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The Study of Organic Removal Efficiency and Membrane Fouling in a Submerged Membrane Bioreactor Treating Vegetable Oil Wastewater

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The characterizations of vegetable oil wastewater (VOW) are unpleasant odor, dark color, and high organic contents, including large amounts of oil and grease (O&G), chemical oxygen demand (COD), fatty acids and lipids. Therefore, VOWs should be treated efficiently to avoid the environmental pollution. The aim of the present study was the investigation of VOW biological treatment using membrane bioreactor (MBR) in terms of organic pollutant removal performance and membrane fouling. During 30 days MBR operation at hydraulic retention time and solid retention time of 48 h and 20 days, respectively, there was a consistently low turbidity (<2 NTU) in the MBR effluent. The COD and O&G removal efficiency from the wastewater using the MBR were 85.0±1.3% and 82.7±1.4%, respectively. With decrease in aerobic metabolic activity and hence the activated sludge growth rate during the MBR operation, the MLSS and MLVSS decreased and led to accumulation of O&G and soluble microbial products (SMP) inside the bioreactor. The effluent COD value and the transmembrane pressure during the MBR operation remained in the range 88.7±11.5 mg L^{-1} and 1-2 kPa, respectively. The current study shows that the MBR has a very good potential for treatment of VOW, both in terms of removal performance and membrane fouling.

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1. INTRODUCTION

The extraction and processing of oils and fats from vegetable sources such as soybean, corn and sunflower are the important processes in vegetable oil industry. Crude vegetable oil refining contains degumming, neutralization, bleaching, deodorization and further refining. Refining of vegetable oils particularly by chemical methods, generates large volumes of wastewaters - approximately 10–25 m^3 per metric ton of product - which can be hazardous to the environment due to their high concentrations of organic and oily content [1, 2]. Depending on the processing technology applied and the raw material processed as well as its quality, the volume of wastewater may vary. The characterizations of vegetable oil wastewater (VOW) are unpleasant odor, dark color, and high organic contents, including large amounts of oil and grease (O&G), biochemical oxygen demand (BOD), chemical oxygen demand (COD), fatty acids, phenolic chemicals, pectins, lipids and a diversity of other pollutants [3-5]. Hazardous nature of O&G in oily wastewater causes significant problems to the soil, water, air and human beings [6]. Since discharge of poor quality final effluents has a negative effect on natural water sources, hence in order to limit the environmental impact, it is necessary to treat them to an adequate level before discharging to the receiving water bodies [3].

Jamaly et al. and Pintor et al. [2,6] reviewed the present and newly developed methods for the treatment of oily wastewater including primary, secondary and tertiary treatment such as electrochemical methods (electrocoagulation and electroflotation), membrane filtration (ultrafiltration (UF), microfiltration (MF) and reverse osmosis (RO)), biological treatment, hybrid

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technologies, adsorption (using adsorbents such as polypropylene, activated carbon, chitosan-based polyacrylamide and biosurfactants), ultraviolet radiation, and flotation and coagulation (use of zeolites and other natural minerals) [2, 6-8].

Physical or physico-chemical treatment methods for vegetable oil refinery wastewater have the major drawbacks of difficult sludge management and high chemical costs [4]. Also the soluble COD removal is poor in physicochemical treatment processes [9]. Therefore, biological methods are preferred because of the simplicity, low cost and compatibility with the environment.

The literatures show that the discharged fatty materials from food industries are easily biodegradable, and hence ready to be treated by biological methods [3]. Aslan et al. [10], observed that despite the high organic load in sunflower and corn oil wastewater, 93% and 96% of their total COD, respectively, usually contained biodegradable COD. Some authors have achieved complete COD removal using biological treatment by providing an upstream physicochemical unit of coagulation/flocculation and sedimentation or dissolved air flotation [11].

