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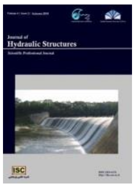

A comparative study on the use of membrane bioreactor (MBR) and activated sludge followed by ultrafiltration (CAS/UF) processes for advanced treatment of industrial wastewater

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Abstract

With increasing industrial wastewater reuse, due to inadequate water resources, membrane technology has shown results of very high efficiency in a wide range of reuse purposes and reverse osmosis (RO) pre-treatment. In the present study, the performance of two pilot-scale CAS/UF and MBR to polished effluent wastewater of an industrial town treatment plant were evaluated and compared in a continuous 43-day period. According to the test results, the removal efficiency of total suspended solids (TSS) for both reactors was nearly 100%. Nevertheless, the MBR improved the Chemical oxygen demand (COD) and the total dissolved solids (TDS) by nearly 3% and 5%, respectively. Total nitrogen (TN) and total phosphorus (TP) removals of approximately 31% and 20% in The MBR-based process and 24% and 18% in the CAS/UF module were obtained. Analysis of heavy metal concentration indicated that Cr, Pb, and Ni, which were in both soluble and particle forms, could be adequately eliminated by each system, while Cu, which was mainly in a soluble form, had a lower removal rate (32% and 51% in CAS/UF and MBR, respectively). Besides, since the silt density index (SDI) value for most of the samples was less than 3, both reactors can be used as RO pre-treatment systems.

Keywords: Membrane bioreactor (MBR); CAS/UF; Heavy metal; Industrial wastewater; Organic micropollutant.

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1. Introduction

The increasing dependence on water for numerous aims such as non-potable applications and industrial activities attracted considerable attention to alternative water resources especially in the Middle East, where the scarcity of water supply is of paramount concern [1, 2]. To solve this shortage, various studies have been done using different methods to purify water and remove

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contaminations from effluent wastewater [3, 4]. The deficiency of conventional wastewater purification methods to remove organic micropollutants (OMPs) led to the advent of advanced technologies, e.g. modified activated sludge [5], membrane bioreactor (MBR) [6], microfiltration or ultrafiltration (UF or MF) membranes [7], reverse osmosis (RO) [8], and conventional activated sludge (CAS) followed by UF membranes [9].

Recently, membrane separation mechanisms have become a practicable option for municipal and industrial wastewaters treatment with an acceptable effluent quality [10]. In UF/NF procedures, although permeate flux reduction by membrane fouling is still one of the most considerable challenges to achieving suitable potable water [11-13], they can be applicable due to the combination of biological wastewater treatment with physical separation processes, which leads to less space occupation and expenditure [14]. Additionally, using this method, the toxicity of wastewater and non-potable water can be reduced through using low-dose disinfection [15]. Meanwhile, membrane filtration can be widely utilized as pre-treatment for removing suspended solids [16], a wide range of contaminants, endocrine-disrupting compounds [17], heavy metals [18], and landfill leachate [19, 20]. Also, a group of researchers used ultrafiltration and microfiltration membranes as a tertiary treatment [21]. The results indicated that ultrafiltration could be more efficient compared to the microfiltration process, especially in eliminating viruses.

Another well-known membrane application for wastewater reclamation is membrane bioreactor (MBR) which combines biological degradation process with physical separation [22]. In this approach, merging the second and third purifying steps caused a smaller footprint, lower power requirements, and less sludge generation [23-25].

Similar to the CAS/UF process, the MBR mechanism works through biodegradation, adsorption to the biomass or membrane, and volatilization [9]; Nevertheless, MBR systems are comparatively operated with long SRT [26], which is appropriate in terms of higher biomass retention, the dominance of slow-growing microorganisms, and more macro-molecules consumption [27,28]. Another advantage of the MBR is a great efficiency of COD removal because of the remaining high concentration of the mixed liquid suspended solids (MLSS) in such systems [29]. Lerner *et al*. represented that the MBR technology compared to the conventional activated sludge process (ASP) has a superior output quality in terms of organic matter, suspended solid (SS), and nutrients [30]. Moreover, according to an investigation, the MBR process showed a suitable performance in the elimination of some metals such as iron, lead, copper, nickel, and aluminum (approximately 70%) [31].

The ultrafiltration membranes were applied as CAS-UF and MBR in order to identify and compare the quality of physicochemical and microbiological parameters [10]. According to the results, although the effluent of the two systems had similar physicochemical characteristics such as turbidity and suspended solids, the COD and coliphage reduction were more efficient in the MBR.

Kent *et al*. attempted to detect the differences between the performances of pilot-scale MBR and CAS-TMF. The results showed that COD, TOC, and polysaccharide concentrations were lower in the MBR permeate due to its higher SRT and retention of soluble microbial products [31]. Besides, utilizing them simultaneously in the RO pretreatment uncovered that the RO fouling with MBR pretreatment was nearly half of that with CAS-TMF pretreatment.

Kitanou *et al*. aimed to investigate the difference between an external pilot-scale MBR and a conventional activated sludge process (ASP) plant. Based on the results, the MBR could remove total suspended solids (TSS), BOD_5, COD, total phosphorus (TP), and total nitrogen (TN) far more effectively than ASP [32]. The concentrations of the heavy metals in the ASP and MBR effluent met the irrigation reuse standards; however, in the latter process, since the membrane could keep the suspended solids, the metal ions attached to sludge flocs, and were removed properly.

