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Treatment of oil production wastewater by membrane bioreactor

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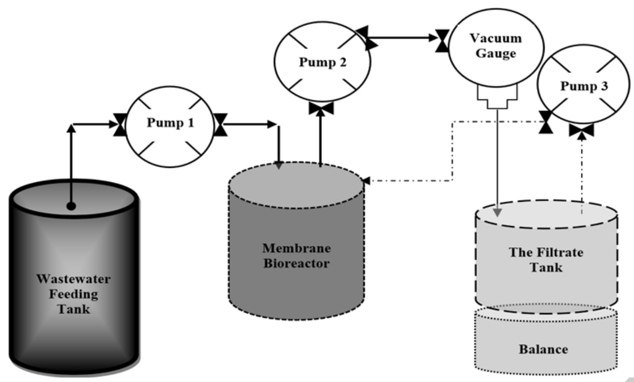
Graphical abstract



Abstract

Wastewater generated in oil production and refinery industries contains high levels of pollutions. Water from the oil production field is the largest source of wastewater in the oil production and refinery industry. It can cause environmental problems on a global scale due to its high volume and pollutant properties. The treatment of produced water and refinery wastewater is traditionally carried out by physical, chemical, and biological treatment processes. In recent years, it has been focused on the use of membrane technologies. In the treatment of oil production wastewater, membrane bioreactors are used as pre-treatment and/or direct treatment methods. In this study, the treatability of wastewater generated in the oil production field by membrane bioreactor was investigated. The system has been operated for 70 days and its efficiency has been evaluated. Treatment parameters such as chemical oxygen demand, total solids, total nitrogen, total phosphorus, Alkalinity, and conductivity were analyzed. In addition, flux and transmembrane pressures were measured. Result, chemical oxygen demand, total solids, total volatile solids, suspended solids, total phosphorus, total nitrogen and sulfate, average removal efficiencies 65%, 65%, 58%, 73%, 70%, 50%, and 57% was obtained, respectively. It has been determined that the system used can be used in petroleum production wastewater treatment.

Keywords: Membrane bioreactor, oil production wastewater, wastewater treatment, flux, pollution removal

1. Introduction

The modern lifestyle depends on a reliable source of energy. of the various energy sources, fossil fuels have been the least expensive for over a century and remain the primary source of energy for humankind today (Adham, 2015). Deriving the etymology of the word, the word "petroleum" is derived from the Latin, petro (rock) and oleum (oil), meaning oil from rocks. The first oil wells were drilled in the United States of America (USA), and the demand for oil has continued to grow since the late 1850s when Edwin Drake drilled the first oil well (Coday *et al.*, 2014). It is estimated that the daily global consumption of oil will increase from 95 million barrels per day to 106.6 million barrels by 2030 (Alzahrani *et al.*, 2014). Oil and gas are an important source of energy and income for many countries today. Its production was described as one of the most important industrial activities of the twenty-first century (Li *et al.*, 2017). The rapid growth of the oil and gas industry is closely related to the vital role of industrial civilization in its current structure and maintenance (Diya'uddeen *et al.,* 2011). Almost all economic sectors are highly dependent on oil and/or gas. Regarding the energy problem, many countries around the world still rely on oil-based sources as their main source of electricity and transportation fuels (Abas *et al.*, 2015; Igunnu *et al.,* 2012). This ever-evolving industry has been a concern for many countries, mainly because their operations generate large quantities of liquid waste (Guerra *et al.,* 2011). Despite its importance, oil is produced in large quantities from waste, accounting for more than 80% of wastewater and up to 95% in old and mature oil fields. In general, the oil/water volume ratio is 1: 3 (Nasiri *et al.*, 2017; Houcine, 2013). As a result, all wastewater generated during oil and/or gas exploration, production, and treatment is called produced water or treated water, and the produced water is the largest wastewater generated from this activity (Yu *et al.*, 2013; Schnabel *et al.,* 2013). Oil field wastewater or produced water contains various organic and inorganic components, and the discharge of produced water can pollute the surface and groundwater, and soil (Igunnu and Chen, 2012; BP energy outlook energy, 2017).

