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Osmotic Membrane Bioreactor for Oily Wastewater Treatment using External & Internal Configurations

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Abstract

The present work aims to study the treatment of oily wastewater by means of forward osmosis membrane bioreactor process. Side stream (external) configuration and submerged (internal) configuration of osmotic membrane bioreactor were performed and investigated. The experimental work for each configuration was carried out continuously over 21 days. The flux behavior of forward osmosis membrane in an osmotic membrane bioreactor (OMBR) was investigated, using NaCl as the draw solution and CTA as FO membrane. The effect of mixed liquor suspended solids (MLSS) concentration and TDS accumulation of bioreactor on water flux and membrane fouling behaviors was detected. The accumulation and rejection of nutrients in the bioreactor (Nitrate, COD, and Phosphate) were investigated over the days of the experiment. Water flux and membrane fouling were not significantly affected by MLSS concentration at low level and this effect increase with increasing MLSS concentration (4000–10000 mg/L). Besides, water flux was severely affected by elevated salinity of the aeration tank. OMBR showed high removal of COD (96%) and FO membrane revealed high retention of phosphate (97%) but retention for nitrate was relatively low (72%). The sparingly soluble salts in the influent, bioreactor, draw solution, and RO effluent were detected through the experiment. The results showed flux decline with time to about 47% from the initial flux and two osmotic backwashing were applied at day 7 and 14 during the operation and the flux restored approximately 30% of its loss. Side stream and submerged configurations revealed nearly similar response over the experiments while side stream module showed better water flux (7.0 LMH) than submerged (6.1 LMH). The results showed that the concentration of inorganic ions is below the limits that may cause severe scaling.

Key words: Forward Osmosis, Water Flux, Fouling, Osmotic Membrane Bioreactor.

Introduction

Generally, wastewater comes from two major sources: human sewage and process waste from manufacturing industries including oil refineries. Oil, grease and hydrocarbons are the essential contaminants of oil refinery wastewater. Innovative processes based on membranes coupled with biotreatment for wastewater, especially oily wastewater treatment were recently proved as promising technologies to produce high quality water that can be reused as well as free of toxic and harmful constituents for the living organisms when discharged to the environment. Membrane bioreactor (MBR) innovation initially studied and reported for wastewater treatment application around 40 years ago, is presently effectively utilized in municipal and industrial wastewater treatment [1–6]. MBR has become well-known because of some particular favorable circumstances contrasted with traditional wastewater treatment processes, such as, decreased footprint, low creation of excess sludge [2], great effluent quality and high concentration of sludge [1]. The major drawback of this process is high energy consumption during application due to extra aeration required for mixing as well as to control membrane fouling [4, 7, 8]. Two configurations were used in MBR process: external and internal configuration. In external mode, the membrane cited outside the bioreactor, while in internal mode, the membrane cited inside the bioreactor. As of late there has been expanding attention for a novel MBR which incorporates forward osmosis (FO) and the biological procedure for wastewater treatment in a technique usually known as osmotic membrane bioreactor (OMBR). OMBR is a perfect technology which include multi-barrier that can be utilized for indirect and direct reuse applications of potable water [9–15]. Forward osmosis (FO) membranes utilized in OMBRs are applied to withdraw water through a dense, semi-permeable layer from a low-salinity waste feed into a high-salinity draw solution (DS). In some applications an RO process is utilized to re-concentrate the diluted draw solution and at the same time produce high quality water. Osmotic pressure difference over FO membrane between feed (activated sludge) and the draw solution is the driving force in OMBR. The essential benefits of using FO membrane over other membrane separation technologies with respect to wastewater treatment are the low fouling propensity and the perfect rejection of macromolecules, trace organic compounds TOrCs, and ions [16–24].

Previous researches have highlighted the points of interest and uses of OMBRs [9-13]; besides, they have additionally identified the accumulation of total dissolved solids (TDS) and other dissolved components in the aeration tank as an essential disadvantage of the OMBR operation [9, 14, 15]. The high rejection of TDS and nutrients by FO membrane lead to accumulation of these constituents in the bioreactor as well as reverse salt flux by diffusion from the draw solution into the bioreactor. The TDS and hence salinity of activated sludge increases with time which in parallel decreases the osmotic pressure difference over the membrane (reduce driving force and hence water flux) and can unfavorably influence microbial activity and usefulness in the bioreactor which would advance effect membrane fouling [25, 26]. Besides, the interactions of the inorganic particles (particularly divalent cations) as well as organic foulants, and additionally the scaling of low dissolvable salts (e.g. gypsum, calcium carbonate and calcium phosphate [30]) under relative high strength of ions offer ascent to higher complex fouling issues on FO membranes [27–29].