In recent decades, scarcity of water resources has made the reuse of industrial wastewater economically justifiable. Therefore, using an appropriate method as the RO pretreatment seems vital in sustainable water and wastewater management. With these considerations in mind, the chief aim of this work was to compare the performance of industrial wastewater treatment with pilot-scale CAS-UF and MBR. The elimination of certain pollution indicators such as COD, TSS, TDS, TN, and TP as well as some heavy metals (Ni, Cu, Pb, and Cr) was precisely monitored over a six-week period. Moreover, the fouling potential of the RO pretreatment was examined using the silt density index (SDI).

2. Experimental procedure

2.1. Effluent characteristics

The influent utilized in this work was received from Shokouhieh industrial wastewater treatment plant (WWTP), Qom, Iran, where there are several dairy, welding, metal finishing, beverage, etc. factories. After conducting the pretreatment in this plant, the municipal and industrial wastewater is transferred to the treatment plant, which consists of screens, equalization tank, anaerobic reactor, aerobic tank, sedimentation, sand filter, and a disinfection system (see Fig.1). However, flaws in design, performance, and elimination process have caused a high level of contaminations and heavy metals concentration in the effluent wastewater. Indeed, mixers designed for equalization tank, before the anaerobic reactor, do not work properly, and also fine diffusers at the bottom of the aerobic tank become frequently clogged, which disrupts the aerobic reaction. The physicochemical characteristics of the used wastewater, which was collected from the outlet of sand filtration (before chlorination unit), are shown in Table 1.

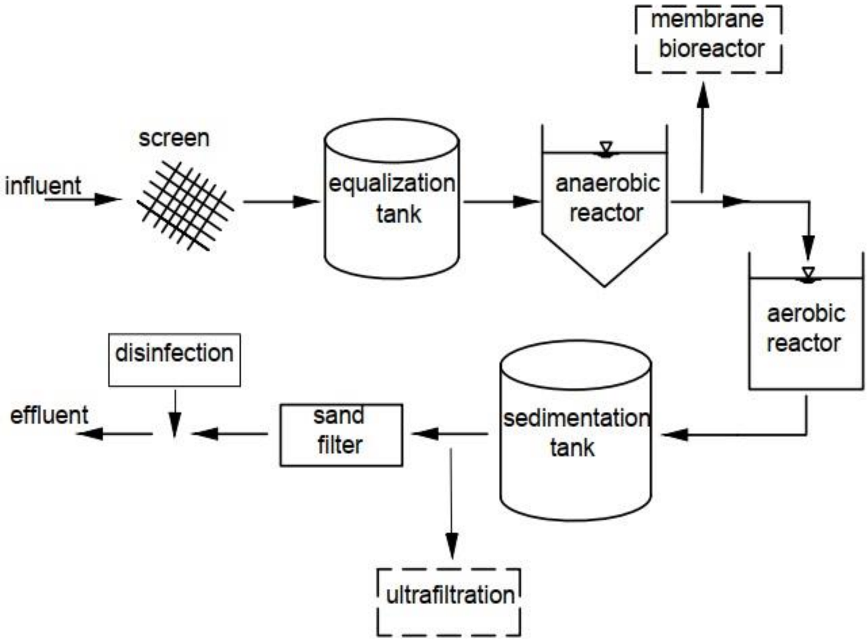


Figure 1. Flow diagram of Shokouhieh industrial wastewater treatment plant

2.2. Pilots configuration and operating conditions

In the current investigation, two continuous pilot-scale UF and MBR (see Fig. 2(a) and (b)) were designed and implemented, and the experiments in this study were conducted in the reactors. Each membrane was made of Plexiglass with a total volume of 32 L. For the filtration process, two polyethersulfone (PES) ultrafiltration membranes were used and placed in the reactors. The membrane specifications can be seen in Table 2.

Table 1. Typical characteristics of feed wastewater

Parameter	Unit	Value
pH	–	7.3 ± 0.62
TSS	mg/L	223 ± 32
COD	mg/L	250 ± 64
Al	µg/L	250 ±70
Fe	µg/L	180 ± 80
Pb	µg/L	340 ± 190
Cu	µg/L	610 ± 170
Ni	µg/L	160 ± 90
Cr	µg/L	225 ± 105
TKN	mg/L	51 ± 30
Alkalinity as CaCO3	mg/L	212 ± 43

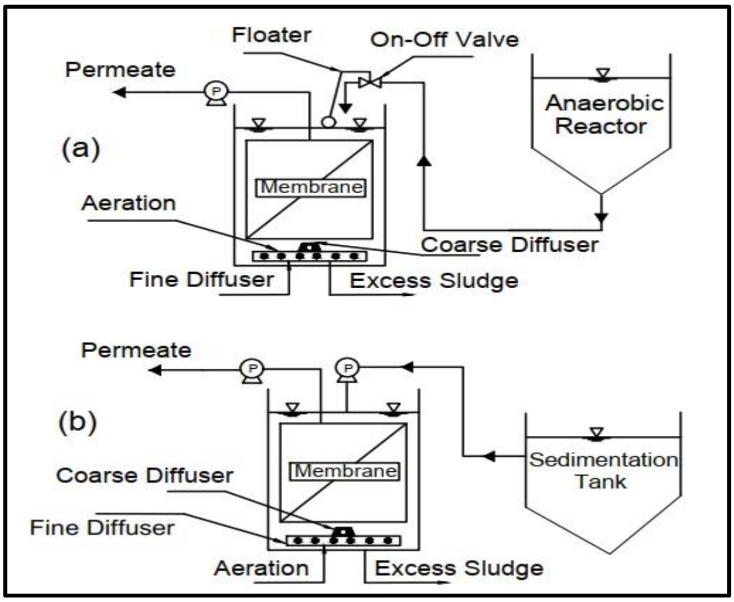


Figure 2. Schematic diagram of (a) MBR and (b) CAS/UF

Table 2. Specification of the membrane used.