VOW can be treated biologically in aerobic, anaerobic or mixture of both conditions. Saatci et al. [12], observed that in an upflow anaerobic sludge blanket process, the COD and O&G removals were above 85% at organic loading rate (OLR) between 1.62 and 7.83 kg COD m^{-3} d^{-1} and hydraulic retention times (HRT) of 2 and 2.5 days. The activated sludge process (ASP) is extensively applied for the biological treatment of VOW [13]. Reddy et al. [14] reported that 81% of COD in wastewaters from a sunflower oil processing plant was removed using an ASP, at a sludge retention time (SRT) of 15 days and HRT of 24 h. However, in ASP, due to lack of filamentous growth and subsequent poor floc formation, high effluent total suspended solids (TSS) is a persistent problem. An amendment of ASP is sequencing batch reactor (SBR) which has been successfully applied for the biological treatment of VOW [13]. Mkhize et al. [15], used an anaerobic/aerobic SBR for the treatment of edible oil effluent. The influent COD and oils and suspended solids removal was 75% and 90%, respectively.

To meet the increasing stringent legislations on water quality and the need for water reuse/recycling, as well as the reduction of the wastewater disposal cost, more advanced wastewater treatment processes are required. The membrane bioreactor (MBR) is becoming a reliable technology for biological treatment of municipal and industrial wastewater [16, 17] and has a high potential for vegetable oil refinery wastewater treatment and oil removal. In MBR system which is a combination of biological treatment and filtration, the sedimentation tank of ASP is exchanged with a usually submerged membrane module. The benefits of MBR are high retention of biomass and particulate COD, low effluent turbidity and small footprint [18].

Nevertheless, currently the reduction in membrane performance caused by membrane fouling is the main issue in continuous operations that limits the extensive application of MBRs [19, 20].

To the knowledge of the authors there is only one previous report on the treatment and reuse of VOWs using MBR technology [4]. In that study VOW was treated using a submerged MBR, employing a hollow-fiber polyvinylidene fluoride (PVDF) membrane. The result showed that the pollutant removal performance of MBR was good. However, in that study as a result of complete sludge retention and therefore increase in MLSS concentration, membrane fouling was clearly discernible. As a result, for the implementation of MBRs treating VOW in large scale, detailed study on operational parameters and membrane fouling seems to be necessary.

The aim of the present study was the investigation of biological treatment of real VOW (taken from the wastewater treatment plant of Kourosh vegetable oil factory) using a MBR in terms of organic pollutant removal performance and membrane fouling at HRT equal to 48 h and a moderate sludge retention time (SRT) of 20 days. Furthermore, the MBR performance in treatment of VOW was compared with that reported with the combined system of conventional ASP and moving bed bioreactor (MBBR) used in Kourosh vegetable oil factory.

2. MATERIALS AND METHODS

2. 1. Vegetable Oil Wastewater and Sludge Used

The VOW samples used in this study were collected from the wastewater treatment plant of a local vegetable oil processing plant (Kourosh vegetable oil factory, Alborz, Iran). The sampling was carried out after the preliminary treatment which consisted of a CPI (corrugated plate interceptor) oil separator, bar screen, grit chamber, aeration, mixer 1 (alum + lime + polyelectrolyte), mixer 2 and sedimentation. The samples were then stored at 4 °C. All samples were equilibrated to room temperature before feeding. Analysis of the VOW was carried out several times and the average composition is shown in Table 1. The C/N/P ratio of the medium was adjusted to approximately 100:10:1 by the incorporation of suitable concentrations of NH_4Cl and KH_2PO_4 into the VOW. The activated sludge, used as inoculum for the MBR, was collected from the aeration tank of the Kourosh VOW activated sludge treatment plant.