The aim of the current work is to investigate the feasibility of OMBR process, which combines FO process and MBR process, in treating oily wastewater and produce high quality water. The OMBR process imply investigation of system configuration (side stream and submerged), MLSS concentration, and FO membrane fouling and backwashing, As well as nutrient removal and accumulation. Moreover, the effects of salt accumulation on biological treatment performance with respect to water flux will also be investigated.

Materials and Methods

The OMBR process includes treatment of oily wastewater by means of two configurations, side stream and submerged as shown in Figures 1 and 2. Using reactor with 8 L total volume for submerged and 3L for side stream contain an air distributor connected with blower to supply O_2 at 4 l/min flow rate which is required for growing of microorganisms. 0.6 M NaCl concentration was used as draw solution and 3 l/min feed and draw solutions flow rate, 30 ±1℃ temperature of both feed and draw solutions. The membrane was asymmetric cellulose triacetate (CTA) FO membrane delivered by HTI Albany, OR. The process carried out without usage of mesh spacer in the FO cell to reduce accumulation of biomass on the membrane and cause fouling issues. The feed used in the present study was brought from Alduara refinery (the effluent stream of DAF process in wastewater treatment unit) and the activated sludge brought from aeration tank at the same unit. The level of activated sludge in the reactor was kept constant at 20 cm height for submerged (5 L of activated sludge (AS)) and 15 cm height for side stream(3 L of AS) by utilizing float inside the reactor. The float is connected to a supply tank contain oily wastewater to compensate the reactor with the lack in water due to permeation. The supply tank located in a higher place than reactor (70 cm) to impart feed without need of pump as illustrated in Figures 1 and 2. The duration of experiment for each configuration of biological stage was 21 days included two osmotic backwashing at day 7 and day 14 of the experiment. Sludge was wasted manually using a graduated cylinder from the bioreactor with rate of 150 ml/day for side stream mode and 250 ml/day for submerged mode starting on day 7 of the continuous operation, and the computed solids retention time (SRT) according to this wasting rate is 20 days. The concentration of draw solution was kept constant throughout the run using vessel with flow regulator fixed at the bottom which contains concentrated draw solution (200 g/l). The concentrated draw vessel was placed over the draw solution vessel as shown in Figures 1 and 2. Concentrated draw solution was flow as drops to balance the dilution of draw solution by water permeation and this flow was modified daily according to dilution rate. Samples of draw solution was taken periodically and treated in RO system which manufactured by STERLITECH, GE Osmonics, and USA. Polyamide membrane were used with dimension of 305 x 305mm. Five liters of draw solution were processed for each run in this system with concentration of 35 g/l of NaCl. The applied pressure was 50 bar and the permeate flux was 25 LMH. The RO permeate was collected and submitted to several tests such as: conductivity, COD, and ions concentration by means of UV spectrometer and flame photometer to evaluate permeate quality (treated water). The RO system was located at Ministry of Science and Technology / Environmental and Water Directorate. Samples were taken and analyzed every five days from feed tank; bioreactor, draw solution, and RO permeate.

