

Full-scale studies on the efficiency of combined SBR-RO treatment for pharmaceutical effluents – towards sustainable ZLD (zero liquid discharge)

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ABSTRACT

With enhanced stringent rules for effluent discharges, industries are under pressure and are continuously in search of effective treatment technologies. In this process, a lot of research and development followed by pilot-scale studies are in a continuous process to achieve the treatment of required standards by researchers around the world. These efforts have presented that integrating biological and advanced physical treatment processes provides a viable solution for many industries, among which the pharmaceutical industry in one. Membrane separation processes eliminate all these techniques through selectively permeable barriers retaining a wide range of particulate and dissolved compounds while allowing passage of water and reverse osmosis (RO) that has been run proved to be effective in removing most of the parameters. Further biological treatment techniques like sequencing batch reactor (SBR) as pre-treatment for RO have proved to be viable solutions. In these lines, any industries are nowadays oriented towards recycling of treated water in the premises under mandatory zero liquid discharge (ZLD) as per the regulations. The present chapter focusses on evaluating SBR and RO treatments with SBR as pre-treatment for RO on large-scale for 3 y. Results from the studies we found that SBR-RO to be a viable and proficient combination for the treatment of pharmaceutical wastewaters. Hence, this combination was evaluated on a full scale for 3 y duration. The highest removal of total dissolved solids achieved was 98.74%, Maximum reduction of chemical oxygen demand obtained was 98.96, and Removal of ammonia was utmost with 89.57%. From the present study, it can be concluded that the treatment process has proved to be an effective treatment that has achieved ZLD.

Keywords: Reverse osmosis; Sequencing batch reactor; Zero liquid discharge; Pharmaceutical wastewater

1. Introduction

With increasing pollution, stringent legislations have been put forth to combat pollution from various sources. Industries, particularly, are burdened with these regulations and are continuously in search of treatment technologies that can effectively treat wastewater to meet the required standards. The combination or integration of biological and

advanced physical treatment processes has proved to be a viable solution to the challenges of wastewater treatment from industries, precisely for pharmaceutical industries [1].

Conventional treatment techniques include varied treatments required for removing different categories of pollutants like physical separation process for particle removal; chemical and biological processes for removal of organic matter, dissolved and suspended solids; evaporation

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techniques for volume reduction, or concentration of wastes. Membrane separation processes eliminate all these techniques through selectively permeable barriers retaining a wide range of particulate and dissolved compounds while allowing passage of water [2]. Reverse osmosis (RO) run on a trial basis proved to be effective in removing most of the parameters coming under the pollution index. Further biological treatment techniques like sequencing batch reactor (SBR) and membrane bioreactor (MBR) as pre-treatment for RO have proved to be viable solutions [3].

In these lines, all industries are nowadays oriented towards recycling of treated water in the premises under mandatory zero liquid discharge (ZLD) as per the regulations laid by local and national authorities [4,5]. The literature on ZLD for the pharmaceutical industry is limited. Few studies have reported having three treatment units constituting ZLD, which include multiple effect evaporators, agitated thin film dryer, and RO [6]. The pre/physical treatment process have been discussed in the previous papers multi-effect evaporator (MEE). The present study focusses on evaluating SBR and reverse osmosis treatments with SBR as pre-treatment for RO on large-scale for 3 y. The efficiency of the processes was evaluated in terms of reduction in total dissolved solids (TDS) and chemical oxygen demand (COD).

2. Methodology

2.1. Brief description of the treatment process

Wastewaters produced from the manufacturing process are segregated based on the concentration of TDS into two

streams, one stream with high total dissolved solids (HTDS) and the other with low total dissolved solids (LTDS). HTDS effluent stream contains all parameters in higher concentrations and hence is required to be given a high degree of treatment, while LTDS is treated along with HTDS after HTDS is partially treated (Fig. 1).

Treatment of HTDS is carried out in two stages, that is, pretreatment and biological treatment. Pretreatment includes neutralization followed by TDS reduction using MEE. HTDS condensate from MEE along with LTDS are added to anoxic tank – I for about 24 h of retention time adding return sludge from SBR. Wastewater from the anoxic tank is given biological treatment, that is, to series of SBR – I, II, III, and IV. Retention duration of 20 h is given after the effluent is decanted. Decanted effluent from SBR is fed to anoxic tank – II for removing ammoniacal nitrogen. After anoxic tank – II, the wastewater is fed to RO, that is, for further treatment. Permeate from RO is reused for different utilities while reject is again pumped to MEE for the reduction of TDS, which are again fed to the same process. Characteristics of RO are given in Table 1.

2.2. Sample collection and analytical procedures

Water samples collected from the SBR were collected daily. Samples from the SBR were collected from feed and outlet respectively from SBR, and for RO samples from the feed, permeate and reject were collected.

Wastewater samples were collected once every month with a temporal frequency of morning, afternoon, and evening, the composite sample was taken from the three