Today, the world faces some problems including environmental pollution, the demand for drinking water, and new energy sources (Saad *et al.,* 2016). Industrial wastewater treatment is an important field of study in environmental engineering. Treating wastewater from the oil and petrochemical industries is an effective option to "purify and treat produced water" (Al-zahrani *et al.*, 2014; Kargari *et al.,* 2014). The wastewater from oil production contains a high percentage of pollutants like oil, salt, phenol, etc. Their treatability with membrane systems is important in terms of enabling both wastewater treatment, reuse, and/or reuse (Reynolds *et al.,* 2012). When this wastewater is given directly to the receiving environments, it pollutes the water of the receiving environment and causes severe damage to the aquatic organisms (Abadi *et al.,* 2011). In addition, water consumption is high in oil production. Wastewater reuse is important. Therefore, treatment definitely is required. The main purpose of the treatment is to separate the oil from the wastewater and then apply the demineralization process (Fakhru Al-Razi *et al.*, 2009). According to studies, the concentrations of 16 polycyclic hydrocarbons (PAHs) included in the EPA range in petroleum wastewater production range between 0.7 - 100 mg/L (Abas *et al.*, 2012). In addition, current separation technologies, especially in offshore operations, cannot fully meet oil and grease removal to meet legal limits. In some of these cases, chemicals are used in the treatment processes however, some of these chemicals cause toxic effects. (Abas *et al.*, 2012). (Seureau *et al.,* 2013) examined the biological treatment of wastewater from oil production. The production wastewater was treated with an Aconiti kusnezoffii radix (AKR) system. They reported that biological treatment is not sufficient to treat this wastewater due to its high salinity. As a result of the study, it is understood that new treatment techniques are needed. (Abadi *et al.,* 2011) used the α-Al_2O_3 microfiltration (MF) ceramic cylindrical system to treat typical oily wastewater. They reported that the amount of filtered grease oil in the reactor system decreased by 4 mg/l. This value provided the required discharge parameters. In addition, the effects of operating parameters such as trans membrane pressure (TMP), cross flow rate (CFV) and temperature on permeability, TOC removal efficiency and fouling resistance (FR) were examined. A backwash was used in the system to remove oil droplets and particles that clog the membrane pores and the results showed that backwashing could significantly prevent the filtration flow reduction.

Abbasi *et al.* (2012), applied a tubular ultrafiltration (UF) unit fitted with modified polyvinylidene fluoride (PF) membranes with nano-sized inorganic alumina particles to remove oily wastewater from the oil field and analyze the membrane water permeability to treat ultrafiltration. The results showed that after UF treatment, the oil content is less than 1 mg/L, the suspended solids content is less than 1 mg/L, and the solid particle diameters are less than 2μm. The results showed that the addition of alumina nanoparticles improved the anti-fouling performance of the membrane and the flow recovery rate of the modified films reached 100% washing with 1% wt. surfactant solution OP-10 (pH 10).

Fakhru'l-Razi *et al.* (2009) reported that re-injection of wastewater from oil-producing areas to the field with low osmotic pressure does not improve water quality. A pilot plant was designed with an aeration tank, air float, sand filter, and UF and the performance characteristics of the hybrid process were examined for treating oily wastewater. As a result of the study, a high rate of solid removal was determined. In addition, it was determined that the iron and sulfate contents were met in the exchange standards. In this study, results such as COD removal and salt removal efficiency are absent. (Pearson *et al.,* 2014) used a PVDF-Al_2O_3 (cross-flow process type) membrane to treat oilfield wastewater. After treating the membrane, the flow was increased to two times by improving the permeability performance, and the oil and suspended solids content was 1 mg/L. It was observed that the COD and TCO removal efficiency were 90% and 98%, respectively, and that the filter water quality standard, oilfield injection/drainage was met. (Zhang *et al.,* 2009) applied polysulfone to treat oily wastewater. The results showed 99.16% oil retention and oil concentration at 0.67 mg/L permeability, which meets environmental discharge requirements (<10 mg/L). It can be concluded that the composite membranes developed in the study are highly contamination resistant, thus the developed The Polysulfone (PSF) membranes are applicable in treating oily wastewater.

