at hydraulic shock load conditions (4 h ,2 h, 1.3 h) decreased to 50 -30 %, 51 - 37 % and 64 - 33% respectively in comparison with the steady state condition (HRT 8 h). A decrease in TKN and phosphate removal efficiency to 59 - 41 % and 6 - 7 % respectively was also observed at different hydraulic shock loads.

INTRODUCTION

Production of pharmaceutical product requires many types of solvents namely alcohols, glycol ethers, ketones, esters, glycol ether esters, chlorinated hydrocarbons, aliphatic and aromatic hydrocarbons. The effluent coming out from the manufacturing unit consists of high TDS, TSS, COD, BOD, traces of the pharmaceutical products and Solvents. The production of bulk drugs has recently been identified as an important source for environmental pollution (water) with active pharmaceutical ingredients (APIs). The effluent from pharmaceutical industry are loaded with pathogenic microorganism, solvents, pharmaceuticals partially metabolized, radioactive elements, carcinogens, phenol, sulphate, Total Kjeldhal Nitrogen (TKN), ammonia and other toxic chemical substances. It is worth mentioning that majority of current wastewater treatment plants were not designed to deal with these types of compounds. Therefore the treatment of pharmaceutical wastewater requires some complementary techniques that could efficiently remove pollutants and enable the wastewater to be discharged into receiving water body or be reused for industrial purpose. The increasing volume of wastewater combined with limited space availability and progressively tightening environmental standards has resulted in advanced treatment technology namely the Membrane Bioreactor (MBR) technology. Membrane bioreactors (MBR) are commonly understood as the combination of membrane filtration and biological treatment using Conventional Activated Sludge (CAS) where the membrane primarily serves to replace the clarifier in the wastewater treatment system. The benefits of MBR process over the conventional activated sludge process includes reduction in the space and reactor size requirements, better effluent quality, disinfection, increased volumetric loading and less sludge production. It can retain all the biomass facilitating the control of Solid Retention Time (SRT), better operation reliability, stability, easy automatic control and compactness of the whole system. These advantages make MBR a valuable alternative over other treatment technologies.

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MATERIALS AND METHODS

Characterization of pharmaceutical wastewater

Physicochemical characteristics of the pharmaceutical wastewater taken from a pharmaceuticals industry at Chennai., are given in Table 1. The pH of the wastewater was around 6.8 – 7.5, the BOD and COD was 3640 - 6000 mg / L and 5420 - 9392 mg / L respectively. TSS was around 560 – 230 mg / L. The BOD/COD ratio of the wastewater was 0.67, which indicating the good biodegradability of the wastewater (Metcalf and Eddy 2003). The TKN and Phosphate concentration of the pharmaceutical wastewater was found to be 610 - 290mg / L and 60 – 30 mg / L respectively

Table 1 Characteristics of pharmaceutical wastewater.

Sl.No	Parameter	Unit	Concentration
1	BOD	mg/L	3640 - 6000
2	COD	mg/L	5420 - 9392
3	TSS	mg/L	560 - 230
4	TDS	mg/L	5222 - 4013
5	TKN	mg/L	610 -290
6	Phosphate	mg/L	60 - 30
7	pH	-	6.8 -7.5

The characteristics of the wastewater collected from the Industry vary in a wide range from time to time. Therefore the wastewater was diluted with distilled water and 200 milligram of Di-ammonium phosphate was added per litre of wastewater to obtain constant characteristics. Di-ammonium phosphate was added to supply sufficient nutrient to the microbial growth in biomass. The characteristics of wastewater in MBR are given in table 2 which are analyzed as per "Standard methods for the examination of water and wastewater".

Table 2 Characteristics of MBR influent wastewater.

