Development of sequential batch ozonated adsorptive membrane bioreactor to mitigate fouling with reduced energy consumption

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Abstract−The present study focuses on overcoming the drawback as fouling in a membrane bioreactor (MBR), which can be alleviated by integrating advanced oxidation process, adsorption, and biofilm carriers in the activated sludge process. The optimal sludge retention time, carbon and ozone dosage was 150 minutes, 15 g and 1.5 $Lmin^{-1}$, respectively. The percentage removal was observed to be above 90% for chemical oxygen demand and total organic carbon whereas for total dissolved solids was only 40% under transmembrane pressure of 20 kPa. The increase in permeate flux was 30% as compared to MBR. Sequential batch membrane bioreactor (SBMBR) showed 12% reduction in energy consumption for three hour operation at the flow rate of 0.72 L/h (transmembrane pressure 20 kPa), and it was confirmed in the SEM of carbon, membrane, UV, CV and HPLC also. The energy consumption required also confirms the less internal fouling via the extended backwash of four hours.

Keywords: Sequential Batch Reactor, Biofilm Carriers, Activated Carbon, Ozonation, Permeate Flux, Activated Sludge Process

INTRODUCTION

The main drawback associated with a membrane bioreactor is fouling, which increases the operation and maintenance costs (O&M) [1]. Fouling is due to the presence of hydroxyl groups and carboxyl groups in the wastewater. In particular, the presence of hydroxyl group creates high physical irreversible fouling, because the affinity towards the barrier is higher as compared to carboxyl groups. This affinity may be due to the higher adhesive force, which leads to the generation of a hydrogen bond. Concentration polarization is one of the major drawbacks which influences the flux reduction by means of increasing the osmotic pressure. This can be controlled two ways, either by increasing turbulence on the membrane facade, which increases the back force of the solute from the barrier, or by reducing the solute concentration of influent towards the membrane. This physical irreversible fouling can be avoided by implementing some pretreatments, such as an advanced oxidation process (AOP), coagulation, and adsorption to remove organic and inorganic pollutants. Normally, AOP degrades the pollutants by generating hydroxyl radicals and converting the pollutants into carbon dioxide and water. The most prominent AOP nowadays used is ozonation when combined with biodegradation results in the reduction of organic compounds [2]. Normally, ozonation is an energy-extensive process [3] and is not suitable for highly suspended solids containing wastewater [4]. The main advantage of using ozonation is the eradication of microorganisms such as virus and bacteria, which will also avoid the formation of biofilm on the membrane surface. The main contributors for fouling are extracellular polymeric substances (EPS), soluble microbial products (SMP) and total dissolved salts (TDS), which leads to loss of membrane performance [5]. EPS and SMP are removed by ozonation and ASP, but TDS still remains a concern. Hence, it can be removed before membrane filtration to reduce the O&M costs. Adsorption using activated carbon (AC) is a central component of the wastewater treatment train because of its versatility, in particular for removing heavy metals and dissolved solids. In general, AC adsorbs the organic and inorganic pollutants either by physisorption or chemisorption. So, AC pores should be in the microporous region; if it is in macro- or mesoporous it will be a highway to the pollutants. The limitation in adsorption is the regeneration of adsorbent, so it can be eradicated by making it as bags like tea bags to hang over the bulk fluid. Even though, the fluid left over after adsorption also has some amount of TDS and pathogens. Researchers have already proved that membrane filtration is one the prominent processes for removing TDS and microbes effectively [6]. The treatments prior to membrane filtration alleviate the tendency of fouling by means of reducing the inorganic, organic pollutants and biofilm active pathogens. The presence of less availability of biofilm active compounds tends to lessen adhesive and cohesive nature on the facade of the membrane. So, irreversible as well as reversible fouling is substantially reduced. The reduced fouling cannot be easily identified by the experimental results. So, models can precisely describe the membrane related process that are valuable for prediction, and control of membrane related systems. Hermia models, which are very relevant to practical applications, can be very much helpful in formalizing the benefits of membrane technology [7].