Process Parameters	Unit	Value
Membrane configuration	-	Flat Sheet
Cut off	KDalton	150 KDalton
Pore size	mm	0.4 µm
Dimensions (Width × Height)	mm^2	240 × 200 mm^2
Effective surface area	m^2	0.048 m^2
Material	-	EPS
Membrane charge	-	Neutral
pH resistance range	-	4-11

Temperature, dissolved oxygen (DO), pH, and discharge level were monitored using control instruments in both reactors. The operating temperature was regulated in the range of 22-27 °C via a heater. Fine and coarse plastic tube diffusers were installed at the bottom of the reactors to provide sufficient DO for the microbial activity and prevent the permeate flux reduction of the membrane, respectively. Two pilots were run for 43 days with a hydraulic retention time (HRT) of 8 h and permeate flux of 83 Lm^{-2} h^{-1}. Moreover, transmembrane pressure (TMP) was continuously measured using an analogue pressure gauge. Throughout the experiment, no chemical cleaning was implemented. The membrane biomass was removed daily only after its concentration reached approximately 2000 mg/L, and the solid retention time (SRT) was set to 25 days.

2.3. Analytical methods

The quality of influent and effluent wastewater in the pilots, activated sludge, and mixed liquor were studied utilizing the Standard Methods characterized by the APHA [33]. In this regard, total suspended solids (TDS), total dissolved solids (TDS), chemical oxygen demand (COD), total nitrogen (TN), total phosphorus (TP) as well as four heavy metals (Pb, Cu, Ni, and Cr) were examined. MLSS and COD were measured via Whatman glass microfiber filter (APHA 2540E) and Hach COD reactor (APHA 5220C), respectively. Additionally, an atomic absorption spectrometer was employed to determine the concentration of the heavy metals. In order to reach a desirable amount of MLSS concentration in the initial phase (up to 2000 mg/L), the MBR pilot was run for almost five weeks. After that, sludge was discharged to fix the MLSS concentration in the reactor. The performance of each reactor in terms of the removal efficiency for SS, COD, TN, TP, and heavy metals was determined from the following expression:

$$\%R = \frac{(C0 - C1)}{C0} \times 100 \tag{1}$$

Where R is the total removal efficiency, C0 is the total concentration in influent, and C1 is the total concentration in the effluent. During the investigation, DO and PH were measured by an oxygen electrode (JENWAY-970, England) and a portable pH meter (JENWAY-370, England), respectively. Furthermore, the temperature of the mixed liquor, also the inner and outer of the reactors was obtained via a digital temperature probe (JENWAY, England).

Fouling potential as an indicator of the RO influent was evaluated by the use of Silt Density Index (SDI) analyses. As stated by ASTM D-4189, SDI can be calculated when water is passed through a standard 0.45 µm pore size microfiltration membrane with a given pressure of 30 psi at a constant temperature. The difference between the required time to filter 500 mL before and after forming the silt indicates the SDI value, which is achieved from the following equation:

$$\mathrm{SDI} = \frac{(1 - \frac{t_1}{t_2})}{\mathrm{T}} \times 100 \qquad (2)$$

Where t_1 and t_2 are the time required to filter the first and the second 500 mL sample, respectively, and T is the elapsed filtration time (normally 15 minutes) after the beginning of collecting the first 500 mL sample.

3. Results and discussion

3.1. Removal of COD, TSS and TDS

The comparison of the MBR and CAS/UF performance in terms of the COD, TSS, and TDS removal is indicated in Fig. 3(a-c). Since the values of the concentrations were highly extended, they were plotted on a semi-logarithmic scale. COD, which has been considered the organic pollution indicator in wastewater, was examined throughout the investigation and was used to measure suspended and dissolved organic and inorganic matters. The inlet COD during the study was detected in the range of 661 to 779 mg/L with an average of 723 mg/L, while this value in the CAS/UF and MBR effluent varied from 94 to 125 mg/L and 68 to 95 mg/L, respectively (see Fig. 3(a)). Indeed, the average COD removal efficiency of the MBR process was approximately 88%, which is higher than the CAS/UF average elimination rate (around 83%). Similarly, Laurinonyte *et al*. reported that the COD removal in both the CAS-UF and MBR was very high from the first days of operation [34]. Also, the higher MBR COD removal effectiveness indicated that, compared to CAS/UF, this system was more capable of eliminating organic constituents [32]. In other words, long sludge ages of the MBR system prevented wash-out of slow-growing microorganisms and resulted in providing enough time for the biomass to adapt to different substrates and environments [35, 36].

