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Treatability of household waste plastic garbage bag recycling industry wastewater with membrane

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Abstract. In this study, household waste plastic garbage bag recycling industry wastewater was treated by a membrane process to recycle water for using in the bags washing process. Two different ultrafiltration (UP150 and UP005) and nanofiltration (NF270 and NF90) membranes were tested. The steady-state permeate flux was obtained 14.9 and 19.2 L/m^2.h at 5 bar for UP150 and UP005 membranes, respectively. However, the steady-state permeate flux was 12.9 and 8.9 L/m^2.h at 20 bar for NF270 and NF90 membranes, respectively. The chemical oxygen demand (COD) was also tested for all membrane permeates and the highest COD removal efficiency was obtained for NF90 membrane. Thus, optimization was carried out using NF90 membrane and the effect of transmembrane pressure (10, 20, 30 bar) and solution pH (5, 7, 9) on COD removal efficiency was tested. The results showed that the highest steady-state permeate flux (23.5 L/m^2.h) and COD removal efficiency (95.1%) were obtained at 30 bar and pH 9. After the optimization of the membrane type and operating conditions, 75% recovery was obtained to re-use in the bags washing process. The concentrate stream was treated by an activated sludge process to manage membrane concentrate and to provide discharge standards. The maximum COD removal efficiency in biological treatment for membrane concentrate stream was 96.2% under steady-state condition using a sequencing batch reactor (SBR) operated at 10 days of sludge retention time and 12 h of hydraulic retention time. The proposed combined process including membrane and activated sludge processes was successfully used to recover wastewater.

Keywords: household waste plastic garbage bag recycling industry wastewater; water recovery; water reuse; membrane process; activated sludge

1. Introduction

Plastics are known as materials of the modern age due to their lighter, cheaper, and more durable properties than other materials. Therefore, these materials are used in a wide range of applications such as constructions, automotive, electronics, agriculture, and mostly packaging (Zhang *et al.* 2020). According to Plastics Europe, 60 million tons of plastics were produced in Europe and 27 million tons of plastics were collected as waste material. 31% of this plastic waste was recycled, 42% was incinerated, and 27% was disposed to the landfill in 2016 (PlasticsEurope and EPRO 2016). In addition, it was stated that the recycling plastic rate was higher than landfilled for the first time in 2016. Furthermore, recycling is the first preferable waste treatment option prior to recovery and disposal in EU Waste Framework Directive (2008/98/EC). Therefore, it is clear that the recycling of plastics has an important issue for minimizing environmental problems, preserving the national economy, and compliance with the legal framework.

In recent decades, plastic bags (PBs) have great attention by retailers and consumers because of their low cost and practicality (Rivers *et al.* 2017) and they are mostly used for shopping and household garbage collection and thrown away after being used once. Furthermore, they may be toxic and pollute the environment (Bharadwaj *et al.* 2020) because of containing heavy metals and organometallic compounds (Alam *et al.* 2018). In addition, it is known that PBs are non–biodegradable in aerobic, anaerobic, or semi-aerobic environments (Kang and Zhu, 2014), and they have a negative effect on wildlife, water resources as well as landfills (Ahamed *et al.* 2021). Because of these environmental concerns, the use of plastic bags has recently been tried to be reduced with directives in many countries. Although legal pressure, PBs are currently used and 16.7 million tons of packaging material including plastic bags were collected for mostly recycling (40.8%), energy recovery (38.8%), and landfilled (20.4%) (Plastics Europe Association of Plastics Manufacturers, 2018). According to the Ministry of Environment and Urbanization of Turkey, the domestic consumption of plastic bags was 354,000 tons in 2016.

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Recycling processes of plastics are divided into mainly four categories: re-extrusion (primary-produce products with equivalent properties), mechanical (secondary-produce products requiring lower properties), chemical (tertiary), and energy recovery (quaternary) (Singh *et al.* 2017). Primary recycling is the re-process of scrap plastic in the extrusion cycle (Al-Salem, 2009). Mechanical recycling is an operation that reprocessing plastic for manufacturing a secondary raw material (Almeshal *et al.* 2020). The mechanical recycling process of plastics are generally involved four steps including separation/sorting, washing, grinding, and reprocessing (including re-melting, adding additives, and pelletizing) (Ragaert *et al.* 2017). Water is used in the washing step for cleaning plastic bags from contaminants or glue and quenching steps for cooling mainly (Al-Salem, 2009). Therefore, highly polluted wastewater flows from plastic recycling industries originated from washing. It may have high turbidity, suspending and surface-active agents, suspended solids content, emulsions, catalysts, and Chemical Oxygen Demand (COD) especially for plastic bags containing household waste (Ismail and Al-Hashmi, 2011). However, the absence of any known study on the treatment of household plastic garbage bag recycling industry wastewater. Currently, many technologies have been performed for industrial wastewater treatment. Membrane technology is one of the practical technologies for wastewater treatment (Ismail *et al.* 2020) due to its advantages such as stability of effluent (Zheng *et al.* 2015), low operational cost (Pronk *et al.* 2019), less sludge production (Wang *et al.* 2016), and smaller footprint (Macedonio and Drioli, 2018). Membrane fouling is one of the most common problems encountered in the membrane processes that causes a higher energy use, a shorter life-span, and a need to clean more frequently. However, in recent years, there have been many studies performed, especially on membrane modification, to overcome membrane fouling (Sun and Li, 2018; Sun *et al.* 2018). However, membrane technologies generated concentrated waste which needs further treatment (Balcik-Canbolat *et al.* 2019). Furthermore, these concentrates must be managed carefully because of their higher content of salt, organic, and inorganic contaminants (Tomasini *et al.* 2019, Sanmartino *et al.* 2017). Since zero liquid discharge techniques have been developed, many chemical and physical methods such as adsorption, coagulation, oxidation, and precipitation were performed for the treatment of membrane concentrate (Sanmartino *et al.* 2017) (Pramanik *et al.* 2017). However, they are not as effective as biological methods due to high operation costs and secondary pollution (Lan *et al.* 2018).