2. 2. Experimental Set up and Operating Conditions

The bioreactor used in the present study, consisted of a 5 L acrylic tank, in which a PVDF flat-sheet UF membrane (Shanghai SINAP Membrane Tech Co., Ltd., China), with a pore size of <0.1 µm and area of 0.1 m^2 was immersed (Figure 1). Details of the setup and mode of operation have been described elsewhere [18]. The bioreactor temperature was kept constant and controlled by a heater. The pH of the MBR throughout its operation was kept within 6-8 through incorporation of 0.8 g L^{-1} $NaHCO_3$ in the VOW in the feed tank. The VOW was fed to the MBR via a feed tank equipped with a mixer rotating at 120 rpm. The values of operating parameters during the 30 days MBR operation are presented in Table 2. At the start of operation, the MBR system was inoculated with the activated sludge and the concentration of initial mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) were 4100±107 mg L^{-1} and 2162±53 mg L^{-1}, respectively.

2. 3. Chemicals and Analytical Methods All chemicals were of analytical grade and the product of Merck Company (Merck, Germany).

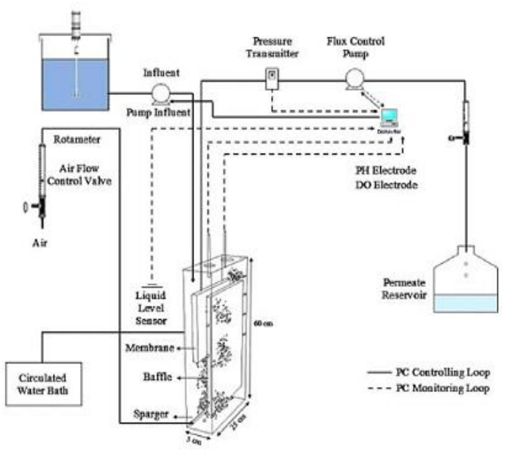

Figure 1. Schematic of the laboratory MBR setup.

TABLE 1. Physicochemical characteristics of VOW used in this work

Parameter	Concentration[a]	Parameter	Concentration[a]
pH (-)	6.9 ± 1	NO_2^{-} [b]	2.9
Turbidity (NTU)	54.1±16	PO_4^{3-} [b]	0.2
Conductivity (mS/cm)	3.3	SO_4^{2-} [b]	1460
COD	676±179	CL^{-} [b]	379
BOD	509±7	F^{-} [b]	6
TDS	2346	Mg^{2+} [b]	3.1
Salinity	2256	Na^{+} [b]	448
O&G	54.0±2.3	K^{+} [b]	7.7
NO_3^{-} [b]	5	Ca^{2+} [b]	442

[a] All mg L^{-1} unless otherwise stated.
[b] All ions were analyzed by ion chromatography (881 Compact IC pro 1, Metrohm, Switzerland).

TABLE 2. The value of operating parameters during the MBR operation.

Items	Values
SRT (d)	20
HRT (h)	48
OLR (Kg COD m^{-3} d^{-1})	0.35±0.09
COD (mg L^{-1})	676±179
O&G (mg L^{-1})	54.0±2.3
Temperature (°C)	30±1

The COD of the samples of the MBR mixed liquor, influent and effluent were determined according to closed reflux, colorimetric method (5220D) of APHA standard methods [21]. The BOD_5 value of the samples was measured using the BODTrakTM instrument of Hach Company. MLSS and MLVSS were determined according to methods 2540D and 2540E of APHA standard methods, respectively [21]. Total dissolved solids (TDS), pH, conductivity and salinity were measured with a Hach apparatus (Hach, HQ 40D). The O&G of the feed and effluent were measured according to the United States Environmental Protection Agency (USEPA) Method 1664, which is a modified version of 5520F in Standard Methods [21]. In cases where foaming and formation of water in oil emulsions occurred, the extract was centrifuged (15 min, 3200 min^{-1}) to separate the water and oil extract. For extraction of O&G of mixed liquor, 5 mL of mixed liquor sample was vigorously mixed with 15 mL of methylene chloride for 10 min. After 20 min the extracts were filtered through anhydrous sodium sulfate to remove water. Subsequently, the samples were dried at 70 °C and weighed on an analytical balance [22]. SMP and the protein and carbohydrate concentrations of the SMP (SMP_P and SMP_C) were determined as described previously [18]. The SMP_{total} was estimated as the sum of these two components. Specific oxygen utilization rate (SOUR) was determined according to Standard Methods [21]. All analysis was performed in triplicate.