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Table 1. Initial characteristics of pharmaceutical effluent

Parameters	Values	Requirement as per TNPCB norms
pH	6.5	6.5-8.5
Total dissolved solids (TDS)	1400 mg/L	200 mg/L
Total suspended solids (TSS)	290 mg/L	50 mg/L
Total solids (TS)	1690 mg/L	-
Chemical oxygen demand (COD)	1920 mg/L	<150 mg/L
Total organic carbon (TOC)	510 mg/L	<50 mg/L
Biological oxygen demand (BOD)	430 mg/L	<50 mg/L
Total alkalinity	312 mg/L	200 mg/L
Total nitrogen	0.4 mg/L	45 mg/L
Turbidity	280.4 (NTU)	-

So, the present research work focuses on the removal of organic/inorganic pollutants in sequential batch ozonated adsorptive membrane bioreactor (SBMBR) for reduced energy consumption and O&M costs. The experimental results were verified with Hermia models to characterize the fouling reduction in the process.

MATERIALS AND METHODS
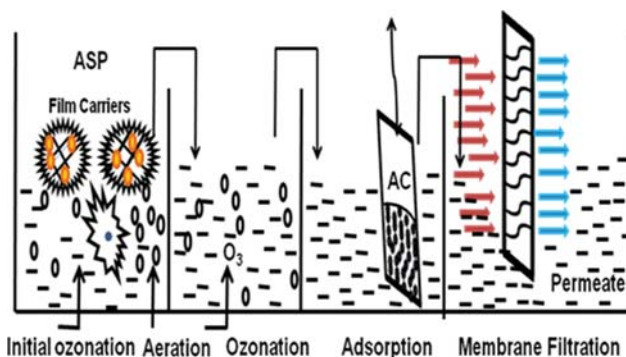
1. Treatment Sample

The synthetic pharmaceutical wastewater used in the experiments was made from waste tablets. Tamilnadu Pollution Control Board (TNPCB) standard discharge norms and the characteristics of the synthetic sample are presented in Table 1. The sludge for inoculation was taken from the secondary clarifier of the treatment plant at Pallavaram, Chennai (CETP). Activated carbon for adsorption was purchased from Sigma Aldrich. MbBRs were purchased from M/s. Lakshmi Ring Travellers Limited, Coimbatore. The typical characteristics of biofilm carriers are presented in Table 2. Cyclic voltammogram and differential pulse voltammetry studies were conducted for TDS removal using Ag/AgCl as reference electrode, glassy carbon and platinum wire as working and counter electrode, respectively. High pressure liquid chromatography (HPLC Agilent 7890B) was carried out for influent and effluent to characterize the fluid.

2. Experimental Setup

The experimental setup to study pharmaceutical wastewater involved using a sequential batch reactor, and the schematic representation is shown in Fig. 1. The reactor was equipped with suction and centrifugal pumps to pump the loaded effluent at room temperature. The reactor is equipped with a fine diffuser to provide

Table 2. Characteristics of biofilm carriers

Parameter	Value
Diameter	13 cm
Density	0.98 g/cm^3
Number of carriers used	2
Maximum filling degree	72%
Intrinsic specific surface	
Total	12.57 m^2
Effective	13.01 m^2

Fig. 1. Schematic representation of sequential batch ozonated adsorptive membrane bioreactor.

necessary oxygen for biological process and to keep the carriers moving along the reactor to provide homogeneity of the effluent. Biofilm carriers of polyethylene material (spherical shaped) with fins and grooves inside were used to obtain the maximum exposed surface area to volume. Ozone generator with the capacity 1-10 g/hr of corona discharge was used for ozonation. Conductivity and pH were measured using an HQ40d unit (Hach-Lange) in the reaction tank. Oxidation reduction potentiometer (HQ40d) was used to monitor aerobic conditions prevailing in the reactor.