All of the above shows that today there are many studies on the treatment of petroleum wastewater using membrane technology, but studies on treating petroleum wastewater using membrane bioreactors and studies that examine the behavior of membrane fouling are limited (Nasiri *et al.,* 2017; Wang *et al.,* 2015). Studies are needed on the feasibility of treating wastewater from oil production using membrane bioreactors. Reusing water obtained from oil production and processing and wastewater refining will reduce water consumption, provide economically added value and protect receiving environments and aquatic organisms (Padaki *et al.,* 2015; Silva *et al.,* 2017). Membrane bioreactor (MBR), an activated sludge process in which wastewater is biologically treated and biomass (activated sludge), has two stages comprising the membrane filtration process where it is separated from the treated wastewater with MF or UF membrane (Alley *et al.,* 2011). The use of a membrane bioreactor has some advantages over traditional methods of producing water treatment, which include better quality of wastewater compared to the conventional activated sludge process, lower energy cost, no chemical additives required, lower sludge production, high loading capacity, and high quality of treated wastewater. Few studies exist to treat wastewater from oil production with MBR (Kose *et al.*, 2012; Li *et al.*, 2017; Alsalhy *et al.,* 2016; Subramani *et al.,* 2015). MBR submerged membrane biofilter has been used in oil production and wastewater treatment in refineries (Ozgun *et al.,* 2013). As a result of the study, 80% removal of COD was achieved. However, there is not enough information about salt removal and other parameters and membrane contamination yet. Additionally, Wang *et al.,*

(2015) conducted a similar study. Here the MBR-UV and MBR-UV/H_2O_2 integrated systems were used. They aimed to investigate the photolysis performance supported by ultraviolet light and hydrogen peroxide (UV/H_2O_2) in order to reduce membrane contamination. 80% removal of COD achieved. It has been determined that the application of photolysis to ultraviolet light and hydrogen peroxide (UV/H_2O_2) increases flux and prolongs membrane use time. It has been determined that the MBR-H_2O_2/UV-NF system enables wastewater reuse for oil refining.

Studies are needed to remove solids, COD, salt, and similar substances. A study to explain this situation was conducted by Razavi and Miri (2015). In this study, the efficiency of the hollow fiber membrane bioreactor (HF-MBR) for treating oil refinery wastewater was examined. The submerged HF-MBR was operated for 160 days under various operating conditions. Chemical oxygen demand (COD), biological oxygen demand (BOD_5), total suspended solids (TSS), volatile suspended solids (VSS) and turbidity removal efficiency were 82%, 89%, respectively. They showed that 98%, 99% and 98% were reached.

This study aims to treat wastewater from oil production that contains high salt and COD content using a membrane bioreactor system. Thus, it is assessed that better-quality water has been obtained and reuse will be enabled. In addition, it aims to determine the elasticity of wastewater for oil production by membrane systems. The advantages and disadvantages of this system have been determined according to the classic treatment methods applied. It aims to determine the membrane model used and the treatment efficiency of the reactor system.

Table 1. Elementary properties of wastewater and inoculate used in experiments

Parameter/Unit	Concentration	inoculation culture
COD (mg/L)	988-2330	16000
pH	6.3-7.6	7.6
Alkalinity (mg $CaCO_3$/L)	290-445	2450
TS (mg/L)	810-1110	8100
TVS (mg/L)	660-888	6800
SS (mg/L)	180-250	7280
Oil-grease (mg/L)	40-90	
TN (mg/L)	360-495	270
TP (mg/L)	4-6	52
Sulfate (mg SO_4/L)	6.3-16.8	36

Table 2. Properties of membrane module in MBR

Membrane type	Hollow fiber – P5, Hydrophobic
Manufacturer	Zena Membranes
Pore diameter	0.1 µm
Membrane material	Polypropylene
Typical flux	150 l/m².h, 1 bar 15° C
Inner diameter/outer diameter	210/280 µm
pH resistance	2-11
Fiber shredding pressure	> 5.5 bar
Fiber breakdown pressure	> 3.5 bar
Effective module areas (cm²)	38