TSS was also determined according to Standard Methods [21]. All turbidity measurements were performed with a portable turbidity meter (Aqualytic AL450T-IR). All ions of influent VOW were analyzed by ion chromatography (881 Compact IC pro 1, Metrohm, Switzerland).

2. 4. Statistical Analysis Statistical analyses of the experimental data, including one-way ANOVA was performed using Minitab version16 (Minitab Inc., State College PA, USA). Determination of the period in which steady state conditions were obtained during MBR operation was performed using one-way ANOVA according to the method described by Abdollahzadeh Sharghi et al. [18].

3. RESULTS AND DISCUSSION

3. 1. MLSS and MLVSS The variation of MLSS, MLVSS and the MLVSS/MLSS ratio during the operation of MBR at HRT and SRT of 48 h and 20 days, respectively, is presented in Figure 2. The results of one-way ANOVA analysis (not presented) showed that after day 22 and 25, respectively, steady state conditions with regard to MLSS and MLVSS were attained. According to the data presented in Figure 2, during the MBR operation the average value of MLSS and MLVSS concentration decreased from 4100±107 mg L^{-1} and 2162±53 mg L^{-1} to 1158±37 mg L^{-1} and 600±52 mg L^{-1}, respectively. The decrease in MLSS and MLVSS concentrations during the MBR operation was the result of the net growth rate falling below the rate of biomass removal that was necessary to maintain the SRT at 20 days. The corresponding MLVSS/MLSS ratio during MBR operation (Figure 2), was almost constant and average value was 0.52±0.05. Therefore, during the 30 days operation of the MBR there was no accumulation of inorganic matter inside the MBR.

In MBR studies, MLVSS concentration reflects the oil adsorbed onto the bacterial flocs as well as the bacterial biomass concentration, whereas MLSS reflects both the inorganic and organic content of the mixed liquor [17]. The low MLVSS/MLSS ratio indicates that a large portion of mixed liquor was inorganic matters. In fact, the use of coagulants in the pretreatment step increased the concentration of inorganic matters [11] in the influent VOW to MBR.

The mixed liquor O&G and COD during MBR operation is presented in Figure 3a. The concentration of SMP_{total} and its components (SMP_P and SMP_C) concentration in the mixed liquor inside the MBR during operation of MBR is also presented in Figure 3b. The trend of change of mixed liquor COD was variable during the MBR operation but an increasing trend was

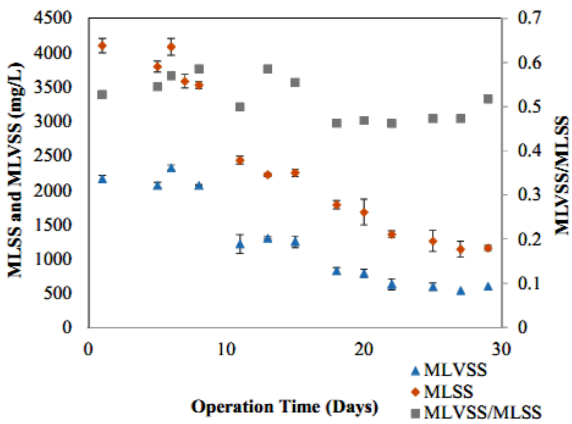

Figure 2. Variations of MLSS and MLVSS concentration and MLVSS/MLSS ratio during 30 days operation of the MBR. Error bars represent standard deviations of three replicates