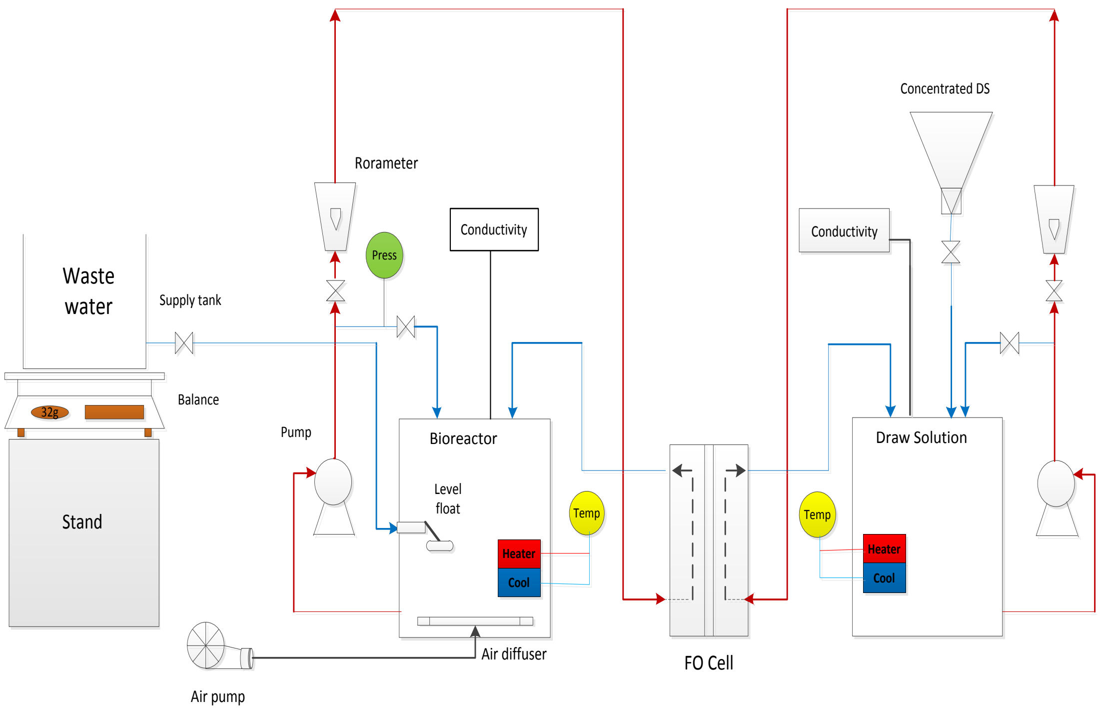

Fig. 1: Schematic Diagram of Side stream Osmotic Membrane Bioreactor Process

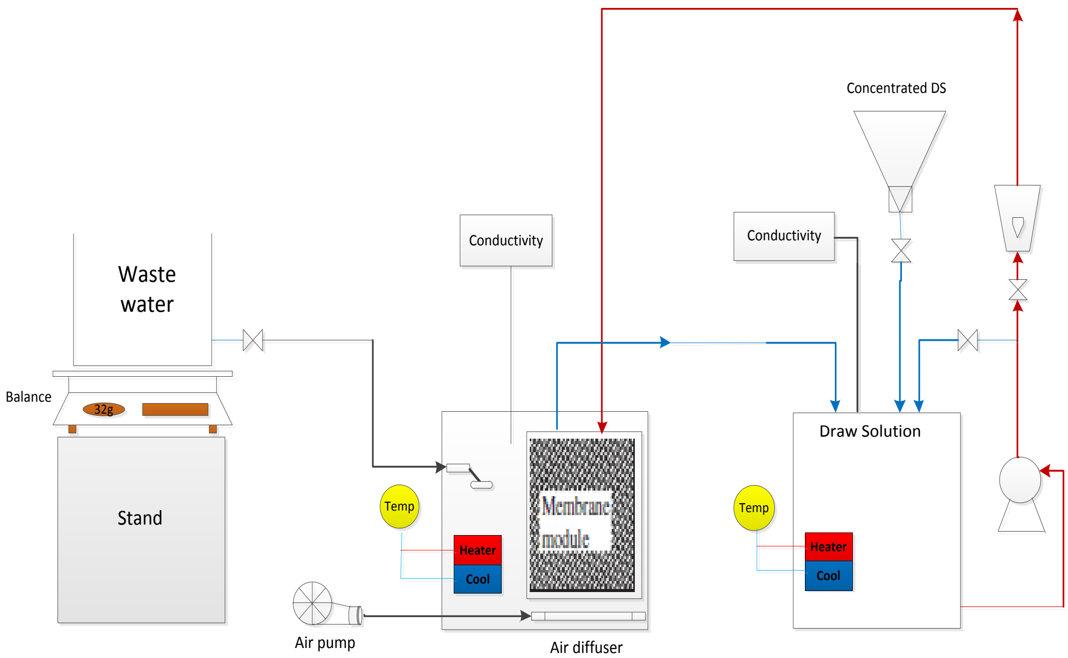

Fig. 2: Schematic Diagram of Submerged Osmotic Membrane Bioreactor Process

Results and Discussions

1. Effect of MLSS Concentration

The influence of mixed liquor suspended solids (MLSS) concentration with respect to water flux using side stream configuration is shown in Figure 3. Three concentrations of MLSS were tested such as 4000, 7000, and 10000 mg/l. By increasing concentration of MLSS the water flux decline had become more severe with time due to increase in concentration of biomass and hence more tendency of membrane fouling. This outcome is compatible with conventional membrane bioreactor (MBR) process [1, 4]. It was documented that membrane fouling resistance is increased proportionally with MLSS concentration in traditional MBR. The reason for this result is due to high MLSS concentration of activated sludge contains excess foulants and hence more intense fouling.

