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Pilot Scale Biological Treatment as Pre-Treatment for Reverse Osmosis

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Abstract

Treatment of pharmaceutical wastewaters is a challenging task owing to their complexity and pollution load, variability in strength of waste streams accompanied with shock loads. Since no single treatment system is a viable option, integration of existing systems with advanced physical/chemical processes has been gaining attention for treatment of pharmaceutical wastewater. In the present study, two biological treatment methods were evaluated for their efficiency as pre-treatment system for RO which are sequencing batch reactor and membrane bioreactor. Efficiency of biological treatments tested SBR and MBR was presented in terms of percentage removal of physico-chemical parameters. Total dissolved solids removal by SBR was 31.82% while MBR showed 29.25% reduction. Chemical oxygen demand removal by SBR was 69.54% while MBR showed 30.35% removal. Efficiency of combined treatments SBR-RO and MBR-RO was presented in terms of removal of total dissolved solids, COD and ammonia. TDS removal was the highest in the combination of SBR-RO with 95.94% removal, while MBR-RO combination resulted in 87.29% removal. Chemical oxygen demand was achieved maximum with the combination of MBR-RO 92.33% while competitive results were achieved with the combination SBR-RO also with 88.62% removal. Removal of ammonia was maximum with the combination SBR-RO 87.5%, while competitive results were obtained with MBR-RO 85.51%. From the results, it can be understood that SBR was efficient in removing ammonia, total dissolved solids and was equally competent in removing chemical oxygen demand. This study concludes that combined treatment of SBR-RO proves to be promising in treating pharmaceutical wastewaters.

Keywords

Reverse Osmosis, Membrane Reactor, Sequencing Batch Reactor, Waste Water

1. Introduction

Treatment of pharmaceutical wastewaters is a challenging task owing to their complexity and pollution load, variability in strength of waste streams with shock loads. These factors govern the effectiveness of conventional treatment systems. Since no single treatment system is a viable option, integration of existing systems with advanced physical/chemical processes has been gaining attention for treatment of pharmaceutical wastewater.

Simplicity in process and single tank design of SBR coupled with flexible technology has been recognized as a desirable treatment option for different types of wastewater [1]-[6]. Unique characteristics of SBR include its competence in removal both organic matter and nitrogen from wastewater [7] [8] [9]. Conditions of both aerobic and anaerobic can be adopted in SBR for removal of phosphorus [10] [11] [12]. These properties of SBR make it viable economically [13].

SBR applied for treatment of olive mill wastewaters showed significant removal of pollution load through 60% removal of total polyphenols and 90% removal of chemical oxygen demand respectively [14]. When used to treat swine wastewater the reactor presented excellent purification of wastewaters. Removal of total phosphorus, total nitrogen ammonia-nitrogen and COD were in the order of 96.2%, 95.6%, 95.7%, and 98.2% respectively [15]. SBR treatment was also adopted for pharmaceutical wastewaters, though less work has been reported the results were promising. Study of wastewater containing antibiotic was carried out by Elmolla and Chaudhuri, 2011 [16].

Modern hybrid wastewater treatment adopting membrane separation technology along with biological process is Membrane bio-reactor (MBR) [17]. Utilized as biochemical engineering processes, MBR can be used as suspended growth bioreactor functioning processes like bio-oxidation, fermentation, nitrification and denitrification along with separation of solid and liquid [18]. Further, this can also be operated at high sludge concentrations [19]. MBRs are well appreciated for their efficiency in removing dissolved solids [20] [21]. Major concern of MBR system is fouling of membrane which indicates accumulation of rejected constituents on the membrane resulting in resistance to water transport through the membrane [22]. Greater than 80% removal efficiency for pharmaceuticals was reported by Radjenovic *et al.*, 2007 [23].

Membrane technologies have gained significant importance in treating wide array of industrial wastewaters during the past decades. Among others, reverse osmosis (RO) has been specifically appreciated as state of art in treatment of wastewater. RO has been adopted for treating industrial wastewaters from petrochemical, chemical, pulp and paper, food industries, electrochemical, municipal wastewater, textile and also for treatment of drinking water [24] [25] [26]. Permeate from this process has high quality with potential to reuse for various processes like dyeing, boilers cooling-towers and cleaning, etc.

However, application of RO faces major hurdle with reference to membrane fouling which impacts efficiency of treatment both qualitatively and quantita-

tively, increasing operation costs due to resistance in filtration, production of corrosive by-products and passage of salts [27] [28]. Owing to fouling reason extensive pre-treatment is required for successful functioning of these systems [29] [30]. Comparative study between conventional biological treatment and MBR as pre-treatment options for RO was studied and revealed that MBR was more suitable [31]. Current research focusses on testing the efficiency of biological treatments SBR and MBR as viable pre-treatment options for RO.