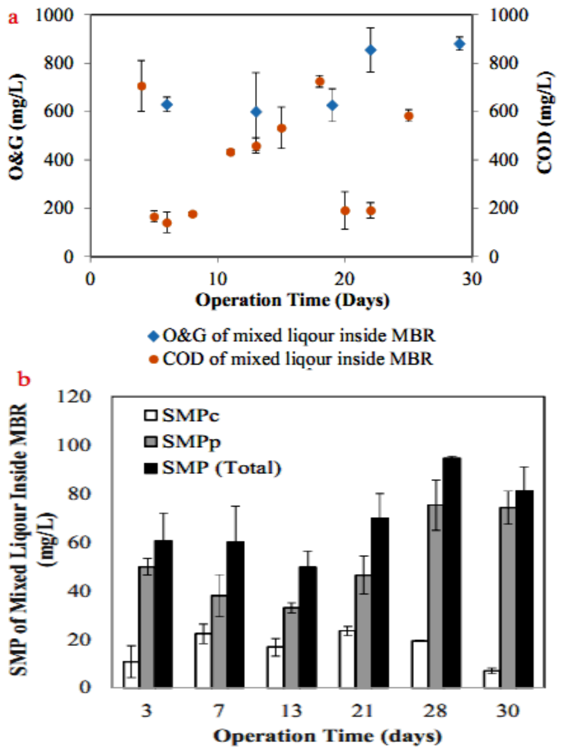

Figure 3. Variations of (a) COD and O&G, and (b) SMP_{total} and its components (SMP_P and SMP_C) concentration in the mixed liquor inside the MBR during operation of MBR. Error bars represent standard deviations of three replicates

observed for O&G and SMP_{total} (mainly SMP_P). The inability of the microbial population inside the MBR to biodegrade the emulsified oil particles and/or the large molecular weight SMPs could potentially lead to the accumulation of O&G and SMP in the mixed liquor [17].

3. 2. Removal Performance of the MBR Treating Real VOW Non-biodegradable fraction of COD is the major problem in the biological treatment of industrial wastewater and limits the maximum COD removal efficiency achieved during a biological treatment process. BOD_5/COD ratio constitutes a good measure of the biodegradability of a wastewater and contaminants. In this study, the BOD_5/COD ratio for VOW was about 0.75. BOD_5/COD ratio of ≥0.4 indicates generally acceptable biodegradability for an industrial wastewater [10, 23].

The variation of COD removal efficiency and concentration in the effluent of MBR is presented in Figure 4. The results of one-way ANOVA analysis (not presented) showed that during MBR operation in terms of COD removal efficiency MBR operation attained steady state condition after day 15. The steady state average COD removal efficiency and effluent COD concentration in the present study were 85.0 ± 1.3 % and 88.7 ± 1.5 mg L^{-1}, respectively.

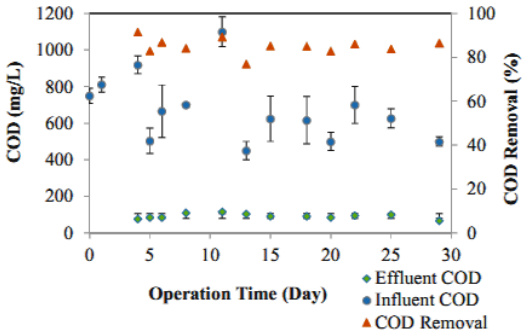

Figure 4. Variations of COD removal efficiency, influent and effluent concentration during MBR operation. Error bars represent standard deviations of three replicates.

As a result of complete maintenance of activated sludge in the bioreactor by immersed membrane, constant concentration of biomass could be sustained despite large variations in the composition of the wastewater.

The concentration of effluent O&G and its removal efficiency during the MBR operation are presented in Figure 5. The average O&G removal efficiency and effluent concentration in the present study was 82.7 ± 1.4 % and 9.3 ± 0.4 mg L^{-1}, respectively.

The results of biological treatment of VOW of Kourosh vegetable oil factory that used combined system of conventional ASP and MBBR showed that at HRT equal to 72 h, the effluent COD concentration was around 190 ± 10 mg L^{-1}. A comparison between the combined system used at the Kourosh vegetable oil factory and MBR system used in present study (effluent COD concentration was around 89 ± 2 mg L^{-1} at HRT equal to 48 h) represents a good performance of MBR in the removal of COD from VOW.