On the other hand, the activated sludge process (ASP) is the most commonly used biological treatment technology in the wastewater treatment plants both domestic and industrial wastewaters (Badejo *et al*. 2017, Nzila *et al.* 2016). In addition to effective degradation and mineralization of a variety of organic pollutants in wastewater (Kassab *et al.* 2010), it could be a terrain treatment option for membrane concentrates due to low cost and energy consumption (Xu *et al.* 2013). To date, various biological treatment methods have been applied in membrane concentrate. In a study, the treatment of reserve osmosis brine was investigated with a biological activated carbon (BAC) column. It was found that 25% of TOC and 39.6% of COD removal were determined with BAC at 40 min empty bed contact time (Ng *et al.* 2008). Balcik-Canbolat *et al*. (2019) was studied to the combination of Fenton and biological process for treatment of textile membrane concentrate. It was reported that after Fenton process, COD removal efficiency was 75% in a sequencing batch reactor which operated at 10 days of sludge retention time and 12 h of hydraulic retention time (Balcik-Canbolat *et al.* 2019). Lan *et al*. (2018) investigated the treatment of coal chemical reverse osmosis concentrate with a membrane-aerated biofilm reactor system. They found that COD and TN removal efficiencies were 81.01% and 70.72%, respectively (Lan *et al.* 2018).

In this study, household plastic garbage bags recycling industry wastewater was treated using membrane process and water was recovered for reuse in the bags washing process. Moreover, membrane concentrate was also treated by an activated sludge process. Two different ultrafiltration (UP150 and UP005) and nanofiltration (NF270 and NF90) membranes were tested to obtain clean water. For membrane studies, the effect of transmembrane pressure and pH were tested on flux and permeate quality. The concentrate collected from NF90 membrane was treated with activated sludge. According to the published studies in the literature, it is the first study on plastic garbage bags recycling industry wastewater treatment and water recovery using the membrane process.

2. Materials and methods

2.1. Plastic garbage bags recycling process and wastewater characteristics

Garbage bags are commonly made from Low-Density Polyethylene (LDPE) and this type of plastic is recyclable. Plastic bag recycling involves chipping the bags into pellets that can be reprocessed into new bags. The garbage bags must be washed before the chipping process and wastewater comes from the washing of bags. A schematic diagram is shown in Fig. 1.

Household plastic garbage bag recycling industry wastewater was obtained from a firm located in Mersin, south region of Turkey. Wastewater was collected for the months between April and June 2019. The characterization of the wastewater is shown in Table 1. Chemical oxygen demand (COD) was measured according to the standard methods of 5220C (APHA, 2005). A closed reflux titrimetric method was used for COD analysis. Total suspended solids (TSS) was measured according to the standard methods of 2540D (APHA, 2005). The conductivity and pH were measured using a pH/Cond 340i Handheld Multimeters, WTW. Total nitrogen (TN) and total phosphate (TP) were determined with Hach Lange LCK 138 and LCK 349 test kits, respectively. Lowry method was used for the measurement of protein content by using a UV