2. Methodology

2.1. Process

Biological treatment using sequencing batch reactor and membrane bioreactor was studied. Efficiency of the process was evaluated in terms of reduction in total dissolved solids and chemical oxygen demand. Treated waters from each of the unit are fed to reverse osmosis for a duration of one month to evaluate their efficiency as pre-treatment to reverse osmosis (**Figure 1**).

2.2. Sampling Site and Sample Collection

All the samples were collected from the treatment plant of the pharmaceutical industry. Water samples from sequencing batch reactor and membrane bioreactor were collected once every day. Samples from sequencing batch reactor were collected from feed and outlet respectively and for membrane bioreactor and reverse osmosis samples from feed, permeate and reject were collected.

2.3. Analytical Procedures

The following parameters for the collected samples were analysed pH, total dissolved solids and chemical oxygen demand. All parameters were analysed as per the standard methods of APHA, 2015 (Table 1).

3. Results

This research presents the results from pilot scale studies focussed on to study the most suitable pre-treatment for reverse osmosis (RO) treatment. Sequencing batch reactor (SBR) and Membrane bioreactor (MBR) were assessed for their suitability as pre-treatment options to RO.

Table 2 illustrates one month (June, 2016) data of SBR tested for its suitability

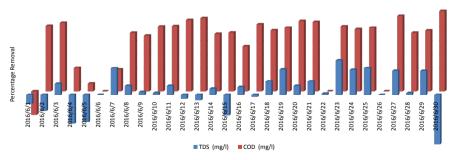


Figure 1. Percentage removal of TDS and COD in SBR pilot trails during June, 2016.

Table 1. APHA methods (2005).

S.No	Parameter	Method of Analysis
1.	рН	APHA Standard Method 4500
2.	Total Dissolved Solids	APHA Standard Method 2540C
3.	Chemical Oxygen Demand	APHA Standard Method 5220

Table 2. Removal of TDS and COD in the SBR pilot trail.

Date	TDS (mg/l)	COD (mg/l)	Date	TDS (mg/l)	COD (mg/l)
01-06-2016	-8.41	-21.88	16-06-2016	7.20	41.51
02-06-2016	-14.95	60.61	17-06-2016	-1.90	61.95
03-06-2016	10.16	63.33	18-06-2016	12.50	56.32
04-06-2016	-26.67	21.64	19-06-2016	23.36	58.84
05-06-2016	-25.00	6.98	20-06-2016	8.33	64.96
06-06-2016	0.00	0.00	21-06-2016	12.50	64.08
07-06-2016	24.66	20.00	22-06-2016	1.27	0.00
08-06-2016	8.33	54.06	23-06-2016	31.82	60.08
09-06-2016	2.78	51.53	24-06-2016	23.08	57.37
10-06-2016	1.90	59.97	25-06-2016	24.82	58.84
11-06-2016	8.33	60.37	26-06-2016	0.00	0.00
12-06-2016	-3.70	65.86	27-06-2016	22.22	69.54
13-06-2016	-4.89	67.55	28-06-2016	1.82	54.08
14-06-2016	5.77	53.13	29-06-2016	22.14	56.12
15-06-2016	-18.93	54.19	30-06-2016	-45.58	74.35

as preliminary treatment for Reverse Osmosis process. One month data on a daily basis was presented. Efficiency of the process was tested with reduction in parameters total dissolved solids and chemical oxygen demand. Further as surrogate for removal of TDS and COD, sludge volume and dissolved oxygen respectively were also recorded. The highest and lowest TDS reduction was recorded as 31.82% and -45.58% on 23rd and 30th June respectively. Reduction in COD was observed to be good in comparison with reduction of TDS the highest and the lowest reduction percentages were recorded as 24.82% and -45.58% on 25th and 30th June respectively. In line with the poor removal of TDS sludge volume has recorded the lowest values ranging from 40 - 280 ml. Dissolved oxygen values ranged between 4.4 to 6.1 mg/l (Figure 1).

Table 3 presents one month trail run of Reverse Osmosis treatment, which was fed with SBR outlet. Percentage removal of TDS from RO treatment was noted at the highest and lowest as 96% and 78.18% respectively. Maximum and minimum percentage of COD removal was 88.62% and 64.62% respectively.

Percentage reduction of ammonia with maximum and minimum was 87.50% and 78.00% respectively (Figure 2).

Table 3. Efficiency of RO with SBR as pre-treatment with reference to reduction in TDS and COD.