Figure 3 shows low effect of MLSS on water flux and this effect increase with increasing MLSS concentration.

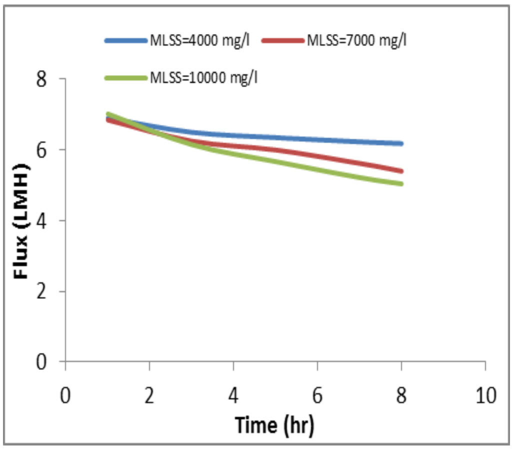

Fig. 3: influence of MLSS concentration on flux decline with time

2. Water Flux

The osmotic membrane bioreactor (OMBR) process was operated with fixed DS concentration which is approximately 35 g/l (0.6 M). Water flux as a function of time over the duration of experiments, which is 21 days, is shown in Figure 4. Throughout OMBR testing, the FO membrane was refreshed by osmotic backwashing process on days 7 and 14 of operation. The initial water flux for side stream mode was approximately 7.0 LMH as illustrated in Figure 4 and for submerged module, the initial water flux was nearly 6.0 LMH as shown in Figure 5. For both configurations, the initial flux is followed by steady decrease in flux over 7 days of operation. The decline in flux might be attributed to decrease in osmotic driving force as well as membrane fouling. The decrease in osmotic driving force across FO membrane was due to increase in bioreactor salinity from approximately 1.0 g/l to 7.0 g/l TDS as illustrated in Figure 6. The salt accumulation in the aeration tank was due to salts incoming with the influent to the bioreactor as well as diffusion of salts to the bioreactor from DS tank (reverse salt flux). The DS concentration over the period of 21 days of operation was kept constant. The difference in concentration across the FO membrane reduced from 35 g/l to 28 g/l during the experiment. The influence of fouling on membrane activity with respect to flux decline was illustrated between day 7 and 14 as shown in Figures 4 and 5 for submerged and side stream configurations respectively, where cleaning take place by osmotic backwashing. Figures 6 and 7 show that after day 7 of operation, the TDS of the bioreactor is nearly stable due to starting of daily wasting from the bioreactor at this period. The increase in salinity (TDS) of the bioreactor for the first 7 days was more than five times of the initial value (1.09 g/l to 5.52 g/l), while for the next seven days after begin of sludge wasting, the salinity of the bioreactor was increased by solely 20% (5.52 g/l to 6.75 g/l). The excess salts accumulated in the bioreactor were withdrawn with daily wasting. This case indicates that the dropping in flux throughout operation was due to membrane fouling not to variance in driving force.

Figures 8 and 9 show and summarize the outcomes of the OMBR experiments which performed to study the fouling of membrane and effect of osmotic backwashing on flux behavior at day 7 and 14 of continuous operation for side stream and submerged configurations. The left and right bars of each pair show the effect of membrane fouling and osmotic backwashing on water flux, respectively. After day 7 of process, the water flux reduced by approximately 47% from the initial flux of new membrane. After osmotic backwashing, the flux restored approximately 30% of its loss, with an overall of 17% loss in water flux which may attributed to irreversible fouling. The second backwashing at day 14 of operation show that the water flux reduced by 23% from day 7 and the second backwashing restore about 87% of the loss in flux. The reduce in flux decline after day 7 is due to daily wasting of activated sludge from the bioreactor which reduce the accumulation of salts and hence enhancing osmotic pressure difference over the membrane. This state gives a suggestion that after a state of irreversible fouling, which is occurring in the first 7 days, subsequent behavior of fouling become more reversible which is consistence with previous studies [9, 14]. The water flux for submerged mode (6 LMH) was less than external mode (7 LMH). The difference in flux between two modes may be due to accumulation of foulants on the active layer of membrane forming thin layer of biofoulants