3. Experimental Procedure

For the experiment, an inlet concentration ranged from 1,920-2,000 mg L^{-1} and the biocenosis of the reactor was grown from inoculated sludge and cultivated over a period of time to reach steady-state conditions. Initially, ozonation was applied for five minutes at a minimum flow rate to enhance the biodegradability index [8]. The mixed liquor suspended solids (MLSS) concentration for activated sludge process (ASP) was maintained at 6,000-9,000 mg L^{-1} throughout the process. The excess sludge was removed from the reactor, and air diffusers were fixed (35 L/min) for aeration to biological system, effective mixing of bulk solution and scouring of the membrane surface as shown in Fig. 1. The sludge process was done in 4, 8, 12 and 16 hours of sludge retention time integrated with moving bed carriers. Biodegradability index should be maintained above 0.4 for ASP and the viscosity of effluent was continuously checked during SRT studies. The changes in effluent characteristics, such as COD, BOD and TOC, found with the integration of the SMBR systems are consistent with the theoretical values towards removal of organics. Film carriers were introduced in ASP to enhance the biodegradation in which moving bed reactor gave onsite to microbes for effective degradation [9]. MbBR of polyethylene material had high protected internal active surface area which allows biofilm to adhere whether it is heterotroph or autotroph bacteria. Also, it acts as a shearing device on a coarse bubble, which in turn enhances the oxygen transfer. The effluent from the biological process was subjected to ozonation and adsorption simultaneously to remove particularly dissolved solids and harmful microbes [10]. Adsorbent quantity was initially optimized in a batch and then introduced into the bulk system where an activated carbon bag was hung with a thickness ranging from 2.5 to 3 mm. The maximum residence time allowed for ozonation in wastewater treatment is thirty minutes or else high resident ozone will

change the effluent color (yellow). One gram of high COD requires 5 to 50 grams of ozone per 1,000 liter of wastewater. Ozone concentration was optimized in a batch initially, and it was found that five grams of ozone reduced merely 75 percent of COD and TOC present in the effluent after biological integrated EO. So we split the time interval as 5 and 25 minutes in which two grams of ozone were passed initially to enhance the biodegradability and then suddenly increased to five grams of ozone to destruct the pathogenic microbes and color, after which the effluent was allowed to settle for fifteen minutes. The supernatant was allowed for microfiltration in which two polyethylene sulfone flat sheet membranes (total surface area of N x 100 cm^2) with 0.1 μm pore size were used for the filtration process. The filtrate from modules was withdrawn from suction pumps operated at constant suction pressure (SZ037DB, Yuehua Company, China).

RESULTS AND DISCUSSION

1. Optimization of Individual Process (ASP, Ozonation and Adsorption)

ASP is an eminent indicator of biological degradation systems [11]. So, ASP was carried out for various SRTs such as 4, 8, 12 and 16 hours to find the maximum survival of sludge with MLSS capacity of 6,000-9,000 mg/L. It is evident from Fig. 2(b) that the initial absorbance value was lower than the treated effluent value. This was due to the turbidity or micro pollutant present in the initial effluent, and this turbidity was decreased by prolonged treatment, which resulted in the increasing absorbance value [12-14]. It was

observed with higher absorbance value on treatment after five hours, followed by decrease in value due to the destruction of microbes resulting in more organic waste. Thus, from Fig. 2(b), we can conclude that the time taken for a biological process of five hours was sufficient for the treatment of effluent. It was reconfirmed by TDS reduction by CV analysis as shown in Fig. 2(a).

Ozonation was carried out for various flow rates like 1.5 and 3 L/m for 30 minutes, shown in Fig. 2(d). Adsorption has also been done by many researchers for the removal of antibiotics [15]. So, adsorption with AC with varying dosage in the range of 10, 15 and 17.5 g was estimated, and the optimum quantity of adsorbent for maximum removal of TOC was found to be 15 g with equilibrium time of 45 minutes as shown in Fig. 2(c). Fig. 2(c) inset shows the optimization of adsorption carried out with the adsorbent dosage of 10 g with an equilibrium time of 60 minutes. Based on the optimization, integration studies were done and checked for chances of integrity.

Fig. 3(a) and (b) shows the activated sludge before and after treatment. The sludge before treatment looks very fluffy but the sludge becomes dense after treatment, which is due to the compact size of bacteria and biopolymers also adsorb the pollutants and promote the flocculation [16]. The sludge attachment may be due to the presence of large amount of hydroxyl and carboxyl groups in the effluent because these groups generate the hydrogen bond easily. And an SEM analysis, UV and CV of sludge and effluent, reconfirms the optimization of ASP, adsorption and ozonation.