	Date	TDS	COD	Ammonia	Date	TDS	COD	Ammonia
	01-06-2016	92.82	64.62	82.17	17-06-2016	87.90	85.70	78.84
	02-06-2016	90.26	74.87	80.23	18-06-2016	87.85	87.72	84.34
	03-06-2016	78.18	78.18	82.61	19-06-2016	87.14	86.22	80.90
	04-06-2016	95.29	81.33	82.55	20-06-2016	85.24	87.23	0.00
	05-06-2016	95.94	82.25	0.00	21-06-2016	87.27	84.52	80.20
	07-06-2016	94.39	0.00	87.50	22-06-2016	84.76	83.06	82.53
	08-06-2016	94.29	74.76	84.86	23-06-2016	86.41	83.40	78.25
	09-06-2016	92.33	74.33	84.26	24-06-2016	81.03	86.67	81.88
	10-06-2016	96.00	75.07	83.39	25-06-2016	88.00	83.21	80.09
	11-06-2016	94.69	76.49	84.62	26-06-2016	84.95	0.00	78.00
	12-06-2016	94.52	88.62	0.00	27-06-2016	0	85.11	79.19
	13-06-2016	93.98	84.88	80.99	28-06-2016	80.48	84.22	81.90
	14-06-2016	94.08	85.51	82.05	29-06-2016	82.22	83.81	-
	15-06-2016	92.94	82.75	81.57	30-06-2016	-	82.06	-
	16-06-2016	-	88.19	79.91	-	-	-	-
-								

Efficiency of Reverse Osmosis with SBR as pre-treatment

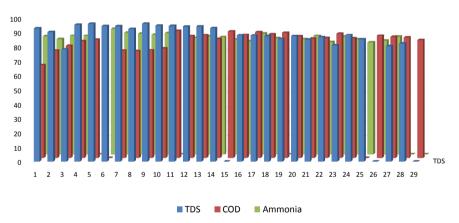


Figure 2. Percentage removal of TDS, COD and ammonia in RO with SBR as pre-treatment pilot trails during June, 2016.

Table 4 presents the biological treatment of pharmaceutical wastewaters using membrane bioreactor (MBR) on pilot trail during the month of August 2016. Efficiency of the treatment is defined in terms of reduction in strength of wastewaters with parameters of total dissolved solids, total solids, chemical oxygen demand and turbidity. The percentage removal of the parameters under test, as understood from the permeate values the TDS removal was recorded to be 1.38% as the lowest and 29.25% as the highest. While TSS has showed 100% removal, the COD removal varied from 4.77% to 30.35%. Turbidity removal was good with 80.90% being the lowest and 99.86% recorded as the highest (Figure 3).

Table 4. Efficiency of MBR treatment presented in terms of percentage removal.

Day	TDS	TSS	COD	Turbidity	Day	TDS	TSS	COD	Turbidity
1	1.38	100	4.77	99.66	16	13.78	100	6.28	99.72
2	3.36	100	11.15	99.50	17	20.27	100	5.08	99.73
3	2.47	100	26.64	99.05	18	12.41	100	8.16	99.44
4	29.25	100	16.00	99.77	19	14.81	100	17.28	99.59
5	6.23	100	19.06	99.79	20	11.62	100	12.65	99.79
6	7.82	100	11.17	99.81	21	10.66	100	26.41	99.33
7	6.09	100	30.35	80.90	22	5.74	100	22.36	99.74
8	16.79	100	11.76	98.95	23	5.98	100	20.09	99.71
9	1.38	100	4.77	99.66	24	9.22	100	10.70	99.79
10	10.82	100	11.36	99.82	25	12.02	100	26.28	99.49
11	11.38	100	5.94	99.68	26	7.30	100	14.42	99.65
12	13.65	100	9.07	99.63	27	10.07	100	16.08	99.51
13	13.95	100	10.10	99.72	28	6.16	100	8.34	99.83
14	9.59	100	15.42	99.73	29	13.30	100	13.65	99.86
15	14.17	100	13.95	99.55	30	8.52	100	9.78	99.57

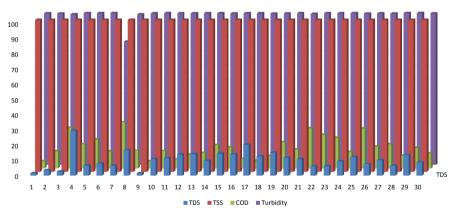


Figure 3. Efficiency of MBR treatment.