Sl.No	Parameter	Unit	Concentration
1	BOD	mg/L	1200 ±100
2	COD	mg/L	1800 ± 50
3	TSS	mg/L	110 ± 5
4	TDS	mg/L	1700 ± 200
5	TKN	mg/L	120 ± 5
6	Phosphate	mg/L	60 ± 5
7	pH	-	6.8 -7.5

The pH of diluted wastewater was 6.8 – 7.5, the BOD and COD was 1200 ± 100 mg / L and 1800 ± 50 mg / L respectively. The TSS was around 110 ± 5 mg / L. The BOD/COD ratio of the wastewater was 0.67, which indicating the good biodegradability of the wastewater (Metcalf and Eddy 2003). The TKN and Phosphate concentration of the pharmaceutical wastewater was 120 ± 5 mg / L and 60 ± 5 mg / L respectively.

Experimental Set up

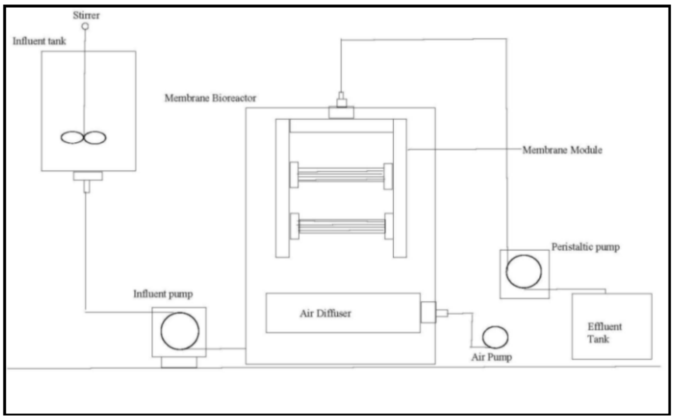

Fig 1 Schematic diagram of MBR

The experimental setup consists of a hollow fibre membrane immersed in the reactor of size about 500 mm × 350 mm × 100 mm with working volume of 6 litres. A schematic diagram of the membrane bioreactor experimental setup is represented in Figure 1. The membrane is Polysulfonate hollow fibre membrane manufactured by Davey Products, Chennai. The specifications of the membrane module are summarized in table 3. An influent tank with a capacity of 20 L was installed at a height above the reactor level and the outlet connected with a peristaltic pump in which flow rate can be adjusted and maintained. The outlet of the membrane module connected to effluent tank through a peristaltic pump. Characteristics of the wastewater collected from the Industry vary in wide range from time to time. Therefore the wastewater was diluted with distilled water and 200 mg of Di-ammonium Phosphate per litre was added to wastewater to obtain constant characteristics. Di-ammonium Phosphate was added to supply sufficient nutrient for the growth of microbial biomass.

Table 3 Specifications of membrane module

Sl.No	Item	Details
1	Membrane Material	Polysulfonate
2	Membrane Type	Hollow fibre
3	Pore Size	0.5 µm
4	Surface Area	0.2 m^2
5	Size of Fibre	1.4 mm OD
6	MOC of Housing	UPVC
7	Operating Pressure	<3 Kg / cm^2
8	Trans Membrane Pressure	2 Kg / cm^2 max
9	Operating pH	1 -13
10	Operating Temperature	45^{o}C
11	Filtrate Flux	50 -100 L/m^2/hr
12	Flow	Out – In

Seed biomass for the reactor was collected from secondary settling tank of wastewater treatment plant in the pharmaceutical industry and was acclimatized with the wastewater in the MBR. The wastewater then fed to the reactor from a feed tank containing stirrer to maintain homogeneity of influent fluid. The effluent is pumped using a peristaltic pump. Air is being supplied through the fine bubble tube diffuser in the reactor. Air flow rate of 0.5m^3/hr was maintained to achieve suspension of the organism, increase contact between organics and organism, avoid scouring and fouling problem and maximize gas liquid solid separation. The effluent from membrane bioreactor was removed using peristaltic pump connected to the membrane module.

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Acclimatization of biomass

Sludge from the secondary settling tank of wastewater treatment plant in pharmaceutical industry at Chennai, with an MLSS concentration of around 5000 mg/L was used as seed inoculums. It was acclimatized and cultivated as fill and draw process in the membrane bioreactor the contents was mixed and aerated by compressed air through diffusers to maintain DO of 1.5 to 3.0 mg/L. Operating cycle of MBR was 1 h fill, 20 h react, 2 h settle and 1 h decant. pH of the reactor was maintained around 6.5 to 7.5.