2. Choice of Integrity (ASP, Ozone & Adsorption and ASP, Adsorption & Ozone)

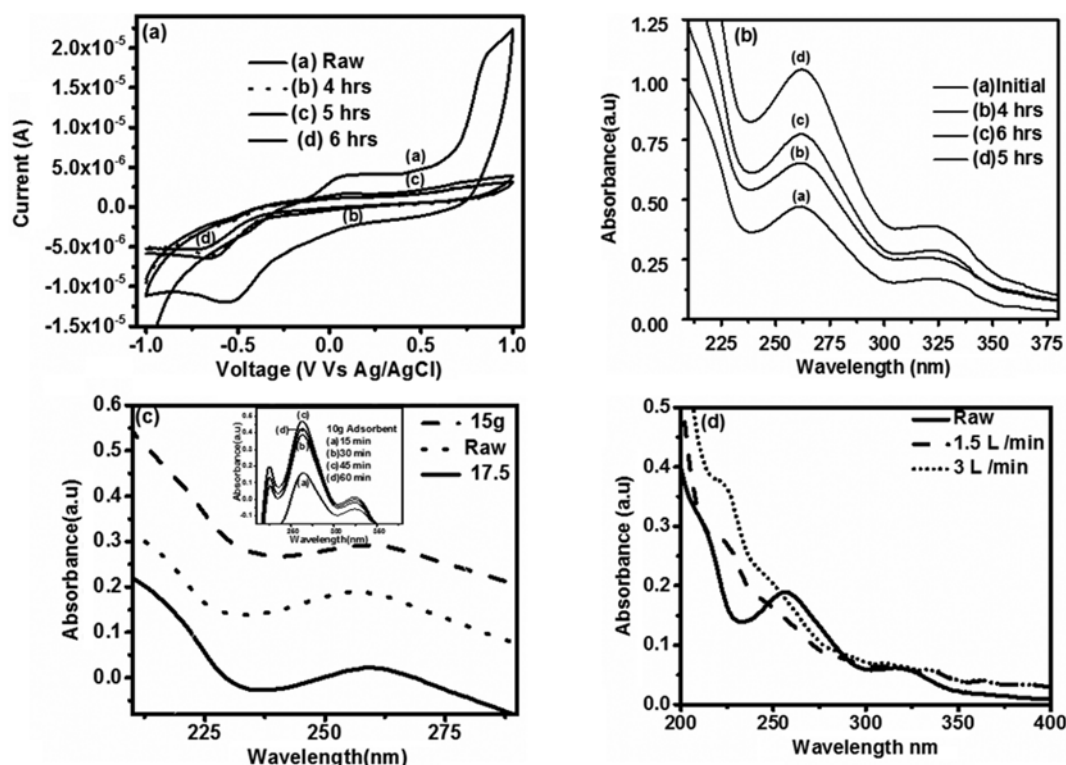The optimized time for ASP, ozone and adsorption was 5 h, 25