Table 5 presents treatment with reverse osmosis with membrane bioreactor as pre-treatment. Feed waters contained high concentrations of all parameters under test. The percentage removal of the parameters under test as understood from the permeate values the TDS removal was recorded to be 76.96% as the lowest and 87.29% as the highest. COD removal varied from 73.03% to 92.33%. While hardness showed 93.33% - 100% removal, ammonia removal was good with 71.46% being the lowest and 85.51% recorded as the highest (Figure 4).

Table 6 illustrates the comparison of total dissolved solids and chemical oxygen demand removal by sequencing batch reactor and membrane bioreactor. Maximum reduction of total dissolved solids was obtained by SBR with 31.82% while minimum was also obtained by SBR with -74.58%. On the other hand,

Table 5. Efficiency of RO with MBR as pre-treatment with reference to TDS and COD Reduction.

Day	TDS	COD	NH ₃	Hardness	Day	TDS	COD	NH ₃	Hardness
1	76.96	83.23	79.63	100.00	14	79.57	76.90	83.93	98.26
2	85.88	83.64	77.81	100.00	15	80.00	81.29	82.85	97.50
3	87.29	82.22	78.46	100.00	16	81.70	76.09	80.28	94.29
4	85.19	84.91	80.71	100.00	17	83.75	82.18	0.00	95.86
5	84.23	85.11	81.51	00.00	18	83.93	85.28	0.00	0.00
6	80.95	88.17	81.88	100.00	19	78.79	86.77	71.46	97.56
7	84.60	86.31	84.29	100.00	20	78.95	88.55	81.25	96.67
8	80.84	86.91	84.48	100.00	21	79.29	89.47	74.29	96.36
9	82.85	84.97	83.75	95.19	22	86.92	90.12	76.25	97.32
10	84.03	86.91	80.27	96.19	23	83.48	89.19	77.69	98.23
11	77.80	73.03	82.35	98.26	24	86.50	90.26	79.00	96.92
12	85.00	79.29	84.29	97.27	25	84.94	91.69	81.97	95.45
13	82.55	77.65	85.51	97.14	26	86.67	92.33	80.15	93.33

Table 6. Comparison of TDS and COD removal after treatment with SBR and MBR.

D	TD	S	СО	D	D	TD	TDS		COD	
Day -	SBR	MBR	SBR	MBR	Day	SBR	MBR	SBR	MBR	
1	-8.41	1.38	-21.88	4.77		7.2	13.78	41.51	6.28	
2	-14.95	3.36	60.61	11.15	17	-1.9	20.27	61.95	5.08	
3	10.16	2.47	63.33	26.64	18	12.5	12.41	56.32	8.16	
4	-26.67	29.25	21.64	16	19	23.36	14.81	58.84	17.28	
5	-25	6.23	6.98	19.06	20	8.33	11.62	64.96	12.65	
6	0	7.82	0	11.17	21	12.5	10.66	64.08	26.41	
7	24.66	6.09	20	30.35	22	-74.58	5.74	0	22.36	
8	8.33	16.79	54.06	11.76	23	31.82	5.98	60.08	20.09	
9	2.78	1.38	51.53	4.77	24	23.08	9.22	57.37	10.7	
10	1.9	10.82	59.97	11.36	25	24.82	12.02	58.84	26.28	
11	8.33	11.38	60.37	5.94	26	0	7.3	0	14.42	
12	-3.7	13.65	65.86	9.07	27	22.22	10.07	69.54	16.08	
13	-4.89	13.95	67.55	10.1	28	1.82	6.16	54.08	8.34	
14	5.77	9.59	53.13	15.42	29	22.14	13.3	56.12	13.65	
15	-18.93	14.17	54.19	13.95	30	-45.58	8.52	74.35	9.78	

MBR present lowest removals with 1.38% and 29.25% as the lowest and the highest. The average removal for TDS was the highest with MBR (10.33%) since it was stable without going to negative values. SBR, even though presented the

highest removal was quite fluctuating in treatment process. Chemical oxygen demand was also the highest (69.54%) with SBR and the lowest also by SBR with –21.88%, while MBR was stable with minimum of 4.77% and maximum of 30.35% removal of COD. The average removal of COD was the highest with SBR with 42.51% and MBR being only 13.96% (Figure 5 and Figure 6).

Comparison of treatment efficiency of reverse osmosis with SBR and MBR as pre-treatment in terms of TDS, COD and Ammonia removal is presented in **Table** 7. Total dissolved solids removal was the highest in the combination of SBR-RO

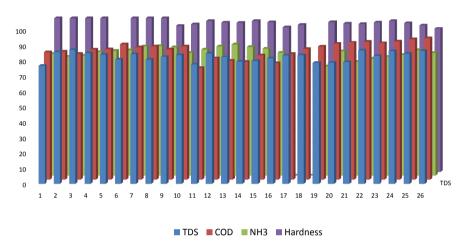


Figure 4. Efficiency of RO with MBR as pre-treatment.