2. Material and method

2.1. Characterization of wastewater from oil production and inoculation culture

Wastewater was provided from petroleum production area that located in the Batman province in Turkey. The inoculum culture used in system was supplied from the digester tank of anaerobic treatment plant of Pakmaya which produce yeast in Amasya, Turkey (Table 1). It was mixed to the reactor system at a ratio of 1/3 inoculate/wastewater. At the end of the adaptation process, wastewater has been started to be fed**.** Inoculate VS value at the start of the reactor was 6800 mg/l.

2.2. System setup

A membrane bioreactor system was two liter and had made from Plexiglass was established for wastewater treatment (Figure 1). The system used the study includes wastewater feed tank, membrane bioreactor, peristaltic pumps, filtrate storage tank, a membrane module, a temperature indicator, a manometer pressure gauge, PVC pipes and compression connection and valves.

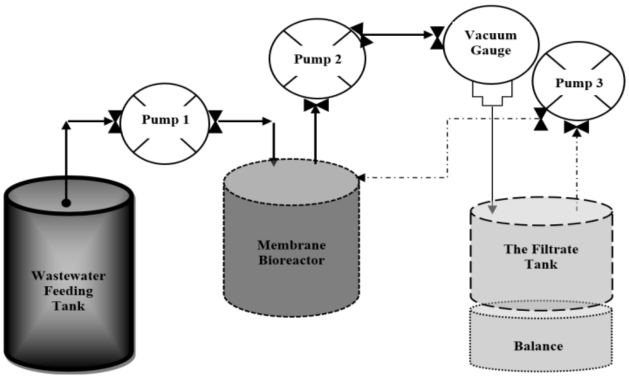


Figure 1. Shematic diagram of the system.

2.3. The membrane module

Membrane module used in the system was made from hollow fibre microfiltration. Its surface area was 38 cm². Membrane module features was given in Table 2. 15 hollow fiber units 30 cm long were cut and prepared by gluing the U-shaped ends together. A special dual-mix adhesive was used to bond the membrane unit (3M ™ Scotch-Weld ™ Epoxy Adhesive 3501).

3. Results and discussions

3.1. Change chemical oxygen demand COD and process efficiency

The membrane bioreactor was fed with oil production wastewater. The system was run for 70 days and the *COD* values were analyzed (Figure 2). The *COD* values of samples taken in flow from the reactor were found in the range of 535-1018 mg/L. It was determined that the *COD* value of the effluent was high for the first two weeks and then decreased. The reason for this increase is the grafting process applied in the MBR system. Additionally, no gas formation or production was observed during this process. According to the analysis results, approximately 50-70% *COD* removal efficiency was achieved with the membrane bioreactor system.

Razavi and Miri (2015) achieved 82% removal of *COD* in a similar study, while Adams et al. (2008) 85% of the *COD* account efficiency. Lower return was obtained in our study. This can be attributed to due to the intensity of membrane fouling and the high content of resolved salts in wastewater. Campos et al. (2002) reported similar removal efficiencies to our data in their study.

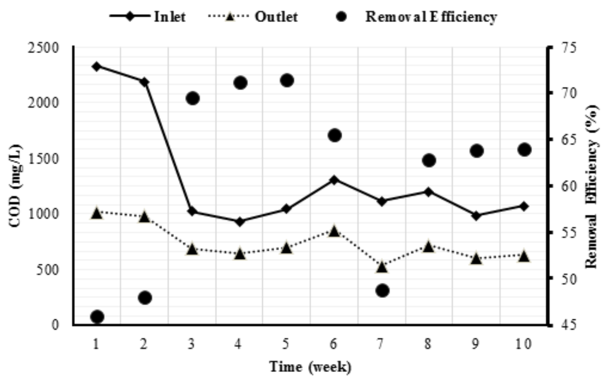


Figure 2. Time-dependent *COD* values and removal efficiency.