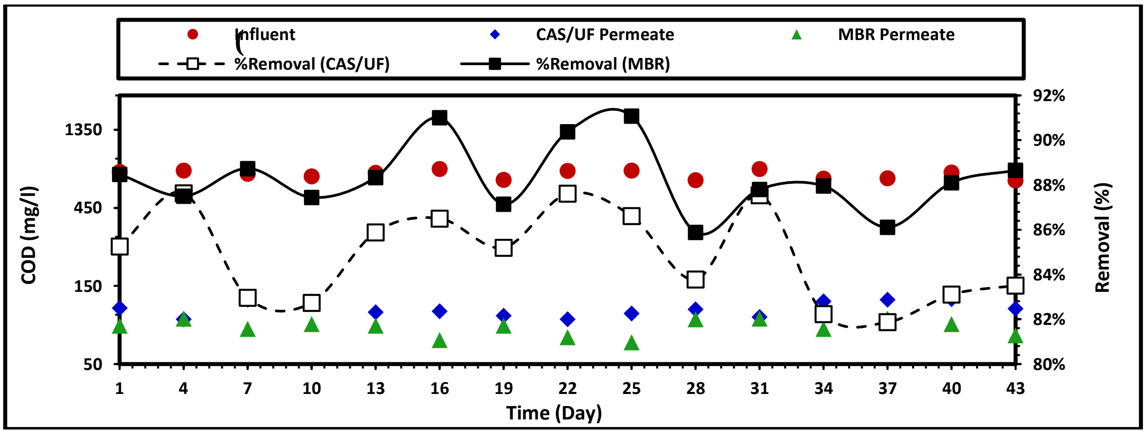
Some other researchers represented a COD removal rate of more than 90% using the MBR technology [32,37], which was higher than the results of the present work. In fact, a significant amount of biodegradable materials was removed in the previous biological processes (aerobic and anaerobic processes); hence, less concentration of organic matters in the reactors led to a lower COD elimination.

Another important factor of the operational behavior in biological wastewater treatment plants (WWTPs) is the TSS concentration. Fig.3 (b) illustrates the inlet, outlet, and the removal efficiencies of the TSS versus the days of procedure in both reactors. As can be seen, the influent concentration of the TSS ranged from 167 to 194 mg/L, and its average was almost 180 mg/L throughout the experiment. This concentration decreased significantly to less than 3 and 2 through the CAS/UF and MBR process, respectively. This result proves that the separation of bio-solids via membranes is independent of the flocculation and settling capacity of bio-sludge.

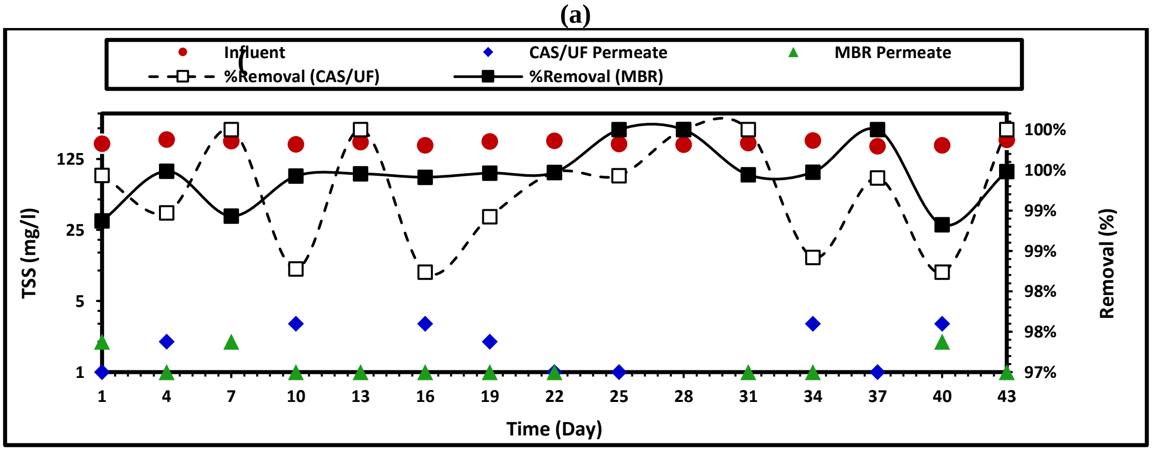
Comparing the MBR and CAS/UF in terms of suspended solids revealed that the performance of the MBR in producing effluent with less suspended solids was more suitable. Indeed, MBR can provide a low TSS concentration, which meets the wastewater discharge standards without requiring filtration. Meyo et al. used an expanded granular sludge bed reactor (EGSB), as a biological pretreatment stage, and a membrane bioreactor (MBR) to treat poultry slaughterhouse wastewater (PSW) [38]. According to the results, the MBR process could increase the removal performance of the TSS and COD up to 95%.

Total dissolved solids (TDS), which refers to the inorganic salts and small amounts of organic substance present in water solution, was also analyzed. As seen in Fig.3 (c), inlet TDS varied from 2645 to 4426 mg/L, and its removal efficiency was around 48% and 52% in CAS/UF and MBR, respectively. With regard to the membrane pore size, some parts of the dissolved solids could not be trapped behind the membrane and emerged in permeate. That is why the elimination rates of TDS were noticeably lower than TSS in both systems.