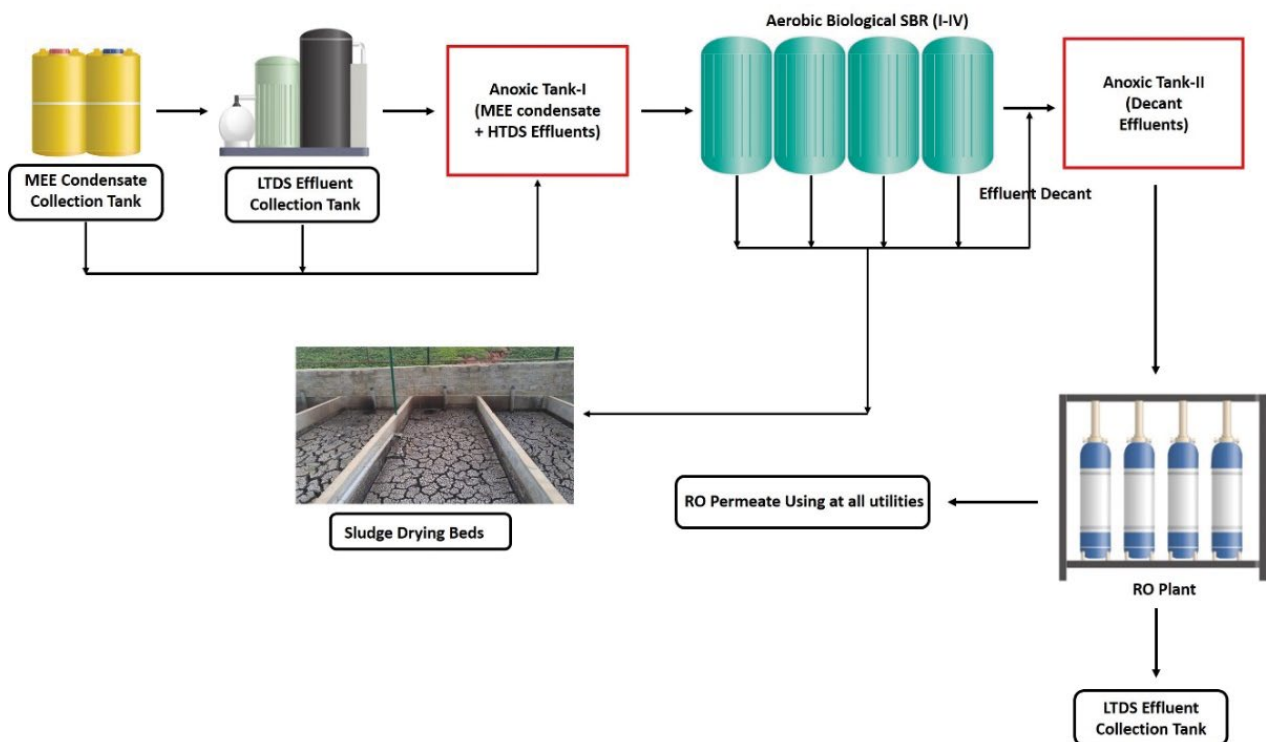


Fig. 1. Process flow chart.

samples. Samples are collected in sterile plastic containers and transferred instantly to the laboratory on the premises of the industry. The analysis was taken up immediately and completed within 48 h of duration. Samples were stored at 4°C for analysis during the following days.

Table 1
Characteristics of the RO membrane used in the study

Details	MHP-90 XXL
Membrane type	Polyamide thin-film composite
Application	Reverse osmosis
Membrane area/module	11.40 m ²
Outer diameter of module	216 ± 2 mm
Inner diameter of module	202 ± 1 mm
Length of module	1,430 mm
Length of the tie rod	1,630 mm
Operating pressure maximum	90 bar
pH operating range	3–11
Rejection as per testing conditions	>98.0%
Maximum operating temperature	40°C

Permeate flux operated in the present study is 273 L/h.

Table 2
Analytical techniques adopted in the present study

S. No	Parameter	Method of analysis
1	pH	APHA Standard Method 4500
2	Total dissolved solids	APHA Standard Method 2540C
3	Chemical oxygen demand	APHA Standard Method 5220
4	Ammoniacal nitrogen as N	APHA Standard Method 4500-NH ₃

Table 3
Treatment of effluent using SBR 2016

Month	Quantity (kl)	SBR feed		SBR outlet			
		pH	COD (mg/L)	pH	COD (mg/L)	SV 30 (ml)	DO
January	47.23	7.30	10,017.42	7.68	1,903.54	233.23	4.71
February	48.69	6.89	11,307.59	8.02	2,305.72	285.17	4.35
March	57.03	7.20	9,805.16	7.89	1,893.19	273.87	4.75
April	77.43	7.53	9,540.53	8.69	1,801.65	189.83	5.07
May	69.76	7.49	7,239.03	8.86	1,611.00	138.71	4.95
June	79.90	7.41	11,379.33	8.79	2,684.26	255.33	4.75
July	79.30	7.02	8,493.55	8.35	1,669.00	255.16	5.93
August	78.61	7.20	6,815.32	8.32	1,352.38	267.26	6.02
September	82.93	7.10	10,112.40	8.05	1,748.00	278.83	4.67
October	67.10	7.20	7,143.06	8.05	1,389.46	278.87	5.69
November	66.30	6.80	8,861.63	8.08	1,328.42	297.67	5.78
December	73.06	7.00	6,815.32	8.32	1,108.26	265.97	4.01

The following parameters for the collected samples were analyzed pH, TDS, and COD. All parameters were analyzed, as given in the section below (Table 2).

3. Results

The present study focusses on the full scale, or large-scale treatment of pharmaceutical wastewaters with the combination tested from pilot-scale studies, that is, SBR-RO for successive 3 y from 2016 to 2018. The combination in pilot-scale has proved to be efficient with SBR removing COD biologically and RO removing TDS, resulting in effective removal of pollutant load from the wastewaters.

Table 3 presents the data on biological treatment using SBR during the year 2016. Since SBR is active for biological degradation, the parameters considered for analysis of treatment efficiency are COD removal and surrogate parameters for successful degradation sludge volume at 30 min settlement (SV 30) and dissolved oxygen (DO).

pH values in SBR feed ranged from 6.8 to 7.53; the least value of pH was reported in November with 6.8, and the highest being 7.53 was witnessed in April. pH values of SBR outlet were observed to increase during all the months, the least value of pH 7.68 was noted in January, and the highest pH 8.86 was observed in May.

Concentrations of COD in SBR feed were observed to range from 6,815.32 to 11,379.33 mg/L, and the least and highest COD values were documented during August, December, and May respectively. Successful degradation of COD resulted in lower COD concentrations in the SBR outlet, the lowest COD was observed during December with 1,108.26 mg/L, and the highest COD was noted during June with 2,684.26 mg/L. Sludge volume at 30 min settlement recorded the lowest value during May with 138.71 ml, and the highest values were observed in November with 297.67 ml. DO values ranged from 4.35 to 6.01 mg/L, the lowest was documented in February, and the highest was recorded in December. The highest COD reduction was noted with 85.01% in November, and the lowest removal with 76.41% was obtained in June.

The percentage removal of the four focused parameters from RO is presented in Table 4. The highest reduction in TDS was obtained in August (97.89%) and lowest in November (95.68%). While the highest of 98.06% reduction was obtained for COD in March, the lowest was registered in August with 95.55%. Ammonia removal was utmost with 88.7% in November and least with 79% in August (Fig. 2).

Overall the parameter that was removed utmost was hardness with 98.70% followed by COD with 98.06%, then TDS with 97.89% and ammonia with 88.7%. The order of removal of parameters during the year 2016 by treatment with RO with SBR as pre-treatment is as follows:

Total dissolved solids > Chemical oxygen demand > Ammonia.

Table 5 portrays the reduction of COD in percentage with the combination of SBR and RO during the year 2016. The minimum percentage of reduction was observed in December with 91.01%, and the maximum reduction was observed in June with 95.44%, respectively.

Table 6 portrays the stage-wise reduction of COD each after SBR and after RO during 2016. The reduction of COD

after SBR was observed as 76.41% in June, which was minimum while the maximum reduction was observed in November with 85.01%, respectively. On the other hand, RO treatment has resulted in the highest reduction of COD all year, which was >99% (Fig. 3).