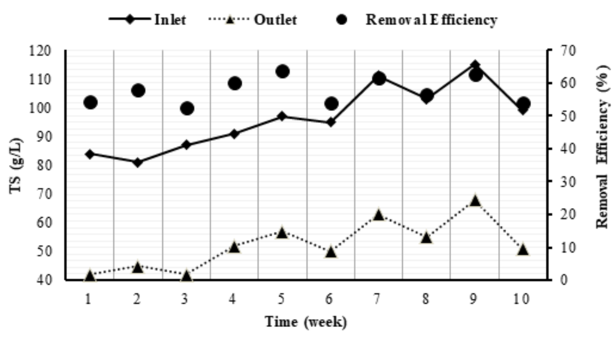


Figure 3. Time dependent *TS* values and removal efficiency.

3.2. Solids

The membrane bioreactor system was run for 70 days and the solids were analyzed (Figure 3). As a result of the analysis, the change of *total solids TS* expense values over time is shown in Figure 3. While the *TS* values measured in the raw wastewater had varied between 80-115 g/L, the *TS* values in the effluent decreased to the range of 42-63 g/L.

On the other hand, the total volatile solids *(TVS)* values changed over time as shown in Figure 4. While the *TVS* input samples analysis result was approximately between 66-88 g/L, the effluent analysis values were reduced to the minimum concentration level of 30-52 g/L. While in the MBR system increases the *TVS* rate and at the same time increases the bacteriological activities, it became clogged in a short time by causing the formation of a cake on the surface of the membrane. The removal efficiency rates for *TS* and *TVS* were 55-70% and 50-63%, respectively.

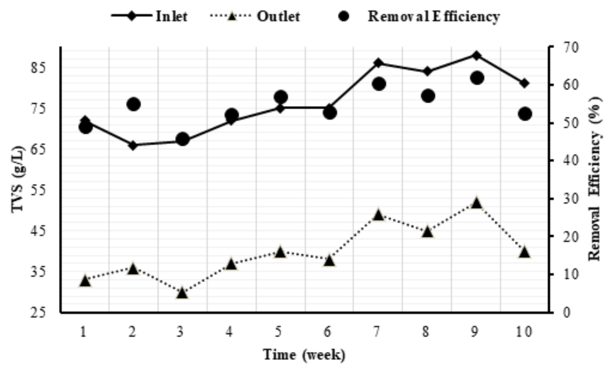


Figure 4. Time-related *TVS* values and removal efficiency.

In the conducted experiments, the change *in suspended solids (SS)* values over time was presented in Figure 5. While the SS values in feeding tank was 1.83-2.45 g/L, *SS* in the permeate reached the range of 1.25-1.6 g/L.

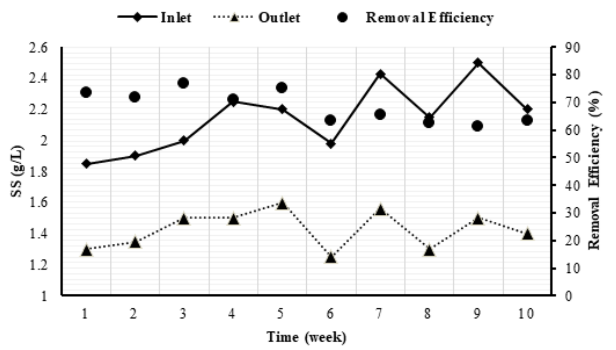


Figure 5. Time-related *SS* values and removal efficiency.

In addition, the concentrations of dissolved solids obtained to the change in over time during operation (as shown in Figure 6). According to the figure, while the total dissolved solids (*TDS)* concentrations in the intake wastewater were observed at levels of 78-93 g/L, *TDS* of liquid treated with the membrane system was recorded in the range of 60-77 g/L. This study showed that the majority of dissolved substances were found to pass through MF membranes. MF membranes are not available for removing dissolved solids.