The mean concentration of the influent and effluent COD, TSS, and TDS elimination efficiency during the operational time are summarized in Table 3. In this period, the average influent of COD, TSS, and TDS concentrations were approximately 723, 180, and 3470 mg/L, respectively. It is worth noting that, throughout the experimental phase, the MBR was able to remove more than 88%, 99%, and 52% of COD, TSS, and TDS, respectively. Comparing these rates with the performance of the CAS/UF process indicated that the MBR can be more suitable than the CAS/UF for wastewater treatment. The obtained results were in line with those reported by Hashemi and Khodabakhshi, who stated that more than 99.9 % of solids were removed with micropore membrane, which mainly contains colloidal solids [39]. Furthermore, added UF filtration after CAS increased the COD and TSS removal by up to 15% and 38%, respectively (see Table. 3). Indeed, such increases in removal efficiency were probably due to adsorption to particulate materials and their removal on the UF membrane surface rather than biodegradation [36].



(a)



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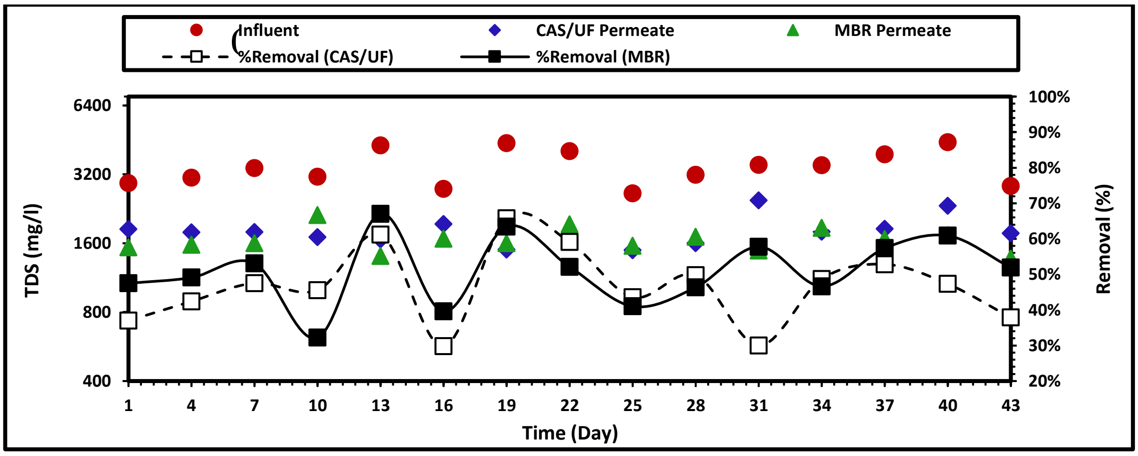


(c)

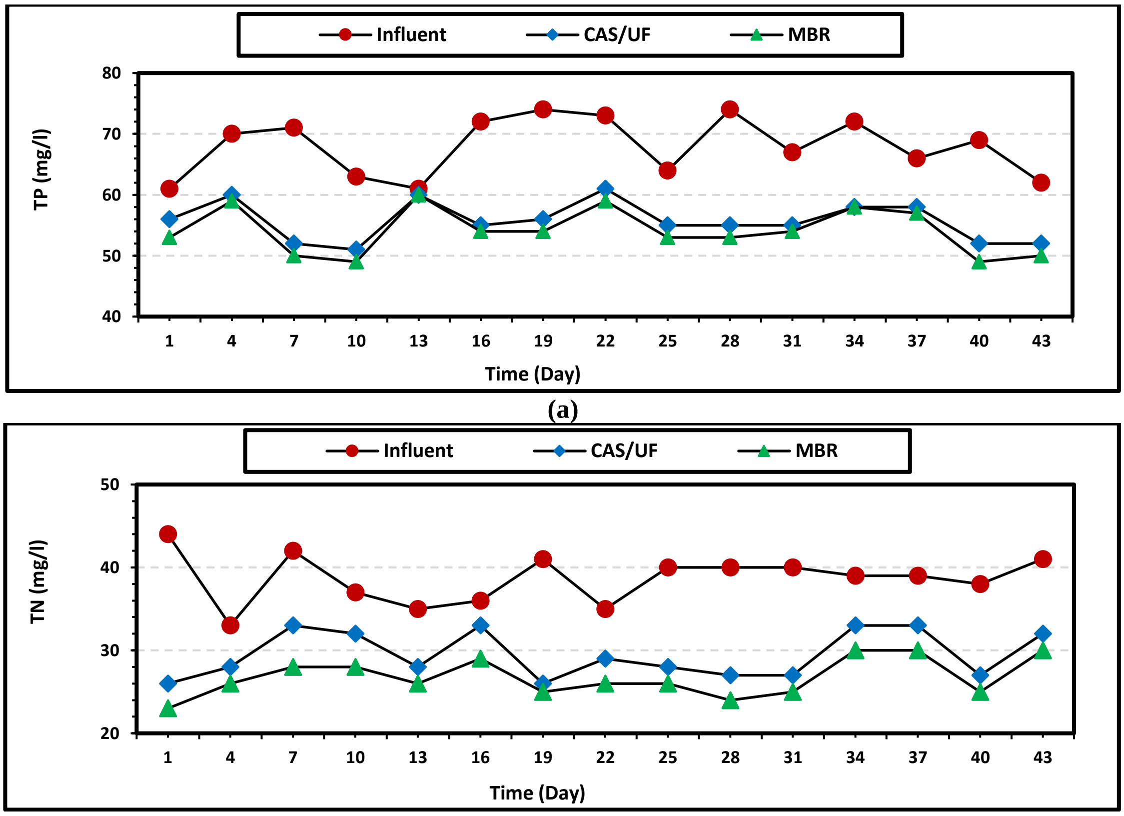
Figure 3. Comparison of (a) COD, (b) TSS, and (c)TDS concentrations in the feed and the effluent of CAS/UF and MBR versus the time of the test and their removal percentage.