Table 7 presents the biological treatment using SBR during the year 2017. pH values in SBR feed ranged from 7.21 to 7.95, the least value of pH was reported in March, and the highest was observed in September. pH values of SBR outlet were observed to increase during all the months, the lowest pH 8.00 was noted in May, and the highest pH 8.67 was observed in February.

Concentrations of COD in SBR feed were observed to range from 6,820.48 to 11,308.87 mg/L; the least and highest COD values were documented during July and March, respectively. Successful degradation of COD resulted in lower COD concentrations in the SBR outlet, the lowest COD was observed during January with 1,109.86 mg/L, and the highest COD was noted during December with 2,460.65 mg/L.

Table 4
Percentage reduction of TDS, COD, NH₃-N and hardness by RO, 2016

Month	TDS	COD	NH ₃ -N
March	96.17	98.06	82.07
April	95.90	97.32	87.48
May	96.40	97.88	86.14
June	96.06	97.19	83.45
July	95.79	96.63	87.55
August	97.89	95.55	79.00
September	97.65	97.80	85.48
October	97.07	96.83	83.97
November	95.68	97.43	88.70
December	97.73	97.43	86.48

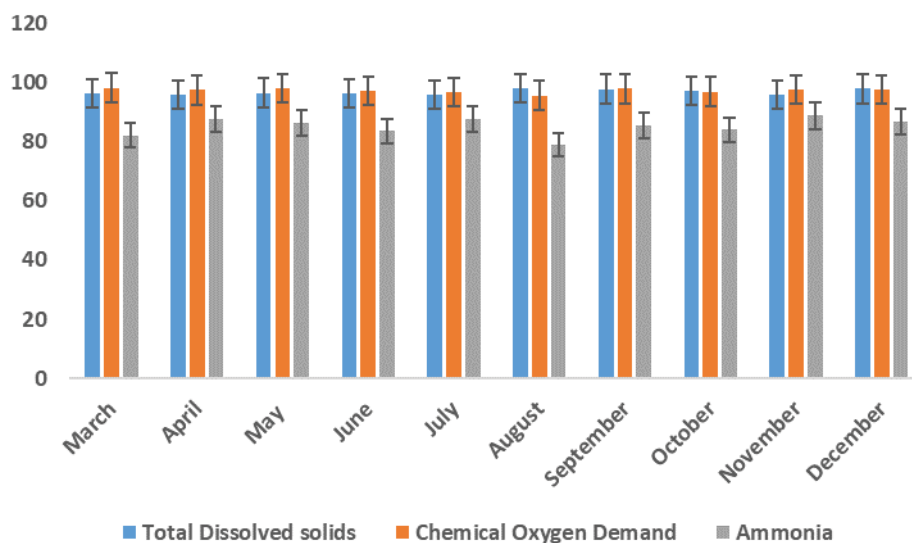


Fig. 2. Percentage reduction of TDS, COD, and NH₃-N by RO, 2016.

Sludge volume at 30 min settlement recorded the lowest value during March with 200.32 ml, and the highest values were observed in September with 345.33 ml. The percentage reduction of COD from SBR is depicted in Table 8. The maximum COD reduction was noted with 85.5% in April, and the lowest removal with 80.64% was obtained in July.

The percentage removal of the three focused parameters is presented in Table 8. The highest reduction in TDS was obtained in September (97.16%) and lowest in April (94.86%). While the highest of 98.96% reduction was obtained for COD in February, the lowest was registered in October with 91.57%. Ammonia removal was utmost with 85.69% in December and least with 78.45% in August. Overall the parameter that was removed utmost was ammonia with 85.69% followed by COD with 98.96%, then TDS with 97.46% (Fig. 4). The order of removal of parameters during the year 2017 by treatment with RO with SBR as pre-treatment is as follows:

Chemical oxygen demand > Total dissolved solids > Ammonia

Table 5
Percentage reduction of COD treated by RO with SBR as pre-treatment 2016

Month	% Reduction
March	94.67
April	93.86
May	94.63
June	95.44
July	93.73
August	91.90
September	94.34
October	94.36
November	93.34
December	91.01

Table 9 depicts the percentage reduction of COD with the combination of SBR and RO during the year 2017. The minimum percentage of reduction was observed in December with 94.51%, and the maximum reduction was observed in August with 97.45%, respectively.

Table 10 shows the stage-wise reduction of COD each after SBR and after RO during 2017. The reduction of COD after SBR was observed as 80.64% in July, which was minimum while the greatest reduction was observed in April with 85.58%, respectively. On the other hand, RO treatment has resulted in the highest reduction of COD, which was 97.45% as the highest and least was 94.51% (Fig. 5).

Table 11 presents the biological treatment using SBR during the year 2018. pH values in SBR feed ranged from 7.22 to 7.90, the lowest pH value was reported in August, and the highest being 7.53 was perceived in March. pH values of SBR outlet were observed to increase during all the months, the lowest pH of 8.05 was noted in February, and the highest pH of 8.93 was observed in April.

Table 6
Stage wise reduction of COD from SBR to RO 2016

Month	SBR	RO
March	80.69	99.18
April	81.12	99.15
May	77.75	98.93
June	76.41	99.33
July	80.35	99.05
August	80.16	98.82
September	82.71	99.18
October	80.55	98.87
November	85.01	99.04
December	83.74	98.77

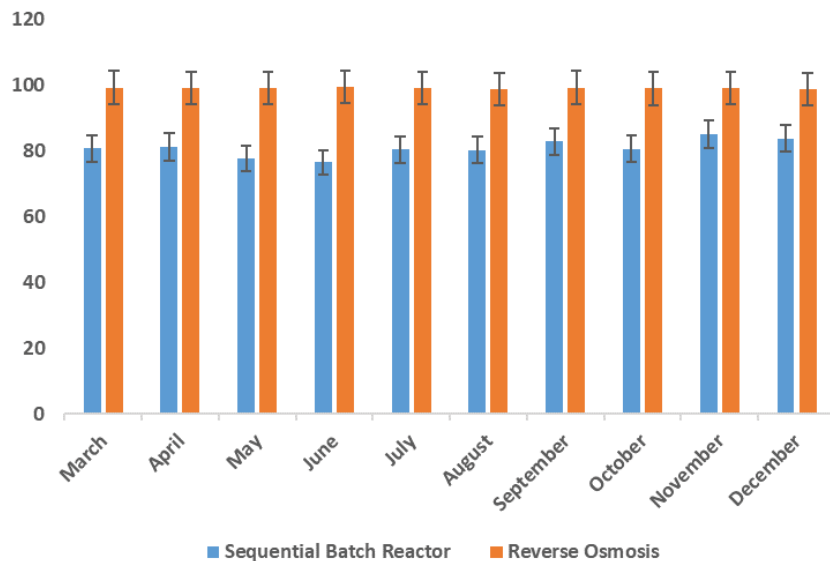


Fig. 3. Stage wise reduction of COD from SBR to RO 2016.