Ma et al. [4] on the treatment of VOWs using submerged MBR with complete sludge retention and different HRTs in the range 16-23 h have reported average COD and O&G removal efficiencies above 86.1 ± 2.1 % and 94.8 ± 1.7 %, respectively. Reddy et al. [14] reported that 81% of COD of sunflower oil

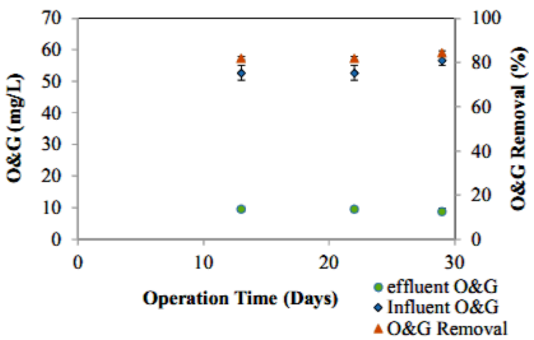

Figure 5. Variations of O&G removal efficiency, influent and effluent concentration during MBR operation. Error bars represent standard deviations of three replicates.

wastewater was removed using an ASP, at SRT 15 days and HRT 24 h. Mkhize et al. [15], used an anaerobic/aerobic SBR for the treatment of edible oil effluent and showed that influent COD and O&G removal efficiencies were 75% and 90%, respectively, at HRT of 24 h and SRT of 30 days.

The recalcitrant organic compounds in the oil that remained in the effluent after biological treatment or microbially produced SMP which can pass through the membrane are the sources of the remaining COD in the effluent of the biological treatment of oily wastewaters [18]. For this reason, the concentration of effluent SMP and its components was measured during the operation of the MBR. The variations of average values of concentration of the SMP_{total} and its components (SMP_P and SMP_C) in the effluent during operation of the MBR is presented in Figure 6. As the O&G of effluent show (Figure 5) unbiodegraded oily compounds are a part of the residual COD in the MBR effluent but production of SMP by the activated sludge inside the MBR is the larger part. Results presented in Figure 6 indicate that throughout the operation of the MBR, effluent SMP is mainly in the form of protein compounds.

3. 3. The Specific Oxygen Utilization Rate (SOUR)

Oxygen utilization rate by microorganisms is expressed by SOUR and indicates the metabolic activity and health of the activated sludge process [3]. In the present study, SOUR of activated sludge has been decreased from 12.8 mg O_2 g $MLSS^{-1}$ h^{-1} to 3.0 mg O_2 g $MLSS^{-1}$ h^{-1} during the MBR operation. This decrease can be attributed to the toxicity and inhibitory effect of the constituents of VOW on the aerobic metabolic activity of microorganisms and therefore the activated sludge growth rate has been decreased during the operation of the MBR. The data of MLSS and MLVSS concentrations during MBR operation (Figure 2) also confirm this result. With decrease in the activated sludge growth rate during the MBR operation, MLSS and MLVSS decreased and led to accumulation of O&G and SMP inside the bioreactor (Figure 3a and 3b).

3. 4. Turbidity Removal Performance

Although the turbidity of the raw VOW was high (54.1±16 NTU), the effluent turbidity was less than 1.5 NTU during the entire MBR operation due to the good separation by the MF membranes (Figure 7). This is indicative of the good performance of the MBR system compared to other biological treatment systems which are not membrane based, especially for the treatment of oily industrial wastewaters [17, 22]. In ASP due to washout of large amount of biomass as a result of sludge-oil aggregation, formation of pin-point floc and great amounts of free-swimming bacteria, significantly high TSS values (with temporary loss in COD removal occurring) have been reported in the treated VOW effluent [14].