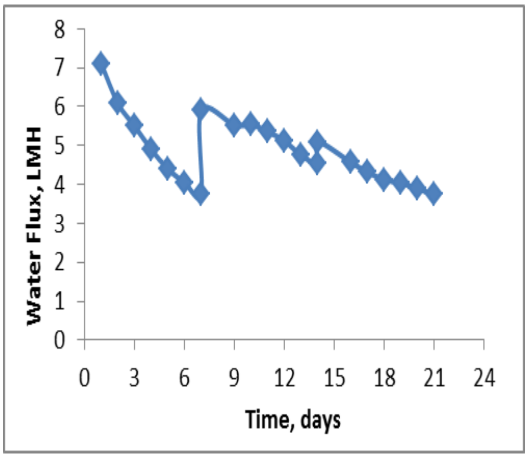

Fig. 4: Water flux over the course of 21 days of OMBR for side stream mode

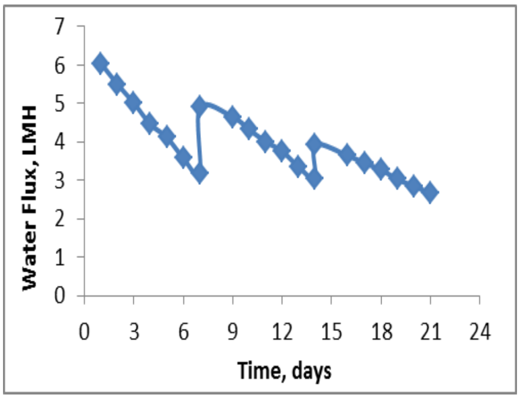

Fig. 5: Water flux over the course of 21 days of OMBR for submerged mode

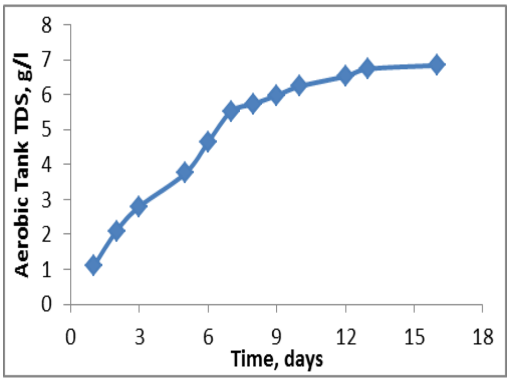

Fig. 6: Bioreactor salinity over the course of 21 days of operation for side stream mode

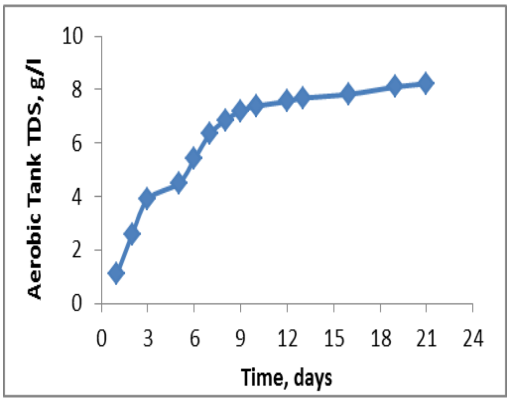

Fig. 7: Bioreactor salinity over the course of 21 days of operation for submerged mode

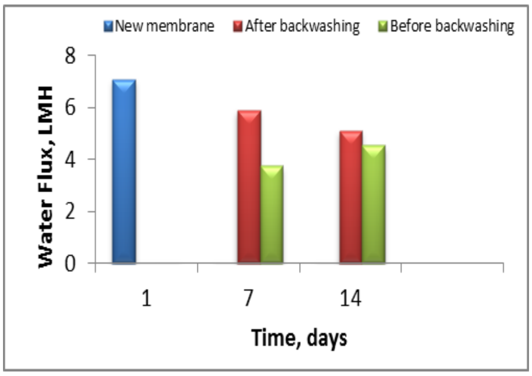

Fig. 8: Effect of membrane backwashing on water flux for side stream mode

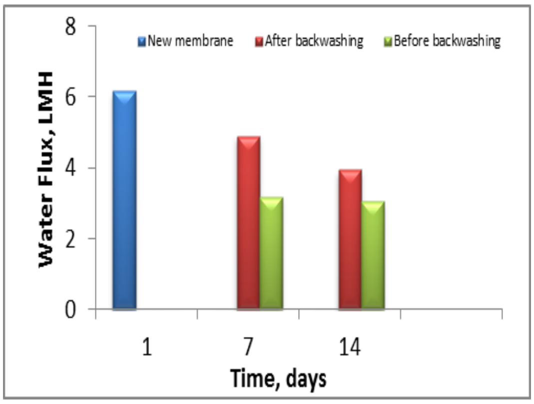

Fig. 9: Effect of membrane backwashing on water flux (submerged mode)