Fig. 2. ASP (a) CV analysis, (b) UV analysis, (c) adsorption with various dosage at 45 minutes; inset shows optimization of time at the dosage of 10 g (d) ozonation with various flow rate at 25 minutes.
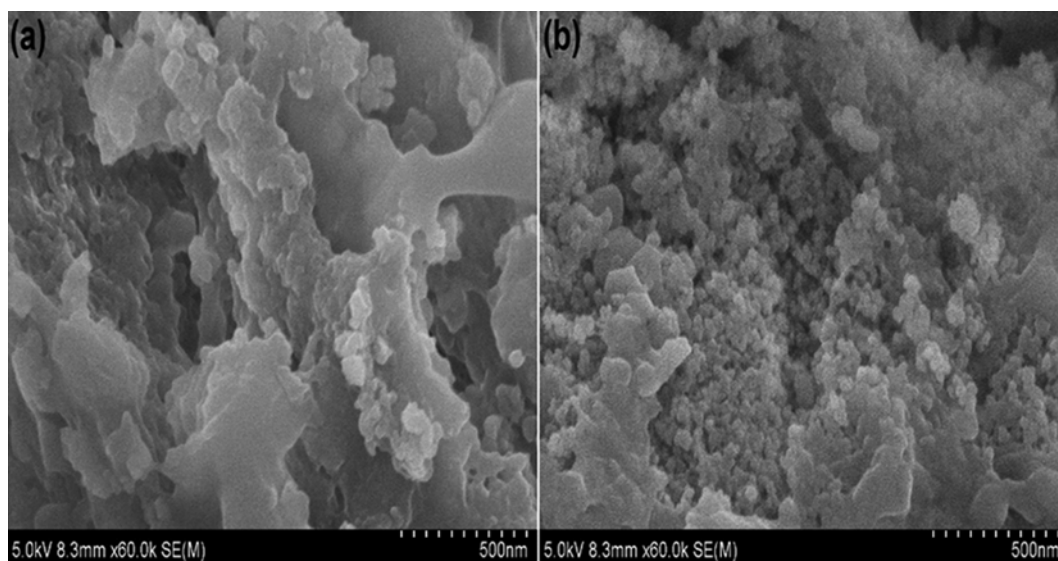
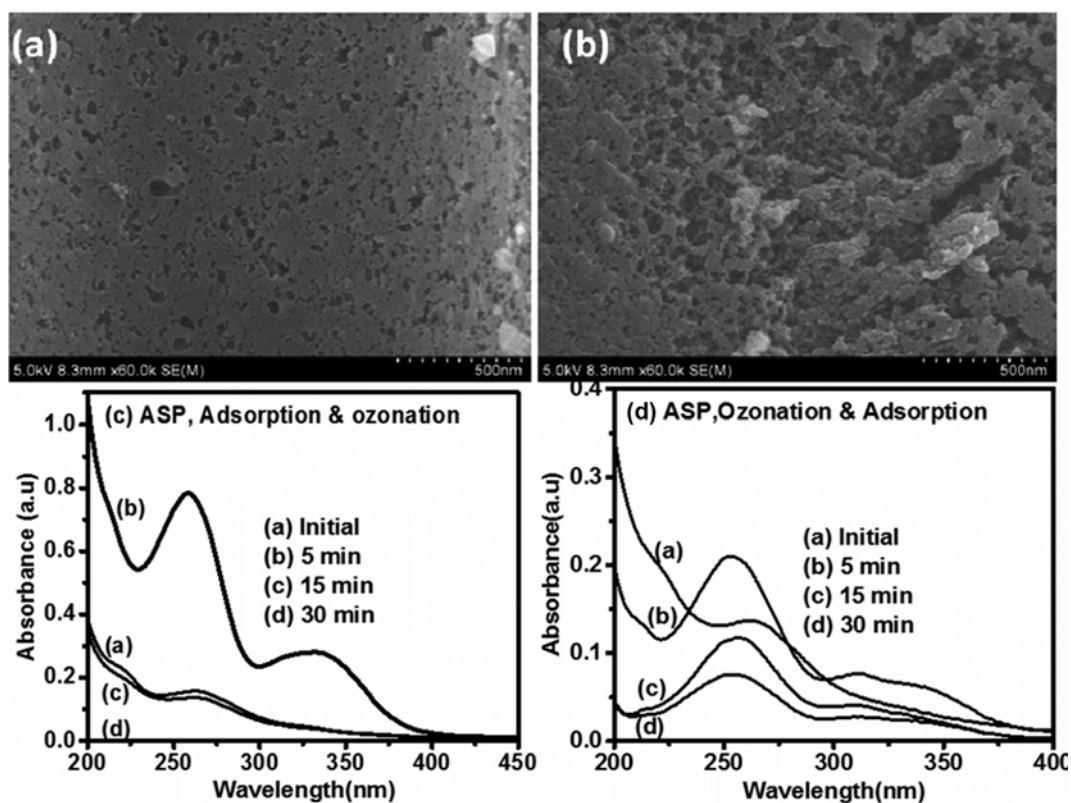**Fig. 3. SEM analysis of sludge (a) before (b) after treatment.**

Fig. 4. SEM images of (a) pure activated carbon, (b) after treatment, (c) ASP, adsorption and ozonation, (d) ASP, ozonation and adsorption.