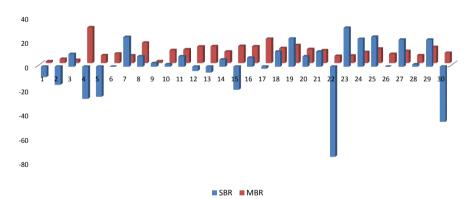


Figure 5. Comparison of TDS removal with SBR and MBR.

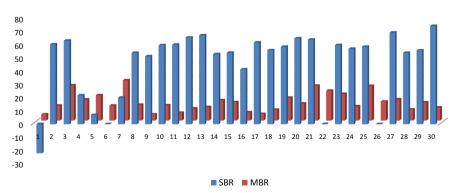


Figure 6. Comparison of COD removal after treatment with SBR and MBR.

Table 7. Comparison of TDS, COD and ammonia removal after treatment with RO using SBR and MBR as pre-treatment.

	TI	OS	CC)D	Amn	Ammonia		
Day -	S-RO	M-RO	S-RO	M-RO	S-RO	M-RO		
1	92.82	76.96	64.62	83.23	82.17	79.63		
2	90.26	85.88	74.87	83.64	80.23	77.81		
3	78.18	87.29	78.18	82.22	82.61	78.46		
4	95.29	85.19	81.33	84.91	82.55	80.71		
5	95.94	84.23	82.25	85.11	0	81.51		
6	94.39	80.95	0	88.17	87.5	81.88		
7	94.29	84.60	74.76	86.31	84.86	84.29		
8	92.33	80.84	74.33	86.91	84.26	84.48		
9	96.00	82.85	75.07	84.97	83.39	83.75		
10	94.69	84.03	76.49	86.91	84.62	80.27		
11	94.52	77.80	88.62	73.03	0	82.35		
12	93.98	85.00	84.88	79.29	80.99	84.29		
13	94.08	82.55	85.51	77.65	82.05	85.51		
14	92.94	79.57	82.75	76.90	81.57	83.93		
15	-	80.00	88.19	81.29	79.91	82.85		
16	87.9	81.70	85.7	76.09	78.84	80.28		
17	87.85	83.75	87.72	82.18	84.34	0.00		
18	87.14	83.93	86.22	85.28	80.9	0.00		
19	85.24	78.79	87.23	86.77	0	71.46		
20	87.27	78.95	84.52	88.55	80.2	81.25		
21	84.76	79.29	83.06	89.47	82.53	74.29		
22	86.41	86.92	83.4	90.12	78.25	76.25		
23	81.03	83.48	86.67	89.19	81.88	77.69		
24	88.00	86.50	83.21	90.26	80.09	79.00		
25	84.95	84.94	0	91.69	78.00	81.97		
26	0	86.67	85.11	92.33	79.19	80.15		
Avg	86.41	82.79	75.56	84.71	72.34	74.39		

 $S-RO-RO\ treatment\ with\ SBR\ as\ pre-treatment;\ M-RO-RO\ treatment\ with\ MBR\ as\ pre-treatment.$

treatment with 95.94% removal. While MBR-RO combination resulted in 87.29% removal (**Figure 7**). Chemical oxygen demand was achieved maximum with the combination of MBR-RO 92.33% while competitive results were achieved with the combination SBR-RO also 88.62% (**Figure 8**). Removal of ammonia was maximum with the combination SBR-RO (87.5%), while competitive results were obtained with MBR-RO (85.51%) (**Figure 9**).

From the results, it can be understood that individually SBR was effective over

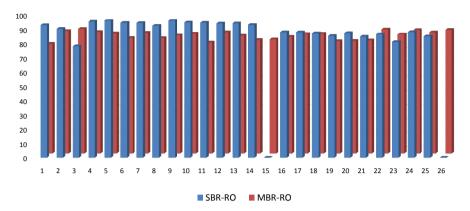


Figure 7. Comparison of TDS removal after treatment with RO using SBR and MBR as pre-treatment.

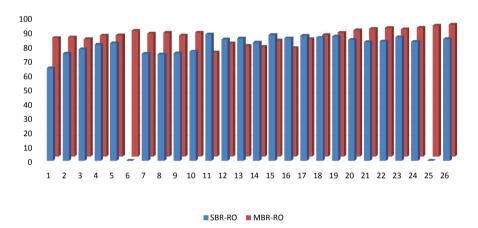


Figure 8. Comparison of COD removal after treatment with RO using SBR and MBR as pre-treatment.