Table 7
Treatment of pharmaceutical effluent using SBR 2017

Month	Quantity (KL)	Feed		Sequential batch reactor (SBR)			
		pH	COD (mg/L)	pH	COD (mg/L)	SV 30 (ml)	DO
January	58.29	7.37	7,399.06	8.32	1,109.86	267.26	5.02
February	56.07	7.53	9,733.43	8.67	1,490.89	275.00	5.08
March	62.19	7.21	11,308.87	8.19	1,735.00	200.32	5.05
April	62.97	7.81	9,084.67	8.26	1,310.34	225.67	4.18
May	54.61	7.70	7,041.81	8.00	1,353.48	303.55	5.44
June	58.93	7.74	8,866.73	8.04	1,420.65	233.00	4.04
July	66.16	7.75	6,820.48	8.24	1,320.21	314.84	4.17
August	70.65	7.64	7,002.26	8.37	1,160.00	254.52	4.02
September	62.67	7.95	8,111.13	8.45	3,118.67	345.33	4.56
October	71.35	7.66	9,005.03	8.26	1,673.55	326.77	3.54
November	58.93	7.80	9,034.67	8.30	2,324.33	311.33	3.68
December	57.87	7.88	9,626.13	8.28	2,460.65	335.16	3.98

Table 8
Percentage reduction of TDS, COD and NH₃-N RO 2017

Month	TDS	COD	NH ₃ -N
January	96.84	98.24	86.54
February	97.14	98.96	98.45
March	95.70	97.82	100.00
April	94.86	97.96	76.40
May	95.30	98.12	87.50
June	96.23	97.46	89.98
July	96.60	96.81	98.60
August	97.00	96.62	100.00
September	97.16	95.98	100.00
October	97.46	91.57	98.89
November	96.46	93.48	83.18
December	97.01	95.74	93.59

Concentrations of COD in SBR Feed were observed to range from 8,989.03 to 11,803.23 mg/L; the least and peak COD values were recorded during December and May, respectively. Successful degradation of COD resulted in lower COD concentrations in the SBR outlet, the lowest COD was observed during December with 1,011 mg/L, and the highest COD was noted during May with 1,639 mg/L.

Sludge volume at 30 min settlement recorded the least value in June with 211.67 ml, and the highest values were observed in October with 281.29 ml. The percentage reduction of COD from SBR is presented in Table 12. The peak COD reduction was noted with 89.72% in November, and the lowest removal with 86.11% was obtained in May (Table 12).

Overall the parameter that was removed utmost was COD with 98.06% followed by TDS with 98.74% and ammonia with 89.57% (Fig. 6 and Table 13). The order of removal of parameters during the year 2018. By treatment with RO with SBR as pre-treatment is as follows:

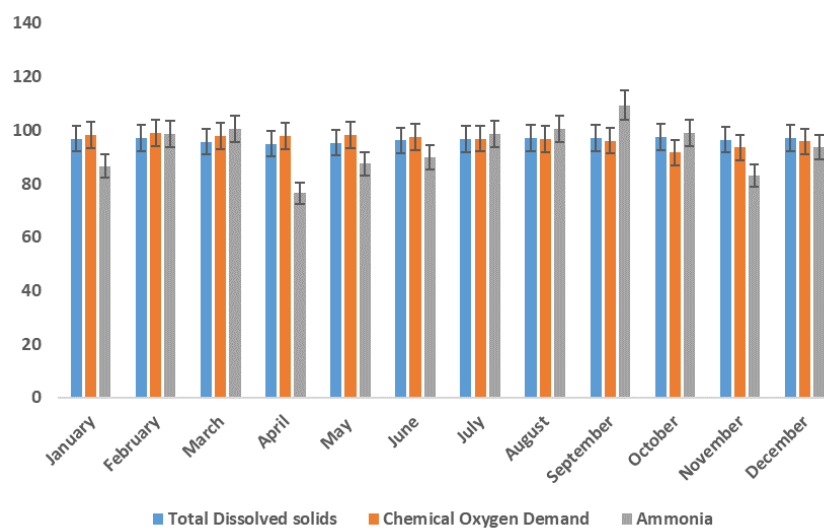


Fig. 4. Percentage reduction of TDS, COD and NH₃-N RO 2017.

Total dissolved solids > Chemical oxygen demand > Ammonia.

Table 14 illustrates the percentage removal of COD with the combination of SBR and RO during the year 2018. The minimum percentage of reduction was observed in May with 86.11%, and the maximum reduction was observed in November with 89.72%, respectively.

Table 15 illustrates the stage-wise reduction of COD each after SBR and after RO during 2018. The reduction of COD after SBR was observed as 89.72% in November, which was minimum while the highest reduction was perceived in May with 86.11%, respectively. On the other hand, RO treatment has resulted in the highest reduction of COD was 99.74%, and the least was 99.41% in July (Fig. 7).

Table 16 illustrates the percentage reduction of TDS, COD, and Ammonia during the 3 y of the study, that is, 2016, 2017, and 2018, respectively.

Among the 3 y, the highest removal of TDS was achieved during 2018 with 98.74% during November. The maximum reduction of COD was obtained in 2016 with 98.96% in February in the year 2017. Removal of ammonia to the

utmost was in the year 2018, with 89.57% during November (Figs. 8–10).

4. Discussion

The efficiency of SBR as pre-treatment to RO at full scale was studied from the year 2016 to 2018. From 2016 to 2018, an increase in the efficiency of treatment was observed. The highest removal of TDS was achieved with 98.74%. A maximum reduction of COD was obtained with 98.96%. Removal of ammonia to the utmost was with 89.57%. The importance of biological treatment lies in its chemical conversion ability of pollutants to end products.