Operation of aerobic MBR and optimization of HRT

MBR was operated continuously and the DO concentration was maintained between 2 - 6 mg/L. The MLSS concentration in the reactor was maintained between 10 – 14 g/L and the pH of the reactor was maintained between 6.5 to 7.5. Aerobic MBR was run at three different HRTs of 8, 6 and 4 h and the performance of the MBR was assessed in terms of BOD, COD, TSS, TDS, TKN, and phosphate removal. The HRT with maximum removal efficiency was chosen as the optimum HRT of MBR. The performance of MBR under stable condition was evaluated in terms of effluent quality (pH, BOD, COD, TSS, TDS, TKN, and phosphate); and sludge characteristics (pH, DO, MLSS, MLVSS).

Evaluation of the performance of MBR at different hydraulic shock load

Performance of the membrane bioreactor was stabilized at optimum HRT with a steady state flow rate. Hydraulic shock load was imposed to this reactor gradually by increasing the influent flow rate in stepwise manner up to six fold (two fold, four fold, six fold,). Each shock was applied for duration of 24 h, after 24 h the flow rate was brought back to steady flow. Effluent samples were collected every 8 h during the shock and once in 24 h after cessation of shock.

The effect of each hydraulic shock load was assessed by comparing the effluent quality, sludge characteristics before and after the shock load. Reactor was operated with steady state HRT till the reactor completely reverts back to steady condition before the next HRT shock load. The ability of the MBR to recover from each imposed hydraulic shock load was evaluated by continuous monitoring of the effluent quality and sludge characteristics, till the reactor performance reverted to steady state condition. Time taken for the MBR to revert back to steady state was calculated for each of the hydraulic shock load.

RESULTS AND DISCUSSION

Cultivation and acclimatization of biomass

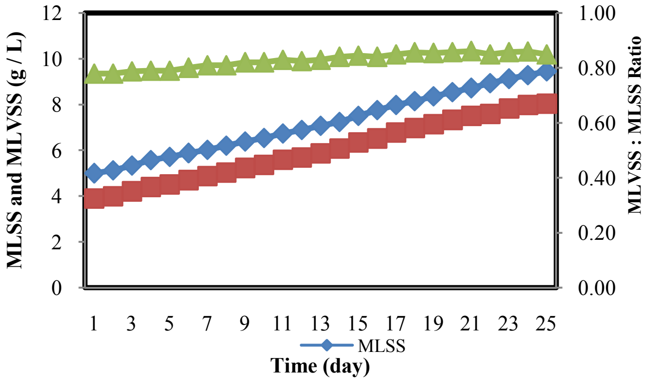

Fig 2 MLSS and MLVSS during acclimatization

Activated sludge collected from secondary settling tank of wastewater treatment plant in pharmaceutical industry at Tamilnadu was acclimatized to the pharmaceutical wastewater by fill and draw process in the lab scale MBR. During the acclimatization process, the pH and the DO were maintained in the range of 7.0 – 7.5 and 1.5 – 3 mg / L, respectively. Figure 2 shows the development of MLSS and MLVSS during the acclimatization process. Initial MLSS and MLVSS were 5.0 g / L and 3.9 g / L, respectively. MLSS increased gradually from 5 g / L to 9.458 g / L in 25 days. MLVSS also showed corresponding increase with MLSS, maintaining MLVSS/MLSS ratio of 0.85.

Operation of MBR and optimization of HRT

Performance of MBR at three different HRTs of 8 h, 6 h, and 4 h with flow rates of 0.75 L / h, 1 L / h, 1.5 L / h are studied to optimize HRT. pH in the MBR varied from 6.5 to 7.5 throughout the experiment. According to Metcalf and Eddy this pH range is e optimum for the proper growth of microorganism. The pH of treated effluent was in the range of 7 -7.5. DO variation during the optimization of HRT illustrated in Figure 3 shows that the DO was maintained between 2.4 – 4.6 mg / L. The DO level decreases with increase in HRT.