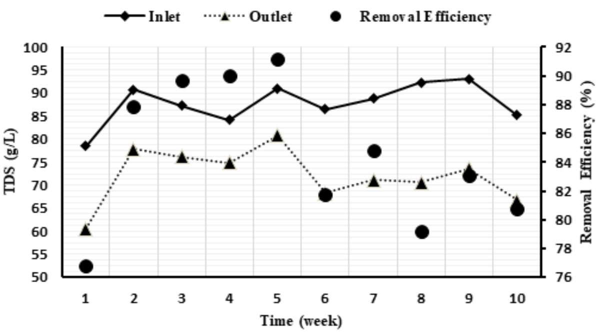


Figure 6. Time-related *TDS* values and removal efficiency

The concentration of *SS* in the filtrate is very low *SS* and *TDS* removal efficiency were about 65-90% in the MBR system (Figures 5 and 6). A significant change was observed in the concentration of dissolved solids. The majority of solids in permeability consist of solutes. This indicates that the membrane filters can pass almost any dissolved solids. It has been observed that the *TDS* removal efficiency was low, and the solutes are completely filtered by the micro-filtration bioreactor system. Razavi and Miri (2015) achieved more than 90% removal of solids in a similar study. Also, they obtained that dissolved salts removing is low.

3.3. Alkalinity and pH

The change of alkalinity values with time is shown in Figure 7. The alkalinity values of raw wastewater for oil production were measured between the range of 290-450 mg $CaCO_3$/ L. The alkalinity values of the reactor exit filter varied between 120-200 mg $CaCO_3$/L. Alkalinity was removed in the range of 36-50% in the system. Since the suspended solids that form alkalinity in the raw wastewater were kept in a filtrate, this removal had provided. The reason for the relatively low removal rate is the high *TDS* content.

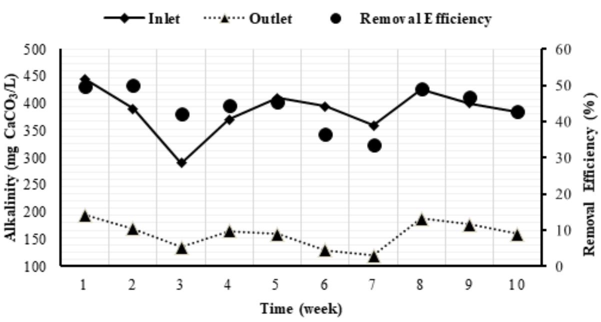


Figure 7. Time-dependent change of alkalinity values and removal efficiency.

In the first stage of the MBR system, while a *pH* of about 6.5-7.5 was measured in raw wastewater, over time the system began to produce alkalinity and the pH values increased in filtration outlet. Parallel to the working process of the MBR system, pH values were recorded in the range 7.7-8.9 (Figure 8). The reason for this increase is the occurrence of anaerobic glycolysis activities. This is also due to the higher alkaline materials compared to the weak volatile acids formed in the reactor system.

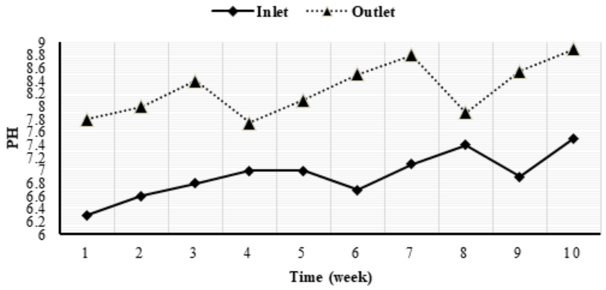


Figure 8. The *pH* values change over time.

3.4. Total nitrogen and total phosphorus

While the initial total nitrogen *TN* values for wastewater were in the range of 360-495 mg N/L, the *TN* values for the exit sample that passed through the HF membrane in the MBR system were measured in the range 165-240 mg N/L. The *TN* removal efficiency of the system was achieved with a removal efficiency of approximately 70% (Figure 9).