and 45 minutes, respectively. The chances of integrity between ASP, ozonation and adsorption (Fig. 4(d)), ASP, adsorption and ozonation (Fig. 4(c)) were done based on the maximum TOC reduction with less time and energy consumption. So the time was reduced to half for ASP, and then the supernatant was allowed either to ozone or adsorption. Ozonation followed by ASP removed maximum TOC and destroyed the leftover microorganisms from ASP,

thereby eliminating the biofilm formation on the membrane facade. Hence, the finalized choice of integration was ASP, adsorption and ozonation [17]. SEM analysis of activated carbon before and after treatment is shown in Fig. 4(a) and (b). The untreated carbon had a clear surface with uniform structure, whereas the treated carbon seemed deposition of pollutants on the surface with asymmetric structure.
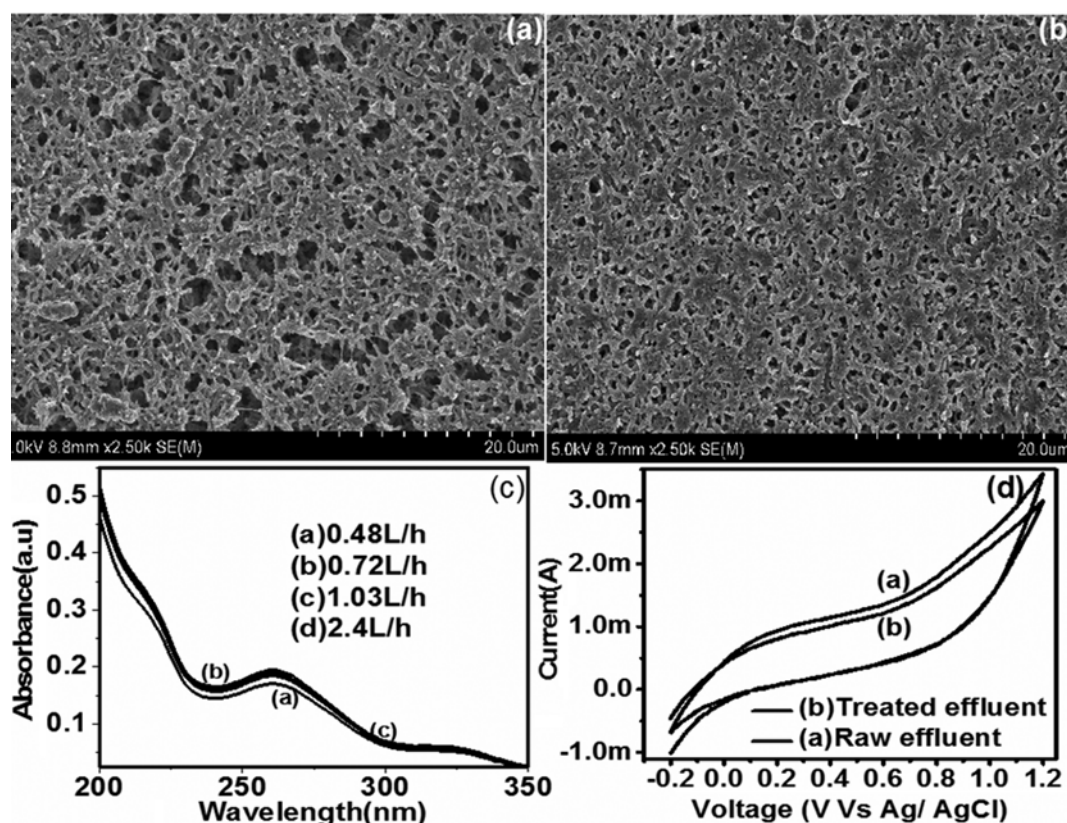
Fig. 5. SEM images of flat sheet membrane (a) before and (b) after treatment, (c) effluent treatment after microfiltration with various flow rates, (d) TDS reduction by differential pulse voltammetry analysis (DPV).

3. Permeate Flux Analysis

The supernatant from ozonation was allowed for microfiltration with constant TMP of 20 kPa for one hour to get maximum permeate flux. The permeate flux with various flow rates was carried out and analyzed for TOC and TDS reduction. The morphology of the pristine and processed membrane was found by scanning electron microscope (SEM), as shown in Fig. 5(a) and (b). The micropores were interconnected with large number of pore openings and connected pores having large void volume. The loss in macroscopic and microscopic area in a membrane was due to the adsorption of micropollutants on the facade as shown in Fig. 5(b). Microfiltration in flat sheet membrane with various flow rates was carried out and optimized 0.72 L/h due to the closer reduction percentage at 0.72 L/h at higher flow rates [18]. The pollutants will sweep off the facade at higher velocity, which in turn reduces the permeate flux drastically. This increased flow rate increases the driving force, cake compression and membrane compaction. Cake formation on the membrane surface is possibly due to the presence of micro and macropollutants in the bulk liquid [19,20].