In contrast to chemical oxidation methods, aerobic methods are proficient in mineralizing bulky organic molecules quantitatively converting them to end products of CO₂, H₂O, and inorganic nitrogen compounds. In this process, the diversity of materials is given from the biomass in the reactor that is referred to as extracellular polymeric substances, which contribute to fouling in membrane processes [7]. It is understood that biological treatment not

Table 9 Reduction of COD treated by RO with SBR as pre-treatment 2017

Month	% Reduction
January	97.40
February	96.49
March	94.83
April	96.79
May	96.62
June	96.59
July	96.25
August	97.45
September	96.31
October	96.81
November	95.65
December	94.51

Table 10 Stage wise reduction of COD from SBR to RO 2017

Month	SBR	RO
January	85.00	97.40
February	84.68	96.49
March	84.66	94.83
April	85.58	96.79
May	80.78	96.62
June	83.98	96.59
July	80.64	96.25
August	83.43	97.45
September	81.07	96.31
October	83.58	96.81
November	84.23	95.65
December	84.30	94.51

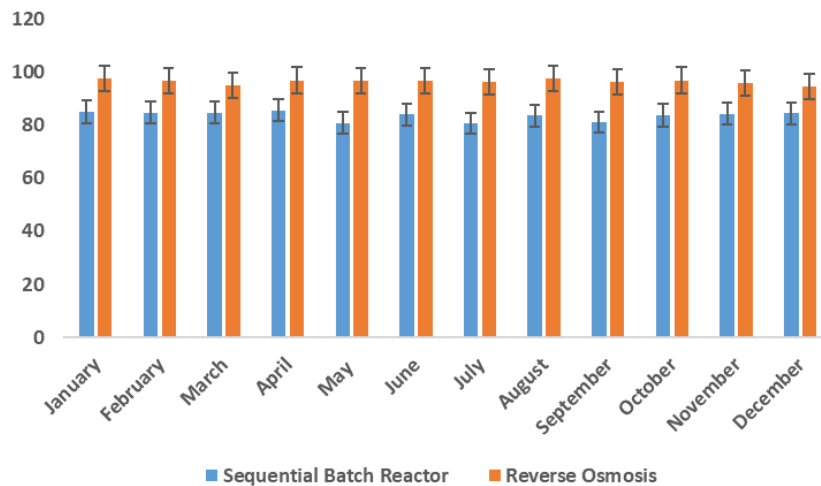


Fig. 5. Stage wise reduction of COD from SBR to RO 2017.

Table 11
Treatment of pharmaceutical effluent using SBR 2018

Month	Feed			SBR			
	Quantity (KL)	pH	COD (mg/L)	pH	COD (mg/L)	SV 30 (ml)	DO
January	100	7.53	10,558.06	8.13	1,432.00	229.00	4.76
February	68	7.65	10,150.00	8.05	1,310.00	227.50	4.86
March	54	7.90	10,906.77	8.40	1,490.00	244.19	3.90
April	83	7.74	9,788.67	8.93	1,298.00	241.67	3.78
May	84	7.58	11,803.23	8.76	1,639.00	241.61	3.85
June	88	7.46	9,678.67	8.55	1,096.00	211.67	4.74
July	117	7.39	9,012.90	8.46	1,183.00	252.58	3.57
August	121	7.22	10,198.71	8.15	1,247.00	257.42	4.15
September	119	7.52	9,297.67	8.22	1,197.00	276.00	5.90
October	98	7.23	9,911.29	8.31	1,128.00	281.29	4.84
November	105	7.58	11,176.00	8.73	1,149.00	231.67	4.20
December	98	7.30	8,989.03	7.50	1,011.00	275.00	4.78

Table 12
Percentage reduction of COD SBR 2018

Month	% Reduction
January	86.44
February	87.09
March	86.34
April	86.74
May	86.11
June	88.68
July	86.87
August	87.77
September	87.13
October	88.62
November	89.72
December	88.75

Table 13
Percentage reduction of TDS, COD and NH₃-N RO 2018

Month	TDS	COD	NH ₃ -N
January	97.02	96.65	83.00
February	96.30	97.07	84.48
March	97.51	97.22	79.74
April	97.11	98.06	77.61
May	97.22	96.53	79.81
June	96.70	96.11	81.48
July	98.23	95.48	82.34
August	97.55	97.22	81.51
September	98.48	98.00	82.26
October	98.60	96.59	86.25
November	98.74	96.10	89.57
December	98.15	97.27	89.28

Table 14
Percentage reduction of COD treated by RO with SBR as pre-treatment 2018

Month	% Reduction
January	86.44
February	87.09
March	86.34
April	86.74
May	86.11
June	88.68
July	86.87
August	87.77
September	87.13
October	88.62
November	89.72
December	88.75

Table 15
Stage wise reduction of COD from SBR to RO 2018

Month	SBR	RO
January	86.44	99.55
February	87.09	99.62
March	86.34	99.62
April	86.74	99.74
May	86.11	99.52
June	88.68	99.56
July	86.87	99.41
August	87.77	99.66
September	87.13	99.74
October	88.62	99.61
November	89.72	99.60
December	88.75	99.69

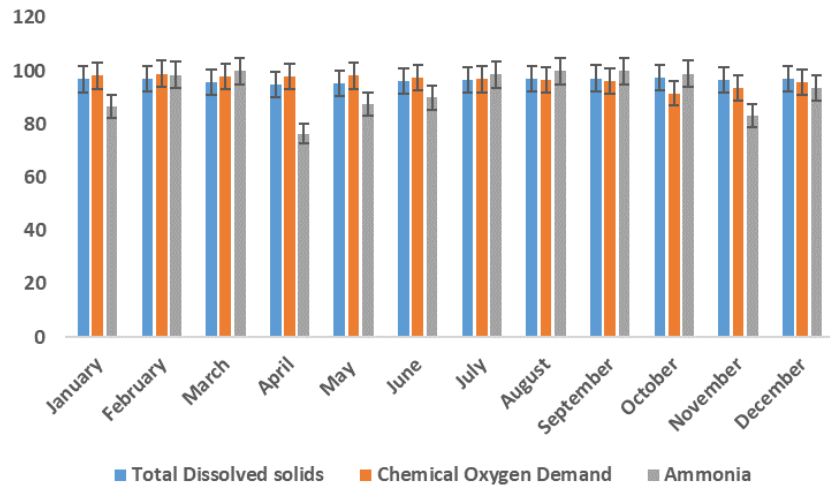


Fig. 6. Percentage reduction of TDS, COD and NH₃-N RO 2018.

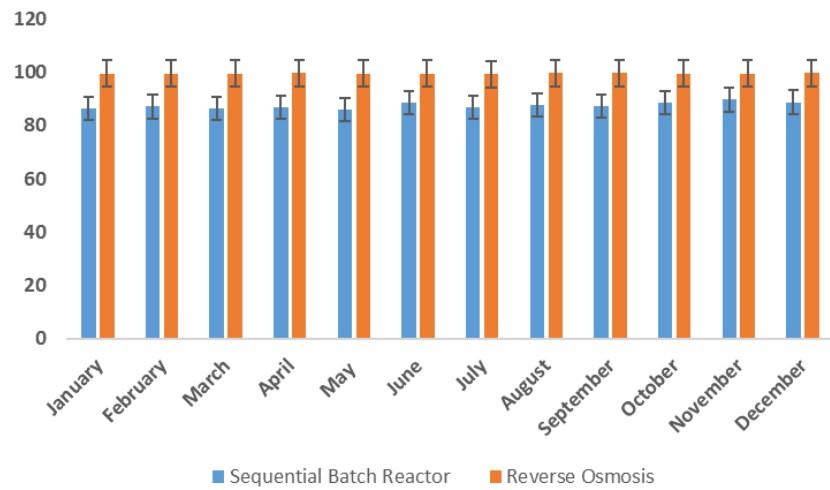


Fig. 7. Stage wise reduction of COD from SBR to RO 2018.