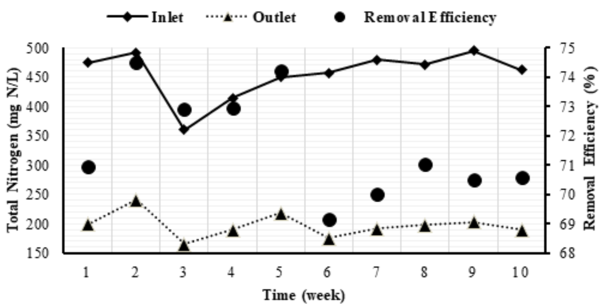


Figure 9. Change of total nitrogen values over time and efficiency of removal.

While the total phosphorus *TP* values of wastewater were in the range of 4-6 mg P/L, the values of 1-3 mg P/L were determined in the system filter. The *TP* removal efficiency of the system was obtained in the range of 40-60% (Figure 10).

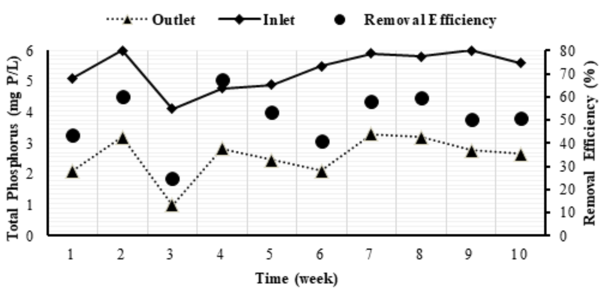


Figure 10. Change of total phosphate values over time and efficiency of removal.

The reason for the high removing of *TP* in the system compare to conventional anaerobic systems is as follows: there are types of microorganisms in the membrane bioreactor system and microorganisms remain in the system during the process. In addition, they need some material to maintain and maintain their reproduction, activity and activity, and because it is one of those materials, the *TP* ratio may cause a decrease in the *TP* ratio in the wastewater inlet, outlet.

3.5. Sulfate

As shown in Figure 11, the amount of sulfate concentration in wastewater from oil production is low. While the sulfate values for wastewater are in the range of 6-16 mg SO_4^-/L, the output values were reached 5-9 mg SO_4^{-2}/L. In the MBR system, the desulfurization rate in the filter water was recorded at 45-60 % within 70 days of operation.

Given the sulfate concentrations in the system, it is understood that the low controlled amount of sulfate in an anaerobic environment leads to the use of sulfate-reducing bacteria in some organic matter in the sulfate reduction stage, and the sulfate is used as an electron acceptor by the sulfate-reducing bacteria and the following bacteria have the property of anaerobic respiration.

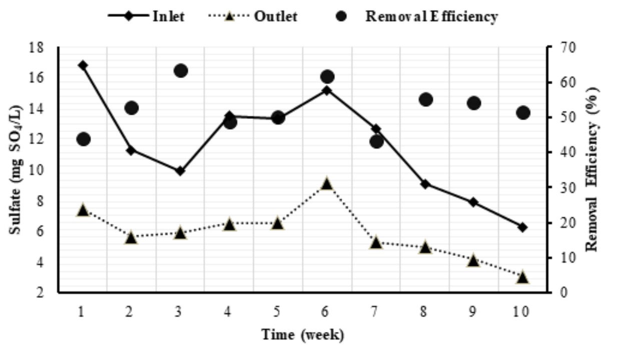


Figure 11. Changes of sulphate values with time and efficiency of removal.

3.6. Conductivity

The results of the seventy days analyze reflecting the conductivity values of oil production wastewater within the scope of characterization studies are shown in Figure 12. The conductivity values of raw wastewater were measured between 100-120 (µs/cm). The range of conductivity values for HF membrane exit filter water was recorded as 75-96 (µs/cm). Removal efficiency of 9-22% was achieved in the system. Since most of the salts that cause conduction dissolve, they have passed through the membrane. For this reason, a lower de-conductivity efficiency was obtained.