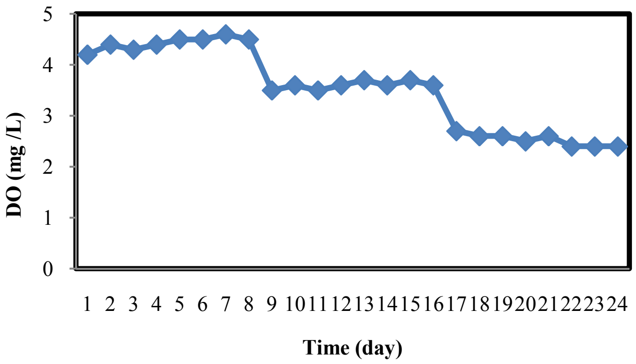

Fig 3 Variation in DO in the MBR at different HRTs

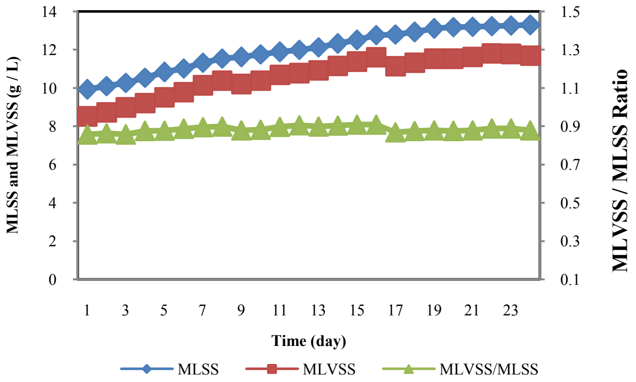

Fig 4 MLSS and MLVSS in MBR

Initial concentration of MLSS and MLVSS was 9.9 and 8.5 g / L respectively. The biomass increased gradually and reached to the MLSS and MLVSS concentration of 13.3 and 11.7 g / L respectively, the MLSS/MLVSS ratio varied in the range of 0.85 – 0.9. The Figure 4.4 represents the development of MLSS and MLVSS during optimization of HRT in MBR. The initial MLVSS / MLSS ratio was 0.85, which later increased to 0.9 with increase in MLSS and MLVSS concentration . This increase in MLVSS/MLSS ratio might be due to high retention of microorganisms within MBR, preventing them from being washed out. More growth of microorganism also contributed the increase of MLVSS/MLSS ratio in the MBR (Ben Aim and Semmens 2003)

Removal of Total Suspended Solids

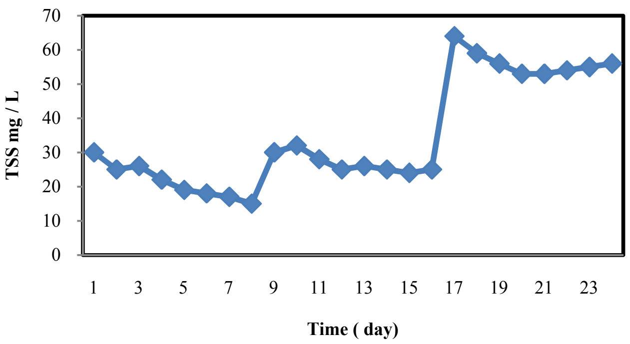

Fig 5 TSS in the MBR effluent at different HRTs

Figure 5 shows the TSS level in the MBR effluent at different HRT. MBR showed higher TSS removal efficiency of 86.72 % at HRT 8 h. TSS removal efficiency at 6 h and 4 h HRTs were 79.92 % and 48.15 % respectively. The average TSS in the effluent was 21 mg / L, 25 mg / L and 56 mg / L for HRTs of 8 h, 6 h and 4 h respectively. Suspended solid concentration in the effluent decreases because the MLSS (biomass) concentration was high, also the high MLVSS shows less death rate of biomass which leads to less TSS in the effluent according to Cote *et al*., (1997).