3. 5. Membrane Fouling In Figure 8, variations of transmembrane pressure (TMP) during MBR operation as a criteria of membrane fouling is shown. This data showed that in the present study the changes of TMP during operation of the MBR was negligible and remained in the range 0.1–1.0 kPa and the need for membrane washing never arose. The membrane fouling rates in the present study was lower than the value reported by Ma et al. [4] on the treatment of VOWs using submerged MBR with complete sludge retention and different HRTs in the range 16-23 h and indicate the very good potential of the MBR employing a moderate SRT of 20 days, in term of membrane fouling, for the treatment of VOW of Kourosh vegetable oil factory.

3. 6. Wastewater Reuse In this study, the water quality of the MBR effluent could meet discharge standards for direct discharges of treated effluents of VOWs to surface water and well. The treated VOW in the MBR also has a potential for reuse in different ways such as agricultural and irrigation watering and different industrial reuse (Table 3).

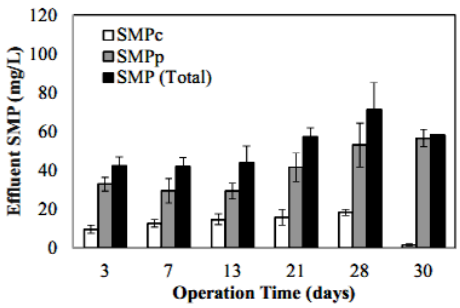

Figure 6. Variations of average values of concentration of the SMP_{total} and its components (SMP_P and SMP_C) in the effluent during operation of the MBR. Error bars represent standard deviations of three replicates

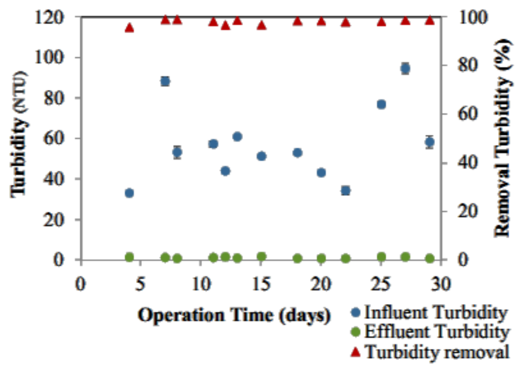

Figure 7. Variations of influent and effluent turbidity and turbidity removal during MBR operation. Error bars represent standard deviations of three replicates

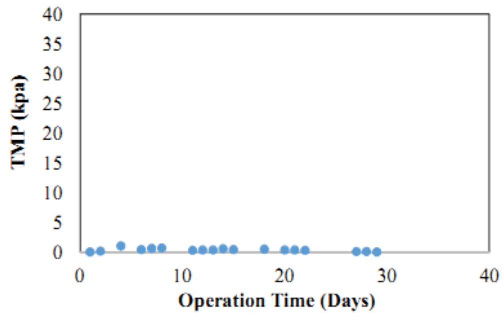

Figure 8. Variations of transmembrane pressure during MBR operation

TABLE 3. Comparison of treated VOW and reuse/discharge limits

Parameter	pH (-)	COD (mg L^{-1})	O&G (mg L^{-1})	TSS (mg L^{-1})	Turbidity (NTU)
MBR treated effluent	6.8± 1	88.7±11.5	9.3 ± 0.4	<2	1.1 ± 0.4
Discharge to surface water[a]	6.5-8.5	100	10	40	50
Discharge to well[a]	5-9	100	10	-	-
Agricultural and irrigation uses[a]	6-8.5	200	10	100	50
Discharge to surface water[b]	6-9	250	10	50	-

[a] Wastewater discharge standards according to the regulations of Iran's Environmental Protection Organization (IEPO) [12]
[b] Effluent guidelines of World Bank Group for direct discharge of treated VOW to surface waters [5]

Effluent guidelines of World Bank Group for direct discharges of treated VOW to surface waters for common application [5] are also given in Table 3. Therefore, using MBR, a large amount of processed water can be reused, which facilitates extra effective water management and concludes to a substantial decline in cost of wastewater discharge.