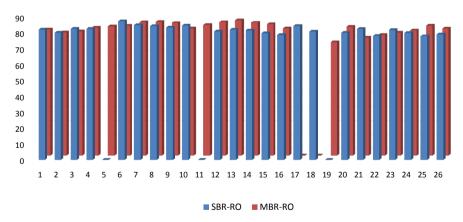


Figure 9. Comparison of ammonia removal after treatment with RO using SBR and MBR as pre-treatment.

MBR, while as pre-treatment the combination of MBR-RO was a little high since SBR-RO was equally competent. Since, reverse osmosis is efficient in removing solids, in biological treatment removal of chemical oxygen demand and ammonia are more desirable to get an overall effective treatment. Hence, the combina-

tion of SBR-RO was tested on a full scale in further studies.

4. Discussion

The prime objective of the current research is to evaluate the efficiency of sequencing batch reactor (SBR) and membrane bioreactor (MBR) as pre-treatment for reverse osmosis (RO).

4.1. Sequencing Batch Reactor

Treatability of pharmaceutical wastewater using sequencing batch reactor (SBR) was tested on a pilot scale. During one of the months *i.e.*, during June SBR outlet was fed to reverse osmosis (RO) to study the applicability of SBR as pre-treatment for RO. Efficiency of SBR is presented in terms of reduction in total dissolved solids and chemical oxygen demand. Details of SBR efficiency during the pilot trails is presented as follows:

> Removal of total dissolved solids

Previous studies have shown that wastewaters containing high TDS when treated by SBR result in substantial reduction of chemical oxygen demand, since high TDS is considered to affect the dissolvability of oxygen in the wastewater [32]. The highest removal of TDS was noted as 27.32% during the month of September while the lowest with negative removal was observed during the month of March with -29.28%. Wu and Maskaly, 2018 [33] stated that at increasing TDS levels aerobic microorganisms and their metabolism are adversely affected leading to failure in the system. Upon effective working conditions removal of TDS is observed which is attributed to the oxidative degradation of dissolved solids [34]. Studies conducted by Mahvi *et al.*, 2008 [35] on wastewater TDS removal reported a reduction of 61.25%.

> Removal of Chemical Oxygen Demand

In the present study, the highest and lowest COD removal was noted as 68.74% and 34.98% during the months of March and May respectively. The reduction in COD is attributed to aeration and digestion processes, which are confirmed by previous studies showing 99%, 90%, 98% and 94% removal respectively [36] [37] [38]. As per the report of USEPA, SBR is effective in removal of COD and BOD along with nitrification, denitrification and suspended solids [39]. Amat *et al.*, 2005 [40] obtained 99% to 86% of COD removal under varied loading factors on a lab scale SBR.

Dissolved Oxygen Profile

2.0 mg/l of dissolved oxygen concentration is required for maximum nitrification rate. When concentration of dissolved oxygen falls below 0.5 mg/l and at a maximum of 1.0 mg/l, it inhibits denitrification. The dissolved oxygen concentration of SBR system during the operation is understood to increase during the react stage as aeration is provided, the concentration decreases in the stages of settle, draw and idle due to ceasing of aeration and mixing processes. Moreover, concentration of dissolved oxygen is substantially related to activity of microor-

ganisms in the system. Further, peak of dissolved oxygen occurs after complete depletion of ammonia which indicates the culmination of nitrification process [41]. An increase in dissolved oxygen is observed when all the organic matter is degraded, reducing the respiration of microbes.

> Sequencing batch reactor as pre-treatment to Reverse Osmosis

One-month trail runs were performed wherein treated effluent from SBR is fed to reverse osmosis. The results were promising with the higher end removal of TDS, COD and ammonia recorded as 96%, 88.62% and 87.50% respectively. Meagre studies are found in context of integrated treatment using SBR followed by RO especially for treating pharmaceutical wastewaters. Gangavarapu *et al.*, 2015 [42] conducted a study on medium scale active pharmaceutical ingredient manufacturing industry that adopted recycling process through zero liquid discharge system. They reported the flow of effluent treatment process consisted of multi-effect evaporator followed by sequencing batch reactor which is concluded by reverse osmosis. They achieved reduction in total dissolved solids, total suspended solids and biological oxygen demand in the order of 99.2, 100 and 100 percent respectively. A combination of membrane sequencing batch reactor with reverse osmosis has achieved 90.9% reduction in chemical oxygen demand, 92% of total organic carbon and 91.5% of oil and grease from produced water of oil and gas field [43].