Removal of Organics

Figure 6 and 7 represents the concentration of BOD and COD in the MBR effluent at different HRTs. The BOD removal efficiency for 8 h, 6 h and 4 h HRT was 86.25 %, 79.92 % and 38 % respectively. The average effluent BOD was 250 mg/L, 334 mg/L and 765 mg/L for 8 h, 6 h and 4 h HRT respectively.

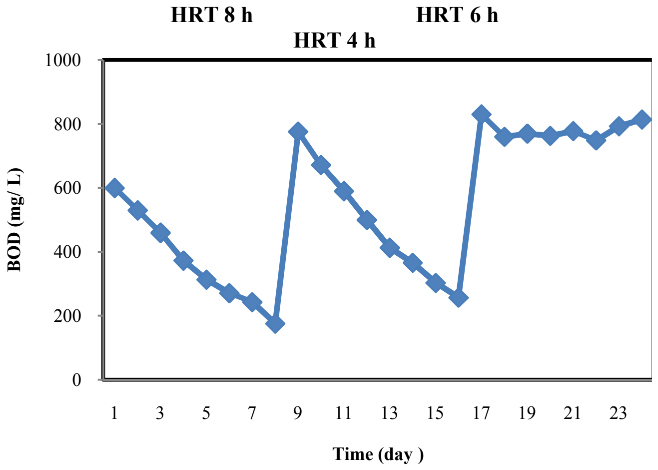

Fig 6 BOD in MBR effluent at different HRTs

Organic loading rate varied from 3.8 g COD/L/d to 7.3 g COD/L/d as the HRT range varied from 8 h to 4 h. Average effluent COD at 8 h, 6 h and 4 h HRT was 286 mg/L, 364 mg/L and 678 mg/L and the removal efficiency was 87.55 %, 83.66 % and 43.9 % respectively.

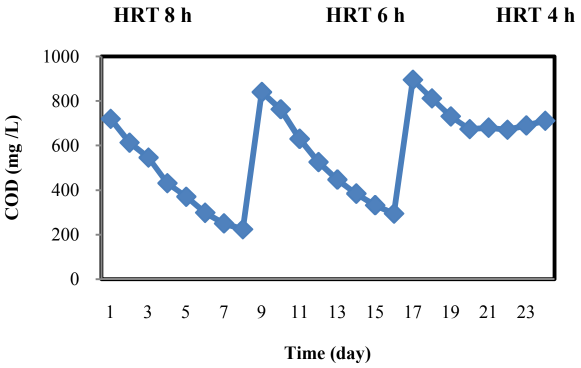

Fig 7 COD in MBR effluent at different HRTs

The high percentage of organic removal can be attributed first to the complete retention of all particulates by the membrane. Zhang *et al*., (2003) reported that high sludge concentration in the system is another important reason for such high organic removal because high MLSS could significantly absorb soluble organic compounds. Further, the washout of microorganism was avoided in the MBR (Rosenberger *et al*., 2002). These conditions could establish specialized microorganism that are able to remove slowly degradable components.

Removal of Nitrogen and Phosphate

Figure 8 represents the variation of TKN in MBR effluent at different HRTs. The TKN removal efficiency of MBR at HRTs 8 h, 6 h, and 4 h was 83.47%, 73.91 % and 51.3 % respectively. The average effluent TKN was 18 mg/L, 31 mg/L and 52 mg/L for 8 h, 6 h and 4 h HRT respectively.