The aeration technique, especially in submerged configuration, is an important factor in fouling control. The rate of air flow utilized in this study was 4 l/min and the air diffuser was fixed in line with membrane with sufficient coarse bubbles which provide scouring of the membrane surface and prevent fouling. Therefore thin layer was formed on the membrane which caused a low resistance to water flux. Another reason of severe membrane fouling through OMBR process is the accumulation of sparingly soluble salts in the aeration tank. The concentration of basic ions in the bioreactor was measured on four occasions over OMBR process and the results are listed in Table 1. The values of ions in this table were implemented in ROSA software to find out the tendency of $CaCO_3$ precipitation in the bioreactor by knowing value of Langelier Saturation Index (LSI). The value of LSI should be less than one to avoid scaling. Figure 10 shows the magnitude of LSI value with time. LSI value was increased from -0.13 to 0.47 after 7 days of operation while there was slow increase in LSI after day 7 due to removing of inorganic ions through wasting. The values of LSI, after 21 days of continuous operation, are less than 1.0 which is consistence with previous studies [33-36]. Although this value of LSI reveals a slight tendency for $CaCO_3$ precipitation, this precipitation may occur in the activated sludge rather than on the membrane. Figure 11 shows that the concentration of $CaSO_4$ after 21 days of OMBR process is below the saturation limit and needs no additives or anti-scalants for scaling control.

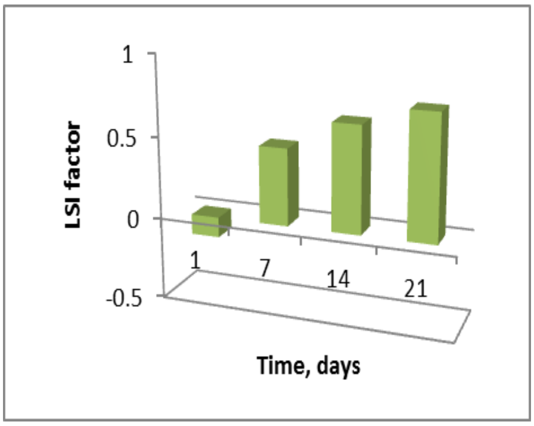

Fig. 10: Langelier Saturation Index (LSI) for the bioreactor change with time

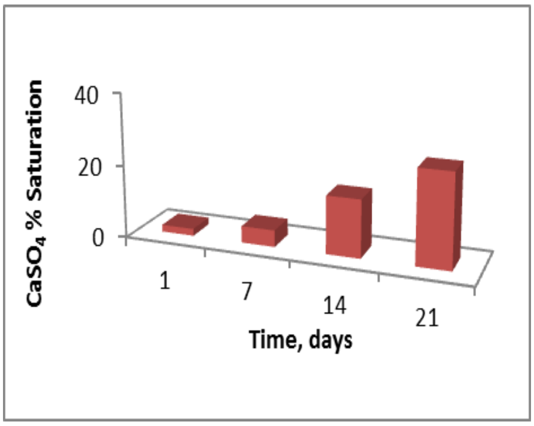

Fig. 11: Saturation percentage of $CaSO_4$ over the course of operation

Table 1: Ions concentration in the bioreactor

Ions	Ion concentration (mg/l)			
	Day-1	Day-7	Day-14	Day-21
Na	148	662	796	833
Ca	56	235	311	357
Mg	65	304	346	388
K	2.6	14.5	16.0	18.3
SO_4	139	635	790	854
CL	123	588	738	890
NO_3	10.5	25.2	30.6	15.6
PO_4	1.5	6.8	7.5	7.9

3. Nutrient Rejection and Accumulation

Phosphate

The values of phosphate concentration during the course of operation in the RO permeate and DS was under the detection limits. Due to high membrane rejection of phosphorus, it will steadily accumulate in the bioreactor over the OMBR operation. After day 7 of process and when sludge wasting is started, the phosphorus concentration is almost leveled off as shown in Figures 12 and 13 for the two configurations.