4. CONCLUSIONS

In the present study, a laboratory scale submerged MBR was evaluated for the treatment of VOW. During the 30 days operation of the MBR at HRT and SRT of 48 h and 20 days, respectively, effective and stable organic compounds removal from the VOW (COD=85.0±1.3 % and O&G=82.7%±1.4 %) using MBR was achieved and

1. Wastewater discharge standards according to the regulations of Iran's Environmental Protection Organization (IEPO) dated 11/26/1994, http://ts.tpww.ir/fa/pfazelab/pf2.

there was a consistently low turbidity in the MBR effluent. Part of the MBR effluent residual COD in the present study was related to unbiodegraded oily compounds but the larger part was related to the production of SMP (mainly SMP_P) by the activated sludge inside the MBR. Decrease in the SOUR of activated sludge during the MBR operation can be attributed to the inhibitory effect of the constituents of VOW on the aerobic metabolic activity and hence growth rate of the activated sludge during the operation of the MBR. With decrease in MLSS and MLVSS concentration during the MBR operation, O&G and SMP accumulated in the bioreactor. The effluent COD and O&G value during the operation of the MBR remained in the range 88.7±11.5 and 9.3 ± 0.4 mg L^{-1}, respectively. Also, the membrane fouling rate during the operation of the MBR was very low and, therefore, the need for membrane washing never arose. The results of the present study showed that the MBR treated effluent is acceptable for direct discharge to surface water and well and for agricultural and irrigation uses and indicate the very good potential of the MBR, both in terms of removal performance and membrane fouling, for treatment of VOW of Kourosh vegetable oil factory compared to the combined system of conventional ASP and MBBR used in this factory.

5. ACKNOWLEDGEMENT

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The Study of Organic Removal Efficiency and Membrane Fouling in a Submerged Membrane Bioreactor Treating Vegetable Oil Wastewater

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چکیده

خصوصیات فاضلاب روغن نباتی (VOW) به طور کلی بوی زننده، رنگ تیره، و محتوای آلی بالا، از جمله مقادیر زیاد روغن و گریس (O&G)، اکسیژن مورد نیاز شیمیایی (COD)، اسیدهای چرب و چربی است. بنابراین، باید برای جلوگیری از آلودگی محیط زیست، VOW بطور مناسبی تصفیه شوند. هدف این مطالعه، بررسی تصفیه بیولوژیکی VOW با استفاده از یک بیوراکتور غشایی (MBR) از لحاظ عملکرد حذف آلاینده های آلی و گرفتگی غشاء بود. در طول ۳۰ روز عملیات MBR در زمان ماند هیدرولیکی و زمان ماند جامد ۴۸ ساعت و ۲۰ روز به ترتیب، کدورت خروجی MBR به طور پیوسته کم (<۲ NTU) بود. بازده حذف COD و O&G از پساب توسط MBR، ۸۵/۰ ±۱/۳ ٪ و ۸۲/۷ ±۱/۴ ٪ به ترتیب بدست آمد. با کاهش فعالیت متابولیک هوازی و در نتیجه سرعت رشد لجن فعال در طول عملیات MBR، غلظت MLSS و MLVSS کاهش یافت و در نتیجه تجمع O&G و محصولات محلول میکروبی (SMP) در داخل بیوراکتور رخ داد. مقدار COD پساب خروجی و فشار عبوری دو طرف غشا در طول عملیات MBR در محدوده ۸۸/۷ ±۱۱/۵ میلی گرم بر لیتر و ۲-۱ کیلو پاسکال به ترتیب باقی ماند. نتایج حاصل از این تحقیق پتانسیل بسیار خوبی از MBR، هم از نظر عملکرد حذف و هم از نظر گرفتگی غشاء را برای تصفیه VOWs نشان می دهد.

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