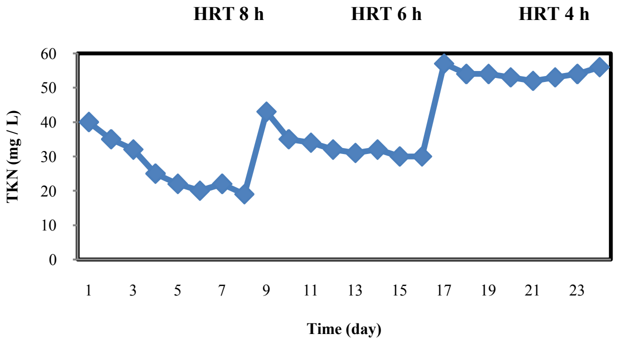

Fig 8 TKN in MBR effluent at different HRTs

Nitrogen can be removed by assimilation into biomass or by nitrification process (Rosenberger *et al.,* 2002). Nitrogen removal by assimilation was evidenced by continuous increase in MLVSS (biomass) concentration. During start-up, the nitrogen assimilation was slow and most of the nitrogen passed into the effluent. In the present study the reduction in TKN in the effluent was observed only on 8th day. This might be due to slow growing nitrifying bacteria which needs long generation time to establish and reach sufficient population to nitrify ammonia in the wastewater. Nitrifying bacteria reproduce very slowly due to the low energy obtained from the oxidation of ionized ammonia and nitrite. Also, high biomass concentration within the reactor and prevention of washout of microorganisms by the membrane filtration would have enabled high TKN removal efficiency. As the membrane completely retained the nitrifying microorganisms in the reactor these autotrophic nitrifies could proliferate without any loss (Lee *et al* 2003). Figure 9 illustrates the variation of phosphate in the MBR effluent at different HRTs.

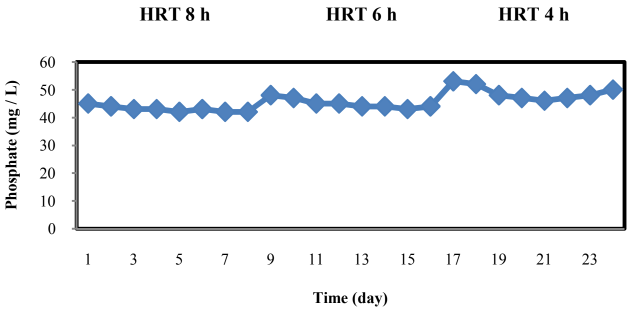

Fig 9 Phosphate in MBR effluent at different HRTs

Phosphate removal efficiency of 31.14 % was the only maximum at 8 h HRT with an average phosphate concentration in the effluent was 43 mg / L. In the HRT of 6 h and 4 h phosphate removal was very less at 27 % and 18% respectively. Phosphate also can be removed by assimilation in to biomass (Rosenberger et al 2002). The reason for low phosphate removal was that the required phosphate was assimilated by the biomass and the remaining is just washed out in the effluent. Also the phosphate was provided in excess with a COD: N:P ration of 100:11:2 (Yogalakshmi 2009).

Performance of MBR at Hydraulic shock loads

Performance of the MBR under a Transient Shock Loads of 4 h, 2 h and 1.3 h HRTs for a duration of 12 h are presented and compared with the steady state performance (8 h HRT). Variation of TSS in the MBR effluent at different hydraulic shock load conditions is represented in Figure 10. In the two fold hydraulic shock load (1.5 L / h)), TSS in the MBR effluent was increased from 14 to 41 mg / L. Similar response of drastic increase in concentration was observed during four (3 L / h)and six fold (4.5 L / h) increase in flow. The effluent TSS at four and six fold increase was 49mg / L and 76 mg / L respectively. Hydraulic shear on the flocks may have lead to the floc breakage which would have been carried away from the MBR with the effluent contributing to increased effluent TSS. Similar observations were also reported by Nachaiyasit and Stuckey (1997) and Yogalakshmi (2009).

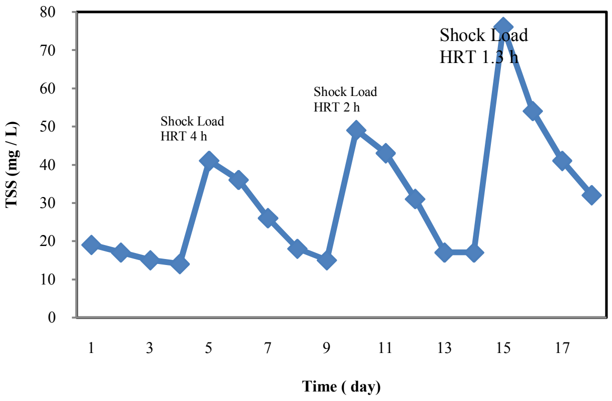

Fig 10 TSS in MBR effluent at different hydraulic shock loadings

When compared to other treatment process such as activated sludge process, the washout of TSS in MBR during hydraulic shock was less. This is because the membrane in the bioreactor prevents the washout of microorganism, which in turn prevents the increase in effluent TSS concentration. TSS concentration in the MBR effluent returned to steady state performance within 2.5 – 3.5 days.