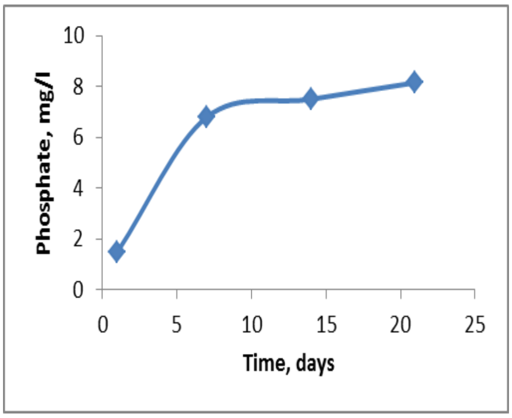

Fig. 12: Concentration of Phosphate in Bioreactor as a Function of Operation Days for Side stream Mode

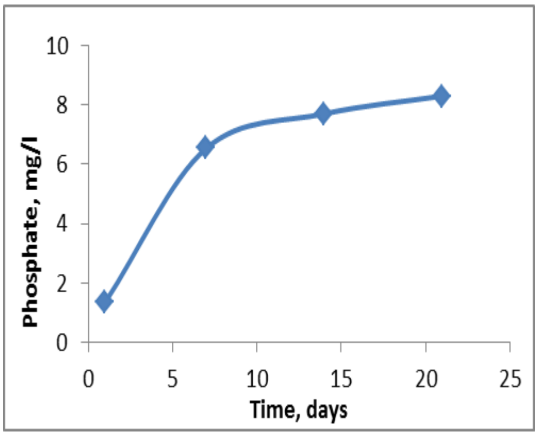

Fig. 13: Concentration of Phosphate in the Bioreactor as a Function of Operation Days for Submerged Mode

The phosphorus rejection via FO membrane was about 98% and the total system (FO + RO) rejection was more than 99%. These results were consistence with earlier studies [31, 32] which achieved more than 99% rejection of phosphate.

Nitrate

From the ionic analysis of aeration tank, draw solution, and RO permeate, the nitrate concentration was found relatively high for the first 14 days of continuous operation as shown in Figures 14 and 15 for side stream and submerged configuration respectively. The high concentration of nitrate is proof of nitrification process (ammonia oxidation) and the denitrification process (nitrate reduction) is limited within this period. During the third week of the OMBR operation, the concentrations of nitrate begin to decrease with time which may be attributed to the increase in bioreactor accumulated salinity. The nitrification process is controlled by bacteria called ammonia oxidizing bacteria (AOB). From the analytical results, the AOB activity was influenced and inhibited by increasing bioreactor accumulated salinity. During the course of 21 days of operation, the nitrate concentration in the draw solution increased with time which is attributed to diffusion of nitrate through the FO membrane from the bioreactor which is consistence with earlier studies [31, 33]. The low rejection of nitrate (72%) by CTA FO membrane is drawback which should be considered in such processes. The nitrate concentration in the RO permeate was relatively low and below the maximum permissible limit of nitrate in drinking water concentration (10 mg/l) which represented as a toxic and hazardous component for health according to Environmental Protection Agency, EPA, which make the water convenient for reuse in most applications.

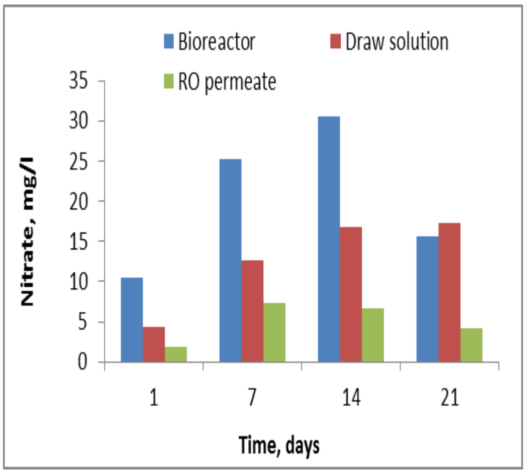

Fig. 14: Concentration of Nitrate in the bioreactor, draw solution, and RO permeate as a function of operation days for side stream mode