4.2. Membrane Bioreactor

MBR processes are experiencing exceptional growth in treating domestic and municipal wastewater during the past decade owing to many advantages like exceptional quality of effluent, lesser production of sludge, lower foot print and flexibility for expansion in future [44]. Industrial use of MBR technology gained attention attributed to the robustness in its process, resistance to high organic loading, ability to treat compounds that are difficult and inhibiting to treat [45]. Pharmaceutical wastewaters manufacturing vitamins were conventionally treated using two units of oxidation process wherein the treated water was unstable with greater amounts of suspended solids among other pollution parameters. Hence Feng et al., 2006 [46] suggested the use of MBR pilot plant to treat these waters instead of the conventional processes.

Removal of Total Dissolved Solids

TDS concentrations of permeate were in the range of 4856 mg/l to 32,000 mg/l. TDS removal was recorded to be 1.38% as the lowest and 29.25% as the highest. There exists a paucity in literature regarding removal of TDS by MBR. Almost all the studies focus on removal of TSS but not TDS. Moreover, TDS removal is always reported when MBR is used as a pre-treatment for RO. Hence, in the present study also the removal of TDS will be dealt in the following section of MBR as pre-treatment for RO.

> Removal of Total Suspended Solids

In the present study, TSS concentrations in permeate were recorded to be zero

0 with 100% removal. Removal of suspended solids effectively performed by membrane separation process, overcoming the limitations related to sludge settling properties [47]. MBR was employed for treatment of pharmaceutical wastewaters on a pilot scale in southern Taiwan. Performance was monitored for a duration of 140 days; it was observed that there was a regular logarithmic progress of viscosity in relation to TSS concentration [48]. 100% removal of suspended solids with high quality effluent was reported by Shahbeig *et al.*, 2013 [49] obtained 98% removal of TSS treated by MBR when ultra-filtration was used as a pre-treatment. Tee *et al.*, 2016 [50] reported 92% turbidity removal when mixed industrial wastewater was treatment in MBR.

> Removal of Chemical Oxygen Demand

COD removal in the present study was attained from 4.77% to 30.35% which was contrary to the previous studies and the reason can be traced to the discussion provided by Di Trapani *et al.*, 2014 [51]. Synthetic cephalosporin antibiotic wastewater water was treated using contact oxidation-hydrolysis-MBR the influent COD concentrations ranged between 2125 - 11,561 mg/L while the effluent concentrations obtained were 79 to 282 mg/L. Further, the effluent BOD obtained was less than 10 mg/L [52]. A diminution in COD from 3000 mg/L to 45 mg/L was obtained by integrated coagulation-contact oxidation—MBR for treating wastewater from pharmaceutical factory producing Lisinopril enalapril.

> Removal of Turbidity

Virtuous removal of turbidity was recorded with permeate values recorded as 0.91NTU as the lowest and 40.1NTU as the highest. Turbidity removal is in the order of 80.90% being the lowest and 99.86% being the highest. Studies elsewhere reported the efficiency of MBR in terms of effective removal of suspended solids, leading to zero turbidity of wastewaters. Removal of specific organic pollutants along with higher removal of COD was achieved by Lipp *et al.*, 2009 [53]. Exceptionally good quality of filtered effluent was obtained after treatment with MBR with turbidity of less than 1 NTU [54].

> Membrane bioreactor as pre-treatment for reverse osmosis

When treated waters from MBR were fed to RO the removal efficiencies of various parameters were found to be significant with TDS removal to be 26.96% as the lowest and 87.29% as the highest. COD removal varied from 73.03% to 92.33%. While hardness showed 93.33% - 100% removal, ammonia removal was good with 71.46% being the lowest and 85.51% recorded as the highest. MBR treatment in combination with traditional methods for treating pharmaceutical effluents containing anti-inflammatory and antibiotic drugs has achieved 90.4% reduction in COD [23]. Latest literature review quoting previous studies acknowledged MBR technology as novel method of pretreatment prior to RO treatment for secondary effluent. Further, it has been reported that complete removal of all PhACs is not achieved by MBR or in fact any single treatment unit and hence advanced treatment techniques like NF, Ozone oxidation, advanced oxidation and RO are to be integrated to achieve

complete removal of pollutants.