Removal of Organics at different hydraulic shock load conditions

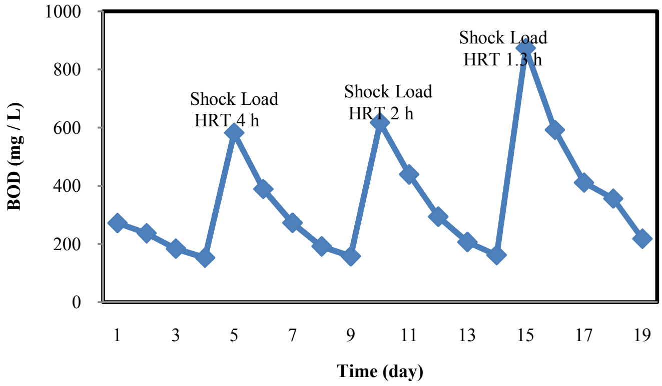

Fig 11 BOD in MBR effluent at different hydraulic shock loadings

Variations of BOD and COD in the MBR effluent under different hydraulic shock load conditions are represented in Figures 11 and 12. At the flow rate of 1.5 L / h, the BOD and COD removal efficiency dropped to 53 % and 60 % with the concentration in the effluent increased to 582 mg / L and 711 mg / L, respectively. BOD and COD removal efficiency dropped further at the flow rate of 3 L / h and 4.5 L / h.

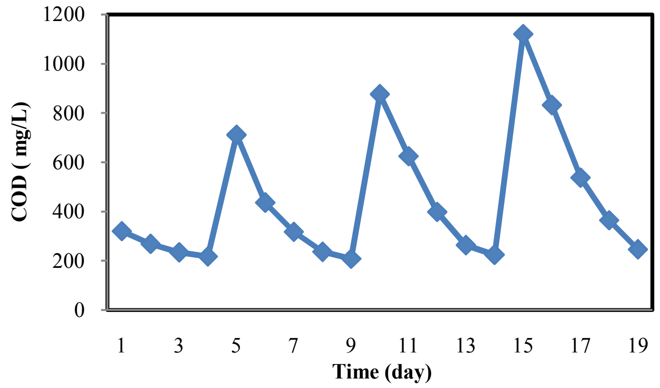

Fig 12 BOD in MBR effluent at different hydraulic shock loadings

The BOD and COD removal efficiency at the flow rate off 3 L / h and 4.5 L / h was 50%, 51.5% and 30%, 37.7% respectively. The BOD concentration in the MBR effluent at the flow rate of 3 L / h and 4.5 L / h was 617 mg / L and 876 mg / L respectively also COD concentration in the MBR effluent at the flow rate of 3 L / h and 4.5 L / h was 873 mg / L and 1120 mg / L respectively. Effluent BOD and COD return to steady state performance within 2 – 3 days.

The main reason for such transient disturbances during both the hydraulic shock loads might be due to release of metabolic products of original carbon source and also due to leakage of carbon source itself (Krishnan and Gaudy 1976). Another reason for such transient disturbances according to Grobicki and Stuckey (1991) was the result of decreased reaction time for biomass to maintain high removal efficiency in the reactor.

Removal of Nitrogen and Phosphate at different hydraulic shock load conditions

Figure 13 represents the variation of TKN in the MBR effluent at different hydraulic shock load conditions. At the flow rate of 1.5 L / h, 3 L / h and 4.5 L / h, the TKN removal efficiency dropped to 59.83 %, 49.57% and 41.02% respectively. At the flow rate of 1.5 L / h, 3 L / h and 4.5 L / h, TKN concentration in MBR effluent increases to 47 mg / L, 59 mg / L and 69 mg / L respectively. The MBR took around 2- 3 days to attain steady state.