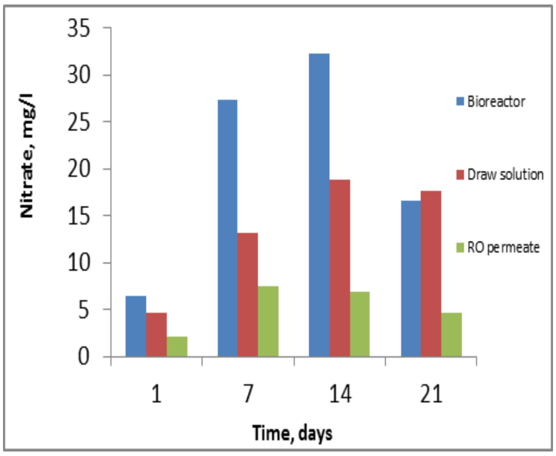

Fig. 15: the concentration of nitrate in the bioreactor, draw solution, and RO permeate as a function of operation days for submerged mode

COD

The COD concentration in the influent, bioreactor, draw solution, and RO permeate were illustrated in Figures 16 and 17 for the side stream and submerged, respectively. The COD concentration in the bioreactor was observed to be increased slightly during the OMBR operation and this might attributed to the increasing salinity of the activated sludge which affects the biological activity and degradation performance.

The COD concentration in the RO permeate was less than 10 mg/l and this result show the advantage of utilizing multi-barrier technology like OMBR.

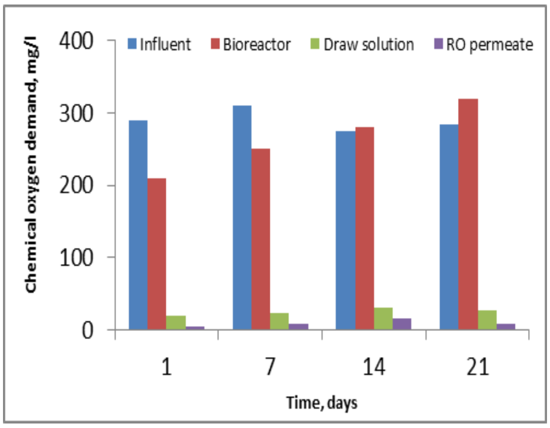

Fig. 16: Concentration of COD in the Influent, Bioreactor, Draw Solution, and RO Permeate as Function of Operation Days for Side stream Mode

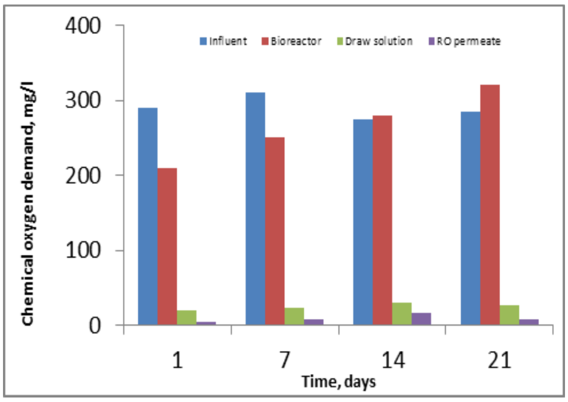

Fig. 17: The concentration of COD in the Influent, Bioreactor, Draw Solution, and RO Permeate as Function of Operation Days for Submerged Mode

Conclusions

This study provides useful information for the determination of appropriate parameters in OMBR operation as well as better understanding of OMBR process with applying two configurations, side stream and submerged. Initial water flux for side stream mode was 7.0 LMH, while initial flux for submerged mode was nearly 6.0 LMH. Water flux and membrane fouling were showed less influenced by MLSS concentration at certain level (4000 mg/l) and this effect increase with increasing MLSS concentration. OMBR system exhibited excellent removal/rejection of nitrogen, phosphorus, and COD from the analysis results which conducted for influent and RO permeate. The average removal of nitrogen, phosphorus, and COD during the OMBR exceeded 72%, 97%, and 96%, respectively, for both configurations. Despite the low nitrogen removal compared to the phosphorus and COD, its concentration in the RO permeate was less than 10 mg/l for the OMBR process. Accumulated sparingly soluble salts in the bioreactor were increased over the course of 21 days to a value below the limits that may cause severe membrane scaling as determined by Langelier Saturation Index (LSI) and $CaSO_4$ saturation. The results revealed sharp flux decline with time for first week for both modes and this decline become less severe when start wasting. Osmotic backwashing found to be efficient way for membrane refresh.

Acknowledgements

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