4.3. Comparison of Sequencing Batch Reactor and Membrane Bioreactor

In the present study maximum reduction of total dissolved solids was obtained by SBR with 31.82%. On the other hand, MBR present lowest removals with 29.25%. Chemical oxygen demand was the highest (69.54%) with SBR, while MBR was stable with maximum of 30.35% removal of COD. Lefebvre et al., 2014 [55] compared the efficiencies of SBR and MBR in treating pharmaceutical wastewaters produced from manufacturing of antibiotic penicillin. They reported better performance of both units at lower organic loading but faced with foaming issues upon increasing the organic load. Further, SBR was good in degrading aromatic compounds while MBR achieved good solids removal. Pilot-scale studies for treating municipal wastewaters using SBR and MBR were studied by Mirbagheri et al., 2017 [56]. They reported that SBR achieved 98.54% of BOD₅ removal while MBR achieved 97.1% of COD. They concluded that both the treatment options were suitable for treating municipal wastewater. MBR exhibited higher removal rates of pollutants over SBR with 99.5% vs. 82% for BOD₅; 70% vs. 47% for COD; 95% vs. 73% for TN and 96% vs. 71.4% for NH₃-N. The study opined requirement of post-treatment for effluents from both units owing to the high strength raw wastewater. Further, they stated that factors like technical expertise, footprint and membrane lifespan are to be considered when assessing the economic burden of adopting MBR treatment technology [57]. Apart from treatment efficiencies one important deciding factor for adopting a treatment technology is the associated cost. Some authors have focused on this aspect and presented their results. With reference to STPs the cost wise hierarchy is presented as MBR < USAB < SBR < MBBR with MBR requiring the highest and MBBR requiring the lowest cost. With this and removal efficiency it can be concluded that SBR would be most economical and efficient treatment technology that can be adopted in India. Further, MBR requires 25% more economy in comparison to SBR. Moreover, with reference to treatment efficiency SBR is equally competent to MBR [58] [59].

4.4. Comparison between SBR and MBR as Pre-Treatment for RO

Total dissolved solids removal was the highest in the combination of SBR-RO treatment with 95.94% removal. While MBR-RO combination resulted in 87.29% removal. Chemical oxygen demand was achieved maximum with the combination of MBR-RO 92.33% while competitive results were achieved with the combination SBR-RO also 88.62%. Removal of ammonia was maximum with the combination SBR-RO (87.5%), while competitive results were obtained with MBR-RO (85.51%). High paucity in literature exists in comparing pre-treatment options for RO. Some of the workers elsewhere voted for SBR + RO/UF over MBR attributed to the cost-effectiveness associated with SBR.

From the results and discussion, it can be understood that individually SBR was effective over MBR. Though as pre-treatment the combination of MBR-RO was a little high, SBR-RO was equally competent. Since, reverse osmosis is efficient in removing solids, removal of chemical oxygen demand and ammonia in biological treatment is more desirable to get an overall effective treatment. Hence, the combination of SBR-RO was tested on a full scale in the further studies.

5. Conclusions

Removal of chemical oxygen demand from wastewaters requires adopting biological treatment. Available conventional biological treatments are not able to treat to the required standards. Literature review stated the use of hybrid processes or combination of various treatment technologies to achieve discharge standards. Hence, in the present study a combination of advanced biological process and advanced physical membrane separation process were chosen to test their combined efficiency in treating pharmaceutical wastewaters. The biological treatment methods evaluated for their efficiency are sequencing batch reactor and membrane bioreactor, further their suitability as pre-treatment to reverse osmosis treatment was also evaluated.

Efficiency comparison among biological treatments SBR and MBR: Total dissolved solids removal by SBR was 31.82% and MBR showed 29.25%. SBR even though presented the highest removal was quite fluctuating in treatment process. Chemical oxygen demand removal by SBR was 69.54% and MBR showed 30.35% removal of COD. Since the prime objective of biological treatment is removal of chemical oxygen demand and SBR was efficient over MBR, SBR was selected for biological treatment of pharmaceutical wastewaters.

Efficiency comparison among combined treatments of SBR-RO and MBR-RO: Removal of total dissolved solids was the highest in the combination of SBR-RO treatment with 95.94% removal. While MBR-RO combination resulted in 87.29% removal. Chemical oxygen demand was achieved maximum with the combination of MBR-RO 92.33% while competitive results were achieved with the combination SBR-RO also with 88.62% removal. Removal of ammonia was maximum with the combination SBR-RO 87.5%, while competitive results were obtained with MBR-RO 85.51%. From the results, it can be understood that SBR was efficient in removing ammonia and total dissolved solids and was equally competent in removing chemical oxygen demand. Since, reverse osmosis is efficient in removing solids, in biological treatment removal of chemical oxygen demand and ammonia are more desirable to get an overall effective treatment. Hence, the combination of SBR-RO was tested on a full scale in the further studies.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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