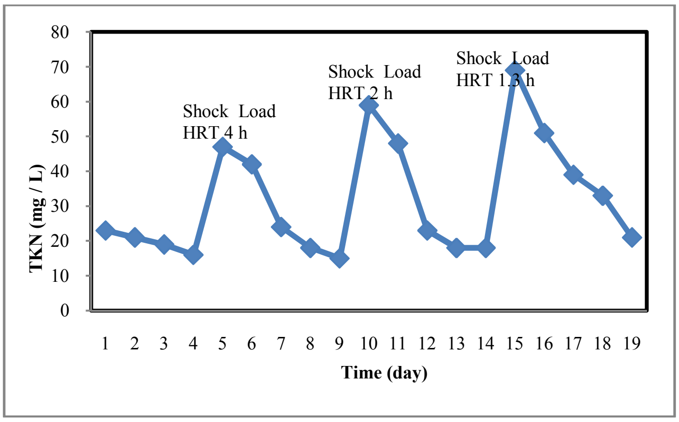

Fig 13 TKN in MBR effluent at different hydraulic shock loadings

During the hydraulic shock load the transfer rate of substrate into the biomass might be limited due to decrease in contact time between substrate and biomass (Grobicki and Stuckey 1991). Metabolism rate is also very slow at short HRT because of very short contact time of biomass with substrate (Nachaiyasit and Stuckey 1997 b). Another reason for increase in TKN in effluent was due to nitrification inhibition

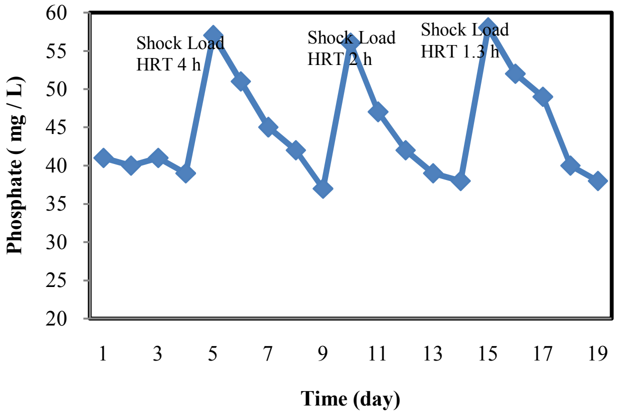

Fig 14 Phosphate in MBR effluent at different hydraulic shock loadings

The variation of phosphate in the MBR effluent under different hydraulic shock load conditions is represented in figure 14. The effluent phosphate increased to 57 mg / L reducing the removal efficiency to 7 % at the flow rate of 1.5 L / h. At the flow rate of 3 L / h and 4.5 L / h, the effluent phosphate was 56 mg / L and 58 mg / L, respectively. During these shocks the system was seriously affected showing leakage of substrate as such without being metabolized. Lack of contact time would also be the reason for such poor phosphate removal. Overall, the phosphate was simply washed through without being metabolized because of too short contact time during the hydraulic shock (Nachaiyasit and Stuckey 1997). Steady state was obtained after 2 – 3 days for all hydraulic shock loads.

CONCLUSIONS

Result of the present study reveals the robustness of the aerobic MBR to hydraulic shock load of pharmaceutical wastewater. High retention capacity of the membrane coupled with high biomass concentration has made the MBR robust to hydraulic shock load. It may be further concluded that (i) The performance of MBR in the treatment of pharmaceutical wastewater was good with the removal efficiency of total suspended solids >86 %, biodegradable organics 86 % and nitrogen 83 % enabling the reuse of treated effluent after secondary treatment. (ii) Washing-out of the microorganisms from MBR is prevented which increases the MLSS (biomass) concentration to 14.7 g / L, which is also the major factor in increasing the efficiency of MBR. (iii) Efficiency of nutrient removal was reduced during shock loads. Steady state was attained within 2 – 3 days. (iv) Steady state was attained at faster rate after each shock loads (4 h and 2 h HRT), hence the shock loads does not affect the performance of MBR. Thus the study reveals that the MBR can be one efficient technology for treating pharmaceutical wastewater.

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