The convective flow of solutes towards the barrier and steeper gradient of concentration in a membrane boundary layer provides an extra resistance to flow, which reduces the flux at high velocity [21]. So the optimized critical flux was considered to be 0.72 L·h. It was also reconfirmed with SEM analysis (deposition of pollutants) of the membrane. TDS is directly proportional to conductivity, i.e., amount of salts present. TOC and TDS reduction was analyzed by UV and CV as shown in Fig. 5(c) and (d). The current produced by influent and treated effluent was 4.2 mA and 2.3 mA. The percentage reduction of TDS was approximately 45 percentile. Hence, this sequential batch treatment could be an efficient treatment for all industrial effluents with various characteristics.

4. HPLC Analysis

A liquid chromatographic analysis done for untreated and treated effluent by SBMBR is shown in Fig. 6(a) and (b). Some chromatography peaks were identified from Fig. 6(a) and these peaks dis-
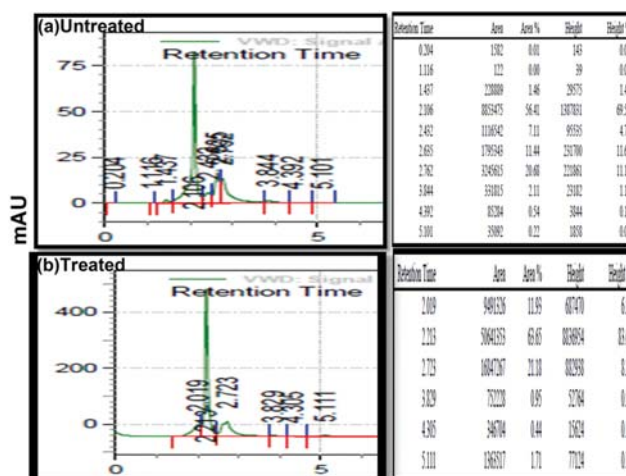

Fig. 6. HPLC analysis for (a) untreated effluent (b) treated effluent by SBMBR.

appeared in Fig. 6(b). The peaks at retention time 2.310, 2.597, 3.490, 3.965 and 5.169 minutes in which the effluent may contain 1 h-payroll 2,3-b pyridine [22], Vildagliptin [23], metalaxyl racemate [24], reserpine [25], flavonoids and vitamins. The peaks in Fig. 6(b) at retention time 2.262, 2.640 minutes may correspond to the degradation of compounds present in the untreated effluent.

5. Specific Energy Consumption

Specific energy consumption is an eminent parameter for characterizing a process in wastewater treatment. Normally, the energy consumption is high in MBR, which can be reduced by hybridizing some unit operations to meet the standard limits. The specific energy comparison between MBR and SBMBR is shown in Table 3. It reveals that the energy consumption for SBMBR was reduced to 12% when compared to MBR in three-hour operation, which may be attributed to the integration of adsorption and other AOP. It's also less than A_2O - MBR, QQ - MBR and RO followed by UV [26,27]. This could be feasible for effluent containing various pollutants and treatment plants facing various climatic conditions.

6. Hermia Model Validation

Generally, Hermia model is used to portray the fouling mechanism in membrane filtration process that includes complete blocking, standard blocking, intermediate blocking, and cake gel layer formation. The model shown in Eq. (1) is used normally for cross flow filtration.

$$\left(\frac{d^2t}{dV^2}\right)=k\left(\frac{dt}{dV}\right)^n \tag{1}$$

Depending on the fouling mechanism, n can be adopted at different values of 2, 3/2, 1 and 0 for complete blocking model, standard blocking model, intermediate blocking model and cake layer formation model, respectively. The present work is focused on constant pressure filtration, and the equations related to the above fouling mechanisms are summarized in Table 4. The decline in the flux and rise in the pressure resulting from the increase in the filtration resistance behaviors were investigated in constant rate filtration. This internal as well as external fouling automatically increases the resistance to permeate, which in turn increases the permeate flux decline. The relation between permeate flux and hydraulic resistance is given by

$$J=\frac{\Delta P}{\mu R} \tag{2}$$

The main fouling mechanism can be confirmed based on the relevant standard error by calculating the experimental data in the