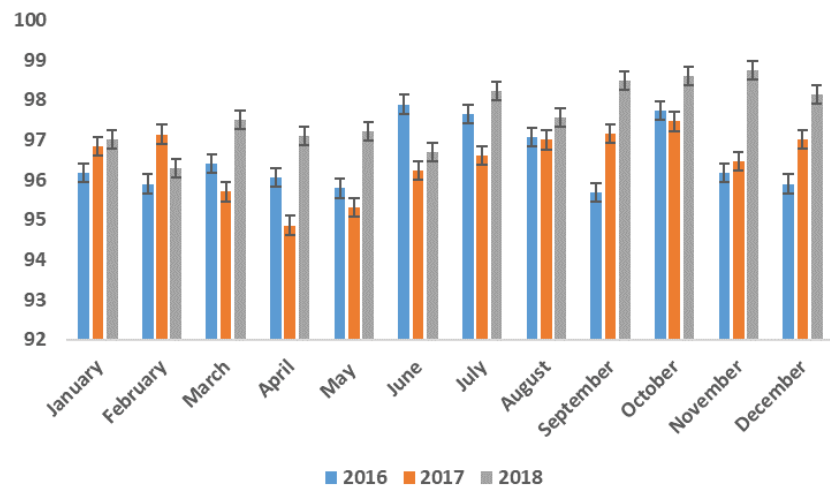


Fig. 8. TDS removal trends in the 3 y.

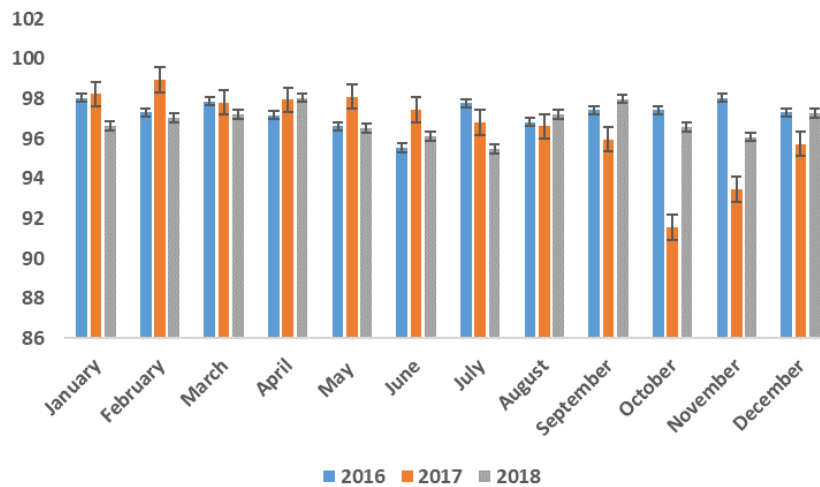


Fig. 9. COD removal trends in the 3 y.

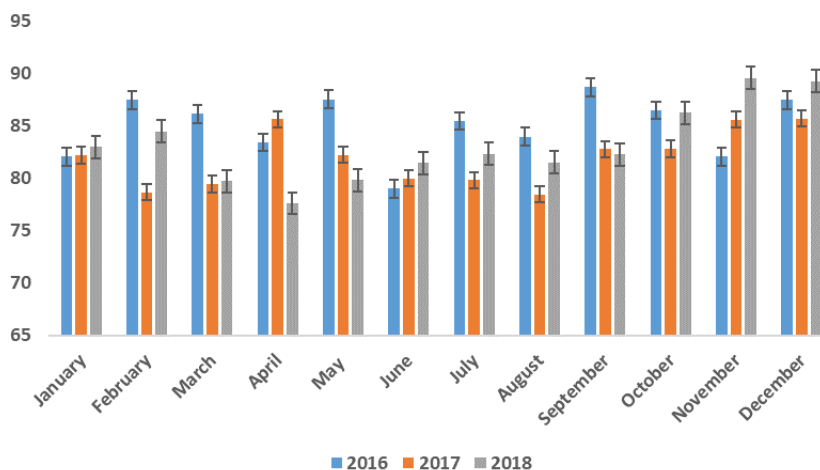


Fig. 10. NH₃-N removal trends in the 3 y.

only improves RO performance but also has advantages like low energy requirements [8]. Further, complete removal of organic, biological, and colloidal substances in pre-treatment or RO feed water is considered as key for the successful and efficient working of the RO plant [9].

Especially for the treatment of pharmaceutical wastewaters, several membrane technologies like Nanofiltration, ultrafiltration, electrodialysis, RO, and their combinations were studied both in the pilot and full scale [10–12]. Inadequate studies are reported on the use of RO for pharmaceutical wastewater treatment. Individually RO with different configurations presented effective elimination of 36 endocrine-disrupting chemicals and personal care products, which included lipid regulators, antibiotics, oral contraceptives, hormones, analgesics, and antiseptics [13].

RO was also efficient in treating high strength wastewater, which requires careful focus to treat up to required standards. Further, high strength wastewaters with complex composition exert pressure on treatment units resulting in high costs for maintenance and inefficient treatment of the

wastewaters. For example, leachate is categorized as high strength wastewater. To produce water, fit for reuse, the leachate is initially treated using a conventional wastewater treatment plant, which was subsequently treated using a SBR followed by RO. Further, RO yield is evaporated for additional concentration. They stated that the treatment process applied was effective in treating leachate, with RO effectively reducing the volume of the leachate. They have also quoted that the measured approach was eco-friendly [14]. Another high strength wastewater is of swine wastewater. Zhang et al. [15] evaluated the treatment of swine wastewater with the following treatment units in the sequence as follows: one anaerobic SBR followed by two SBRs followed by sludge settling tank and sand filter. Finally, the effluent from the sand filter was fed to the RO unit. After treatment, the reduction in COD and solids were in the order 89% and 97%, respectively.