3.2. Removal of Nutrient

The performance of the TP removal efficiency in the MBR was compared with that of the CAS/UF process. The TP concentration in influent varied from 61 to 74 mg/L during the 43-day investigation (see Fig. 4(a)). In the CAS/UF and the MBR permeate, this parameter was found to be from 51 to 61 and 49 to 60, respectively. Meanwhile, the average TP concentration in the inlet was 67.9 mg/L, which shifted to 55.7 mg/L at the CAS/UF outlet and 54.1 mg/L at the MBR effluent procedure. That is, nearly 18% and 20% of the phosphorus was eliminated by the CAS/UF and MBR (see Table 3).

The influent TN concentration within the investigation was detected in the range of 33 to 44 mg/L, which decreased to 26 to 33 mg/L in the effluent of the CAS/UF process and 23 to 30 mg/L in the MBR permeate (see Fig. 4(b)). In other words, as shown in Table 3, the mean concentration of the influent TN in the period of the experiment was 38.6 mg/L, which reached 29.4 and 26.7 mg/L in the CAS/UF and MBR effluent, respectively. The reduction in the nitrogen concentration during the operation of the reactors can be justified by the presence of the organic constituents hydrolysis and cell disintegration [32], which results from the process of nitrification and denitrification [40]. Likewise, the mean removal rate of the TN was obtained almost 24% and 31% in the CAS/UF and MBR processes. In another study, two lab-scale MBR and CAS systems were compared in terms of the removal efficiency of wastewater pollutants. The results indicated that the MBR's performance in reducing the contaminants in raw water was superior to that of CAS [41]. Additionally, Similar results have been reported by Laurinonyte *et al*., who used MBR as an alternative to CAS/UF for the Fischer–Tropsch process [34]. They indicated that the removal rate of the TN and TP for both treatment systems was more than 82%.

It should be noted that the main reason for the elimination of the nitrogen and phosphorous is the biodegradation of such nutrients by microorganisms in aerobic and anaerobic tanks and their separation through the membrane. Since this study used neither the anaerobic tank nor anoxic process, and given that the wastewater used in this study was the effluent of the activated sludge (before the disinfection process), the removal rate of the TP and TN was lower than those of other investigations [1,35,42].



(a)

(b)

Figure 4. Concentration of (a) TP and (b) TN in the feed and the effluent of CAS/UF and MBR versus the time of the test.

Table 3. Average concentration of the analytical parameters in the CAS/UF and MBR permeate throughout the wastewater quality characterization.

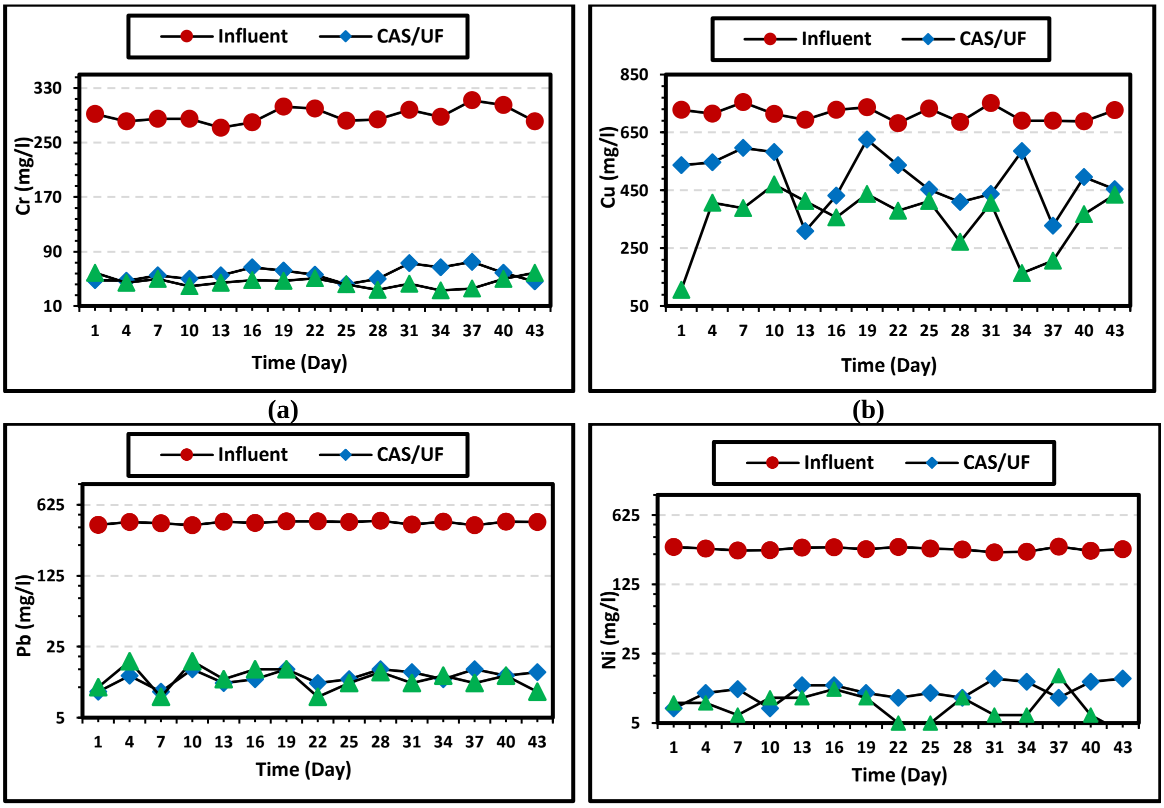
			CAS		CAS/UF		MBR	
Parameters	Unite	Influent	Effluent	Removal (%)	Effluent	Removal (%)	Effluent	Removal (%)
COD	mg/L	723.4	221.6	69.3	109.1	84.9	84.2	88.3
TSS	mg/L	180.5	71.5	60.3	1.3	99.2	1	99.4
TDS	mg/L	3469.3	1764.5	49.1	1811.1	47.7	1655.6	52.2
TN	mg/L	38.6	33.6	12.9	29.4	23.8	26.7	30.8
TP	mg/L	67.9	58.8	13.4	55.7	17.9	54.1	20.3
Cr	mg/L	289.8	236.1	18.5	56.8	80.4	45.2	84.4
Cu	mg/L	714.3	574.3	19.5	488.2	31.6	348	51.2
Pb	mg/L	416.9	225	46.1	12.3	97.1	12.4	97.2
Ni	mg/L	283.6	155.2	45.2	10.6	96.2	7.7	97.2