Table 3. Energy consumption for three-hour operation

Process	Units	Energy consumption, KWh/m³	Performance
MBR (ASP& Microfiltration)	Aeration	0.146	Backwash needed for every 3 h with TOC<80% and TDS<30%
	Suction pump	0.023	
	Backwash (10 minutes)	0.08	
	Total	0.249	
SBMBR (ASP, MbBR, Ozone, Adsorption & Microfiltration)	Aeration	0.146	Backwash needed for every 5 h with TOC>90% and TDS<50%
	Suction pump	0.023	
	Ozone Generator	0.05	
	Backwash	No needed for 3 h	
	Total	0.219	

Table 4. Reformulated blocking mechanism by hermia model

n	Fouling mechanism	Reformulated format	Physical concept	Equation
2	Complete blocking	$\ln J=\ln J_o-[K_{cb}]t$	Pore clogging	(3)
1	Intermediate blocking	$\frac{1}{J}=\frac{1}{J_o}+[K_{ib}A]t$	Pore blocking+surface deposit	(4)
1.5	Standard blocking	$\frac{1}{J^2}=\frac{1}{J_o^2}+[0.5K_{sb}A^{0.5}J_o^{-1.5}]t$	Pore constriction	(5)
0	Cake layer formation	$\frac{1}{J^{0.5}}=\frac{1}{J_0^{0.5}}+[2K_{cl}A^2J_o^{1.5}]t$	Surface deposit formation	(6)

Table 5. Standard error values and fitted hermia model parameters

Model	Slope	Model constants	Constant values	Standard error
Complete blocking	K_{cb}	K_{cb} (s⁻¹)	8.2676E-6	0.0072
Intermediate blocking	$K_{ib}A$	K_{ib} (m⁻³)	0.6345	0.0089
Standard blocking	$0.5K_{sb}A^{0.5}J_o^{-1.5}$	K_{sb} (s⁻⁰·⁵·m⁻¹·⁵)	9.648E-3	0.00884
Cake-filtration	$2K_{cl}A^2J_o^{1.5}$	K_{cl} (s·L⁻²)	0.02212	0.00886

above equations. The internal fouling can be calculated manually in the above equations and can be found the lower closer standard error in standard, intermediate blocking and cake filtration as shown in Table 5. The complete blocking appears very poor, but the error also is much less as compared [28]. The pore fouling rate was decreased, which was due to the lower cross-flow velocity. This velocity may enable to form a scale on a facade that easily protects the primary membrane from the fouling agents, such as proteins and polysaccharides. Moreover, effluent may contain some electronegative compounds, resulting in electrostatic repulsion between organics and membrane, which can eradicate the pollutants from adhering on the facade of the membrane. This electronegative charged membrane tends to bridge with the positive ions present in the effluent via metal and metal carbonate, forming a cake gel layer. It indicates that both ASP and ozone can drastically reduce EPS and SMP.

CONCLUSION

A sequential batch membrane bioreactor (SBMBR) was developed by the combination of biological, adsorption, ozonation and microfiltration techniques. It revealed that it was better than MBR, which gave a higher reduction in TOC and TDS. SBMBR showed 12 percentile reduction in energy consumption for three-hour operation at the flow rate of 0.72 L/h (TMP 20 kPa) due to the reduced shear on barrier and fouling. The sludge deposition over the membrane surface was less, which is due to the incorporation of adsorption, biofilm carriers and ozonation. It was confirmed by SEM of carbon, membrane, UV, CV and HPLC. The energy consumption required for SBMBR also confirmed the low internal fouling via the extended backwash for four hours. We recommend that the incorporation of all physio - chemical processes to MBR will be suitable for higher organic as well as inorganic loading, which will lead to zero liquid discharge (less sludge production) and assure that the energy consumption required for the developed reactor will be reduced up to a maximum of two to three times for a month. The maintenance cost also automatically reduced. Hermia models also showed good results by standard error and constant values such that the internal fouling was reduced by incorporating all these pretreatments to membrane filtration in sequential mode. We suggest that increase in SRT, increase in adsorption capacity by doping some metal or metal oxides will reduce the organic and inorganic content drastically, which in turn will directly reduce the pore clogging, energy consumption and maintenance cost.

ACKNOWLEDGEMENT

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