Precisely SBRs performed biological treatment while the sand filter was used as a pre-treatment for RO. Such treated water when fed to RO, it achieved extremely effective

Table 16
TDS, COD and NH₃-N removal trends in the 3 y

Month	TDS			COD			NH ₃ -N		
	2016	2017	2018	2016	2017	2018	2016	2017	2018
January	96.17	96.84	97.02	98.06	98.24	96.65	82.07	82.20	83.00
February	95.90	97.14	96.30	97.32	98.96	97.07	87.48	78.67	84.48
March	96.40	95.70	97.51	97.88	97.82	97.22	86.14	79.46	79.74
April	96.06	94.86	97.11	97.19	97.96	98.06	83.45	85.66	77.61
May	95.79	95.30	97.22	96.63	98.12	96.53	87.55	82.23	79.81
June	97.89	96.23	96.70	95.55	97.46	96.11	79.00	79.98	81.48
July	97.65	96.60	98.23	97.80	96.81	95.48	85.48	79.83	82.34
August	97.07	97.00	97.55	96.83	96.62	97.22	83.97	78.45	81.51
September	95.68	97.16	98.48	97.43	95.98	98.00	88.70	82.77	82.26
October	97.73	97.46	98.60	97.43	91.57	96.59	86.48	82.83	86.25
November	96.17	96.46	98.74	98.06	93.48	96.10	82.07	85.60	89.57
December	95.90	97.01	98.15	97.32	95.74	97.27	87.48	85.69	89.28

separation of nutrients and salts from water. Coking wastewater was treated for removal of COD and TN using a combined sequencing batch MBR-RO system, which resulted in 93 and 96% removal of COD and total nitrogen, respectively [16]. When aerobic SBR was combined with the processes of photo-Fenton followed by RO in reclaiming textile industry wastewater, the treated water was found to be best suitable for reuse internally for various industrial processes [17].

Some studies have applied the combination of biological and membrane treatment in a different sequence of operation processes, as in the study carried out by Shi et al. [18], where they have first applied membrane separation followed by biological treatment. The retentate from RO was treated in SBR. Their results report optimum removal of total nitrogen, which was below the effluent quality standards. Thus, they suggest this sequence of the treatment process to be applicable for wastewater treatment plants. A similar sequence was tested by Madikizela et al. [19], where SBR was used for biological denitrification of RO concentrate containing high conductivity that was obtained from a coking wastewater plant.

High nitrate (93.1%) removal was achieved in the SBR, suggesting effecting biological denitrification. Further, the sequence of SBR followed by RO was used to treat landfill leachate. The study stated that alone biological treatment would not be efficient in the removal of bio-refractory organic substances. Still, when these wastes were treated using RO, this has effectively eliminated bio-refractory organic substances. Hence, they concluded RO was polishing to achieving that good quality effluents after biological treatment is an effective treatment [20].

With the pressure to meet environmental regulation in a challenging way and necessity for the eco-friendly approach, the need to treat wastewaters up to the mark has become mandatory. To achieve this, the strategy of ZLD has been adopted by many industries, especially pharmaceutical industries. ZLD is understood to be an ideal state of the complete closed-loop cycle, in which discharge of any form of liquid effluent is eliminated.

Most of the pharmaceutical industries adopt SBR-RO as one of the effective treatments to achieve ZLD. Parikh et al. [21], after several trial and error runs, have installed the RO pilot plant and then have proposed the following sequence of treatment processes to achieve ZLD. Initially, process water possessing high TDS is fed to the multi-effect evaporator, the shell condensate from MEE will be fed to RO. Permeate from RO is fed to the boiler, and RO rejects given to the Effluent Treatment Plant (ETP) plant along with water from other process plants. After ETP treatment, the water is sent to a sand filter, then to a cartridge filter, and again fed to RO. Permeate now coming from RO is reused while reject is again fed to MEE.

Another study in the similar lines was carried out by Kumar et al. [22] in a moderate scale active pharmaceutical ingredients manufacturing industry. They categorized process wastewaters to HCS and LCS (high concentration streams and low concentration streams). The proposed treatment for HCS is solvent stripper followed by MEE and agitated thin film drier (AFTD). Further condensates from MEE and AFTD after neutralization are fed to SBR and are added along with LCS. For LCS, the treatment sequence is SBR, followed by RO and polishing reverse osmosis. A pilot plant with this sequence resulted in pronounced removal of total suspended solids, TDS, biochemical oxygen demand, and COD in the order of 100%, 99.2%, 100%, and 99.9%, respectively. Most of the previous studies and reviews state that upon the requirement of further advanced treatment in terms of reducing wastewater discharge along with pollution load, then RO process will be a successful and promising treatment option after specific biological treatment [23–25] as SBR in the present case.

5. Conclusions

Results from the studies we found that SBR-RO to be a viable and proficient combination for the treatment of pharmaceutical wastewaters. Hence, this combination was evaluated on a full scale for 3 y duration.

- Highest removal of TDS achieved was 98.74%.
- Maximum reduction of COD obtained was 98.96%.
- Removal of ammonia was utmost with 89.57%.

The present study depicted the efficiency of various treatment units in treating pharmaceutical wastewaters. Multiple effect evaporators were efficient in removing solids from high TDS waste streams. The SBR was efficient over MBR for treating pharmaceutical wastewaters. Hybrid technologies of combining SBR and RO showed promising results.

From the present study, it can be concluded that the flow scheme of effluents presented below has proved to be an effective treatment that has achieved ZLD. Process water having high TDS is fed to the multiple-effect evaporator; the condensate along with low TDS stream will be treated using a SBR and then fed to RO to obtain polished water having high potential for reuse. The reject from RO is again fed to multiple-effect evaporators. The concentrate from the multiple-effect evaporator is dried in an agitated thin film dryer, which is sent for suitable disposal or further treatment.