3.3. Removal of heavy metal

Toxic metals transfer from water resources to the human body may lead to a high risk of diseases; consequently, the elimination of toxic metals should be considered as an important way of increasing the safety and sustainability of water and wastewater [43]. The concentration of heavy metals in the inlet and outlet of both processes during 43 days of the experiment is displayed in Fig. 5 (a-c). It can be seen that the CAS/UF and MBR could reduce Cr, Pb, and Ni concentrations properly; nonetheless, the inlet concentration reduction of Cu was not significant. Meanwhile, the influent Cr concentration was between 272 and 312 μg/L, which decreased in the range of 42 to 75 μg/L and 33 to 59 μg/L in CAS/UF and MBR effluent, respectively. Moreover, the inlet concentration of Pb was observed from 393 to 435 μg/L, which was extensively declined via CAS/UF and MBR to the values of 9 to 15 μg/L and 8 to 18 μg/L, respectively. Similarly, 263 to 300 μg/L of Ni in influent shifted to the range of 7 to 17 μg/L in CAS/UF and 4 to 15 μg/L in MBR. According to an investigation, metal competition, variations in pH, and SS concentrations, plus changes in inlet metal concentrations are the main reasons for fluctuation in heavy metal removal efficiencies.[44]

As mentioned previously, the removal efficiency of Cu in both systems was lower than the other heavy metal throughout the experiment. On average, the MBR and CAS/UF could reduce Cu from 714 μg/L to 488 and 384, respectively (see Table 3). In other words, the mean removal rate of Cu for the CAS/UF was almost 31% and for the MBR was 51%. On the contrary, the average elimination efficiency of Cr, Pb, and Ni in the CAS/UF process were 80.4%, 97.1%, 96.2%, respectively, and they were 84.4%, 97.2%, and 97.2% for MBR, respectively.

It can be concluded that both of the utilized systems could perfectly remove Cr, Pb, and Ni, which, in turn, indicated that these metals were mainly in particular form. Indeed, by attaching to the sludge flocs, metal ions were retained via the membrane. Conversely, since some part of Cu that existed in soluble form could pass through the membrane; therefore, its concentration in CAS/UF effluent and MBR permeate was relatively more than the other metals. This phenomenon is in agreement with that reported by Hashemi *et al.*, who indicated that Cu had lower removal efficiency, in comparison with Pb and Cr, due to higher solubility [45]. Nonetheless, the membrane technology has established its effectiveness in terms of metal removal from wastewater owing to low energy consumption and high selectivity [46].



(a) (b)

(c) (d)

Figure 5. Concentrations of (a) Cr, (b) Cu, (c) Pb and (d) Ni in the influent and the effluent of CAS/UF and MBR versus the operation time

3.5. pH variation

As depicted in Fig. 6, the average pH values of influent varied from 7 to 7.9, which reached 6.8 to 7.4 and 7 to 7.5 in CAS/UF and MBR permeate, respectively. Such reduction in effluent pH can be illuminated by removing alkalinity which leads to a reduction in the buffering capacity. The following reversible equation between CO2, HCO_3^-, H^+, and CO_3^{2-} is expected when CO_2 dissolves in water.

$$H_2O + CO_2 \leftrightarrow H_2CO_3 \leftrightarrow H^+ + HCO_3^- \leftrightarrow 2H^+ + CO_3^{2-} \quad (3)$$

Values of these compounds depend on the pH level. For instance, in pHs more than 7, HCO_3^- and CO_3^{2-} are dominant. Since some parts of the dissolved ions were trapped via the membrane, the concentration of HCO_3^- and CO_3^{2-} decreased which led to the production of more H^+. As a result, the mean value of the outlet pH was lower than the feed. Similarly, Qin *et al*. stated that in pHs higher than 4.3, HCO_3^- tended to be removed by the membrane [47]. In fact, H_2CO_3 can be assumed unchanged when CO_2 is not rejected from the system; ultimately, higher H^+or lower pH would be expected in permeate compared to the feed in order to obtain a constant PKa (Ka = $[HCO_3^-] \times [H^+] / [H_2CO_3]$). However, when PH is lower than 4.3, H^+can attach to the membrane resulting in lower protons or higher PH in the effluent.