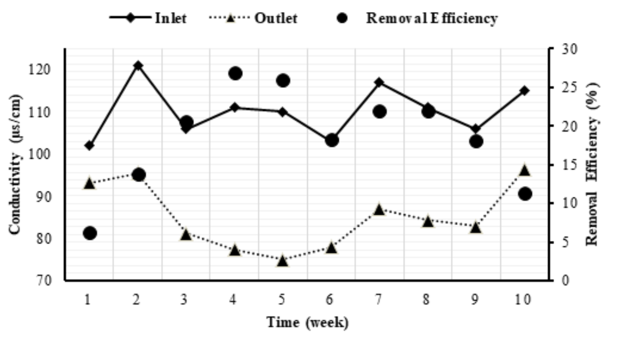


Figure 12. Change of conductivity values depending on time and efficiency of removal.

In general, there is a correct relationship between the conductivity value, dissolved solid and the salinity concentration in the wastewater content. With the exception of the first two weeks, a downward trend was observed in the important position and then no significant change was observed.

3.7. Flux and trans membrane pressure

The results of the measurement of flux and trans membrane pressure in the membrane bioreactor system are shown in Figure 13. In the reactor system, the flux was 3.12 L/m^2 at the start, decreasing and approaching zero on the 26th day. The transmembrane pressure increased in parallel with the decrease in flow. Contaminated components in wastewater, especially suspended solids, have caused membrane contamination, which reduces the flux of filtered water. On the 26th day the raw water was fed by changing the membrane and the situation was repeated. The membrane was altered twice in this process, and the behavior of the flowing and moving membrane was repeated with the same feature. Cake formation is observed on the surface of the membrane and contamination in a short time.

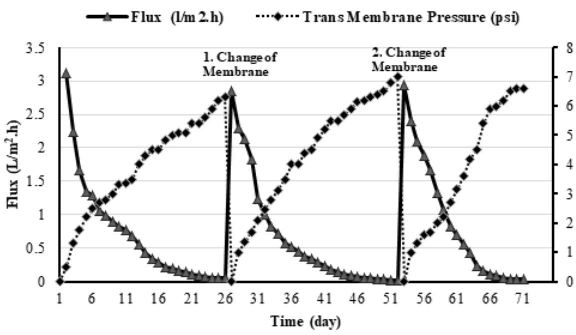


Figure 13. Time-related varying values of flux and trans membrane pressure.

In addition, a significant decrease in flux was observed for the first three to four days each time the membrane was changed. It is reported that the pores of most membranes quickly fill with particle and other components that begin to clog.

4. Conclusions and recommendations

In this study, the treatability of wastewater generated in the oil production field by the hollow fiber membrane bioreactor was investigated. The system has been operated for 70 days and its efficiency has been evaluated. Treatment parameters such as COD, solids, total nitrogen, total phosphorus, alkalinity, and conductivity were analyzed. In addition, flux and transmembrane pressures were measured. Result, COD, TS, MLVSS, SS, TDS, alkalinity, TP, TN and sulphate, average removal efficiencies 65%, 60%, 58%, 55%, 82%, 50%, 72%, 63%, and 59% was obtained, respectively. Also, the results for pH and conductivity values were measured in the range of (7.7-8.9), (75-96 µs/cm), respectively. It has been determined that the system used can be used in petroleum production wastewater treatment. As a result of this study, it was concluded that wastewater from oil production fields can be treated with microfiltration membrane bioreactor (MF-MBR), but it is insufficient for advanced treatment. In this case, by following the said system, UF etc., it is understood that high quality water can be obtained if membrane processes are used.

Membrane technology, which has various advantages, is increasingly used in treating produced water to meet many water quality requirements by replacing traditional technologies. However, fouling and membrane occlusion are a major disadvantage of the membrane processes that must be controlled in this application. Given the variety of characteristics of the produced water, it is imperative to conduct an investigation consisting of different techniques to ensure that the purpose-appropriate purification can be used. Generally speaking, there is still no perfect membrane, as each type of membrane provides unique properties and optimizes various parameters. The search for a membrane that will meet the needs of the industry with high efficiency and low cost continues, and the low flow is one of the major drawbacks preventing the application of large-scale membrane technology in the oil industry.

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