References

- [1] Y.H. Zhao, J. Le, M.H. Abraham, A. Hersey, P.J. Eddershaw, C.N. Luscombe, D. Butina, G. Beck, B. Sherborne, I. Cooper, J.A. Platts, Evaluation of human intestinal absorption data and subsequent derivation of a quantitative structure-activity relationship (QSAR) with the Abraham descriptors, *J. Pharm. Sci.*, 90 (2001) 749–784.
- [2] Y.-Y. Choi, S.-R. Baek, J.-I. Kim, J.-W. Choi, J. Hur, T.-U. Lee, C.-J. Park, B.J. Lee, Characteristics and biodegradability of wastewater organic matter in municipal wastewater treatment plants collecting domestic wastewater and industrial discharge, *Water*, 9 (2017) 409.
- [3] E. Sahar, R. Messalem, H. Cikurel, A. Aharoni, A. Brenner, M. Godehardt, M. Jekel, M. Ernst, Fate of antibiotics in activated sludge followed by ultrafiltration (CAS-UF) and in a membrane bioreactor (MBR), *Water Res.*, 45 (2011) 4827–4836.
- [4] M. Srinivasarao, C.V. Galliford, P.S. Low, Principles in the design of ligand-targeted cancer therapeutics and imaging agents, *Nat. Rev. Drug Discovery*, 14 (2015) 203.
- [5] C. Bernard, S.A. Chandrakanth, I.S. Cornell, J. Dalton, A. Evans, B.M. Garcia, C. Godin, M. Godlewski, G.H. Jansen, A. Kabani, S. Louahlia, L. Manning, R. Maung, L. Moore, J. Philley, J. Slatnik, J. Srigley, A. Thibault, D.D. Picard, H. Cracower, B. Tetu, Guidelines from the Canadian Association of Pathologists for establishing a telepathology service for anatomic pathology using whole-slide imaging, *J. Pathol. Inf.*, 5 (2014) 1–31.
- [6] K.J. Shah, S.-Y. Pan, A.D. Shukla, D.O. Shah, P.-C. Chiang, Mechanism of organic pollutants sorption from aqueous solution by cationic tunable organoclays, *J. Colloid Interface Sci.*, 529 (2018) 90–99.
- [7] R. Reif, A. Santos, S.J. Judd, J.M. Lema, F. Omil, Occurrence and fate of pharmaceutical and personal care products in a sewage treatment works, *J. Environ. Monit.*, 13 (2011) 137–144.
- [8] M.M. Crowley, F. Zhang, M.A. Repka, S. Thumma, S.B. Upadhye, S. Kumar Battu, J.W. McGinity, C. Martin, Pharmaceutical applications of hot-melt extrusion: part I, *Drug Dev. Ind. Pharm.*, 33 (2007) 909–926.
- [9] D.E.B. Isaias, R. Niero, V.F. Noldin, F. de Campos-Buzzi, R.A. Yunes, F. Delle-Monache, V. Cechinel-Filho, Pharmacological and phytochemical investigations of different parts of *Calophyllum brasiliense* (Clusiaceae), *Pharm.-Int. J. Pharm. Sci.*, 59 (2004) 879–881.
- [10] C. Bellona, J.E. Drewes, Viability of a low-pressure nanofilter in treating recycled water for water reuse applications: a pilot-scale study, *Water Res.*, 41 (2007) 3948–3958.
- [11] G.T. Snyder, A. Hiruta, R. Matsumoto, G.R. Dickens, H. Tomaru, R. Takeuchi, J. Komatsubara, Y. Ishida, H. Yu, Pore water profiles and authigenic mineralization in shallow marine sediments above the methane-charged system on Umitaka Spur, Japan Sea, *Deep Sea Res. Part II*, 54 (2007) 1216–1239.
- [12] A.J. Watkinson, E.J. Murby, D.W. Kolpin, S.D. Costanzo, The occurrence of antibiotics in an urban watershed: from wastewater to drinking water, *Sci. Total Environ.*, 407 (2009) 2711–2723.
- [13] A.M. Deegan, B. Shaik, K. Nolan, K. Urell, M. Oelgemöller, J. Tobin, A. Morrissey, Treatment options for wastewater effluents from pharmaceutical companies, *Int. J. Environ. Sci. Technol.*, 8 (2011) 649–666.
- [14] L. Miao, G.Q. Yang, T. Tao, Y.Z. Peng, Recent advances in nitrogen removal from landfill leachate using biological treatments – a review, *J. Environ.*, 235 (2019) 178–185.
- [15] Y.Z. Zhang, L. Duan, B. Wang, Y.L. Du, G. Cagnetta, J. Huang, L. Blaney, G. Yu, Wastewater-based epidemiology in Beijing, China: prevalence of antibiotic use in flu season and association of pharmaceuticals and personal care products with socioeconomic characteristics, *Environ. Int.*, 125 (2019) 152–160.
- [16] J.L. Wang, S.Z. Wang, Removal of pharmaceuticals and personal care products (PPCPs) from wastewater: a review, *J. Environ.*, 182 (2016) 620–640.
- [17] G. Blanco, A. Junza, D. Barrón, Occurrence of veterinary pharmaceuticals in golden eagle nestlings: unnoticed scavenging on livestock carcasses and other potential exposure routes, *Sci. Total Environ.*, 586 (2017) 355–361.
- [18] Y.H. Shi, J.H. Huang, G.M. Zeng, Y.L. Gu, Y.N. Chen, Y. Hu, B. Tang, J.X. Zhou, Y. Yang, L.X. Shi, Exploiting extracellular polymeric substances (EPS) controlling strategies for performance enhancement of biological wastewater treatments: an overview, *Chemosphere*, 180 (2017) 396–411.
- [19] L.M. Madikizela, S. Ncube, L. Chimuka, Uptake of pharmaceuticals by plants grown under hydroponic conditions and natural occurring plant species: a review, *Sci. Total Environ.*, 636 (2018) 477–486.
- [20] S.R.S. Reddy, M.K. Karnena, S. Yalakala, V. Saritha, Biological treatability of low total dissolved solids (LTDS) using SBR as a pre-treatment for reverse osmosis, *J. Water Resour. Prot.*, 12 (2020) 135–154.
- [21] K.J. Parikh, K.K. Sawant, Solubilization of vardenafil HCl in lipid-based formulations enhances its oral bioavailability in vivo: a comparative study using Tween-20 and Cremophor-EL, *J. Mol. Liq.*, 277 (2019) 189–199.
- [22] G.R. Kumar, B.S. Chandrashekar, M.S. Rao, M. Ravindra, K.T. Chandrashekar, V. Soundararajan, Pharmaceutical importance, physico-chemical analysis and utilisation of Indian sandalwood (*Santalum album* Linn.) seed oil, *J. Pharmacogn. Phytochem.*, 8 (2019) 2587–2592.
- [23] S.R. Reddy, V. Saritha, M.K. Karnena, B.K. Dwarapureddi, Combined SBR and RO pilot scale treatment for pharmaceutical wastewater, *Desal. Water Treat.*, 98 (2017) 45–51.
- [24] S.R.S. Reddy, M.K. Karnena, V. Saritha, Pilot scale biological treatment as pre-treatment for reverse osmosis, *J. Water Resour. Prot.*, 10 (2019) 1369–1388.
- [25] S. Vara, M. Konni, M.K. Karnena, Membrane Technology for Treatment of Pharmaceutical Wastewaters: A Novel Approach, A.C. Affam, E.H. Ezechi, Eds., Handbook of Research on Resource Management for Pollution and Waste Treatment, IGI Global Publisher, Hershey, Pennsylvania, USA, 2020, pp. 502–530.