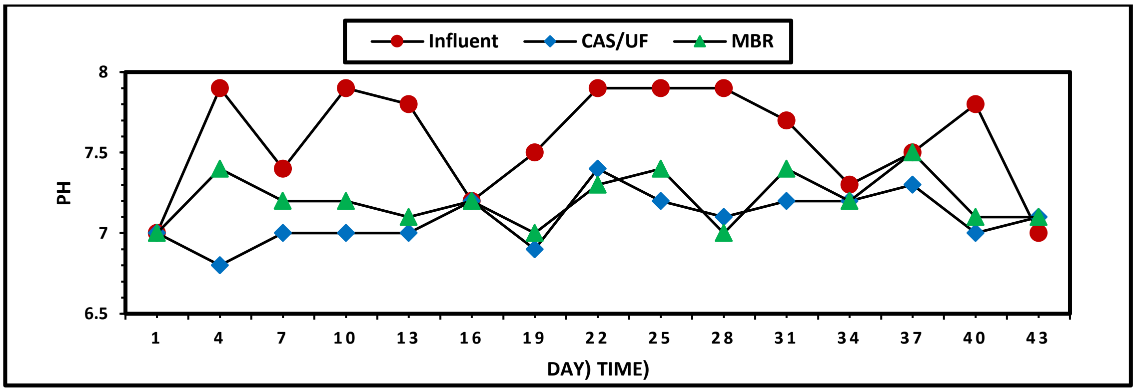


Figure 6. pH variation in the influent and effluent CAS/UF and MBR during the experiment time.

3.6. Silt Density Index (SDI) analysis

In order to assess the water quality of RO influent, silt density index was evaluated on the CAS/UF and MBR permeate. Although RO membrane manufacturers generally recommend an SDI of less than 3, the membranes usually foul at an acceptable rate in the SDI of 5 or less. Based on the previous studies, high levels of colloidal matters, micro-air bubbles, and organic substances have been reported as the main reasons for increase in SDI [48].The SDI values for both module effluents can be seen in Fig. 7. The permeate SDI values for the two systems were less than 5 for all the samples and were below 3 most of the time. The mean SDI values for the CAS/UF and MBR effluent were 2.8 and 2.46, respectively. Hence, CAS/UF and MBR were able to produce excellent water quality, which would be applicable for an RO process.

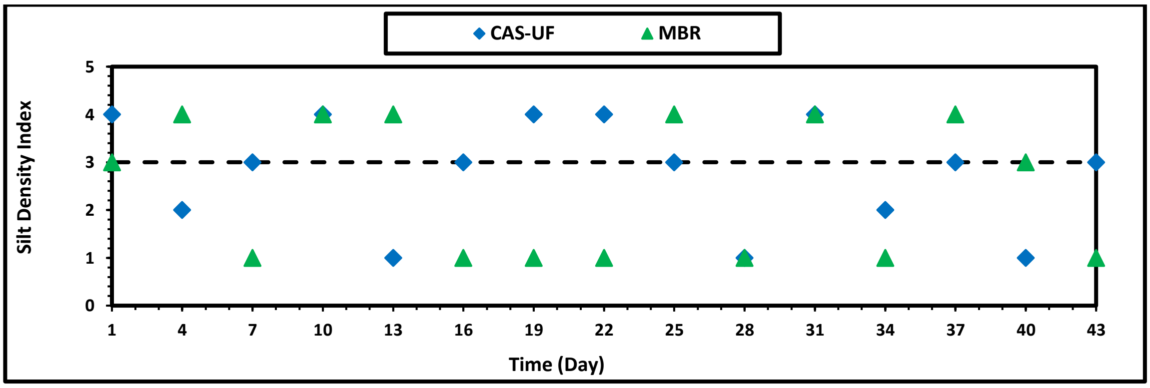


Figure 7. Comparison of SDI value for permeate water between CAS/UF and MBR.

4. Conclusions

In this study, the efficiency of MBR to purify effluent of an industrial wastewater treatment plant was evaluated and compared with that of CAS/UF process throughout around 6 weeks. Effluent water from both reactors has a removal rate of more than 99% for TSS, which demonstrated the application of the membrane technology in solids separation. Also, MBR increased the removal efficiency of the COD, TDS, TN, and TP by 3.4%, 4.5%, 6.5%, and 2.4%, respectively, compared to CAS/UF. The average removal percentages of Pb, Ni, and Cr were high and more than 80%, indicating that these two metals existed in particulate form, while Cu which was in soluble ionic or complex form was not removed completely (almost 32% and 51% by CAS/UF and MBR, respectively). Although the SDI values of MBR were lower, both modules could meet the influent requirement of RO pre-treatment. Further research would be needed to compare the MBR and CAS/UF systems with regard to the technical and economic aspects. Moreover, it is worthwhile to investigate the effects of organic load rate (OLR), hydraulic retention time (HRT), and solids retention time (SRT) on the performance of the processes over longer periods of time.

Acknowledgements

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