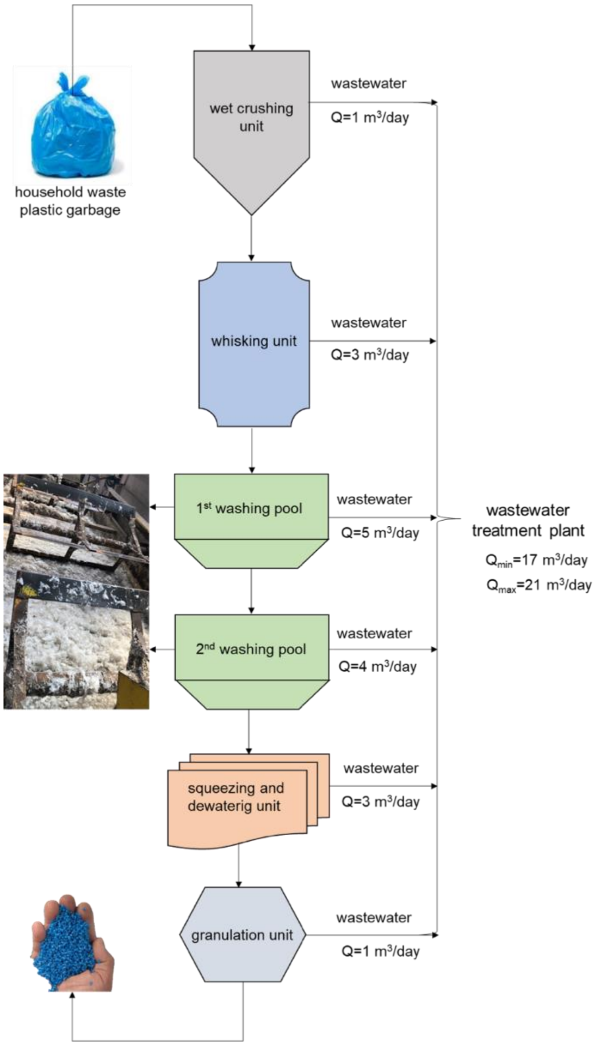

Fig. 1 A schematic diagram of wastewater production during plastic garbage bags recycling process

Table 1 Characterization of the household plastic garbage bag recycling industry wastewater

Parameter	Unit	Value
COD	mg/L	2600 ± 317
TP	mg/L	13.6 ± 1.6
TN	mg/L	31.2 ± 3.5
TSS	mg/L	225 ± 15
Protein	mg/L	83.3 ± 18
Carbohydrate	mg/L	32.3 ± 3
Conductivity	µS/cm	457 ± 12
pH	-	6.9 ± 0.2

vis spectrophotometer (GBC, Cintra-20) at the wavelength of 660 nm (Lowry *et al.* 1951). Bovine serum albumin (BSA) was used as a standard and the results expressed in mg equivalent of BSA per liter. Dubois method was used for the measurement of carbohydrate content at 490 nm (Dubois *et al.* 1956). Glucose was used as a standard and the results expressed in mg equivalent of glucose per liter. Triplicate experiments were performed for all measurements. Standards were prepared in deionized (Millipore Direct-Q3UV) water displaying a resistance of not less than 18 MΩ.

The removal efficiencies (R) were calculated using Eq. (1).

$$\text{Removal efficiency} \quad (\%) = \frac{C_0 - C_t}{C_0} \times 100 \qquad (1)$$

where, C_o is the initial concentration and C_t is the concentration measured at any time t.

The permeate flux was (J) was determined using Eq. (2).

$$Permeate\ flux\ (J_W) = \frac{Q}{Ax\Delta t} \qquad (2)$$

where, Q is the quantity of the permeate sample collected over a period of time (Δt, h) (L); A is the effective membrane area for filtration (m^2).

2.2 Membrane and activated sludge combined studies

Two different UF (UP150 and UP005) and NF (NF270 and NF90) membranes were used for the removal of the organic pollutants from wastewater generated by the household plastic garbage bag recycling industry. Both NF membranes are made of a composite of polyamide thin films. The membranes consist of three layers as follows: a polyester supporting structure, a microporous polysulfone interlayer, and an ultrathin polyamide barrier layer (on the top). The NF270 membrane is a piperazine-based semi-aromatic polyamide thin-film composite, whereas the NF90 membrane is a fully aromatic polyamide-based thin film composite. The properties of the four membranes are shown in Table 2. At the beginning of all experiments, the membranes were immersed in distilled water overnight and they were compressed under a transmembrane pressure (ΔP) of 10 bar for UF and 30 bar for NF membranes for 3 h.

A cross-flow test unit, as depicted in Fig. 2, was used in this study. This system was composed of the following components: a coarse filter, a membrane module, a high pressure pump, a feed tank, the necessary fittings, and an aeration tank for the treatment of membrane concentrate. The stainless steel cross-flow filtration system had an operating volume of 10 L and a filtration area of 0.05 m^2. The system pressure was adjusted via the by-pass valve. Wastewater was pumped across the membrane cell from the feed tank using a centrifugal pump.

Table 2 Characteristics of UF and NF membranes used in experiments

Properties	UP150	UP005	NF270	NF90
Molecular Weight Cutoff (Daltons)	150,000	5,000	~ 200-400	~ 200-400
Material Type	PES	PES	Polyamide Thin-Film	Polyamide Thin-Film
Charge	-	-	Negative	Negative
Temperature (max) (°C)	95	95	45	45
pH range	0-14	0-14	2-11	2-11
Water Flux ($L/m^2.h$)/psi	40.8/58	571.4/29	122.5-166.7/130	78.2-102/130
Supplier	Microdyn Nadir	Microdyn Nadir	Dow Filmtec	Dow Filmtec

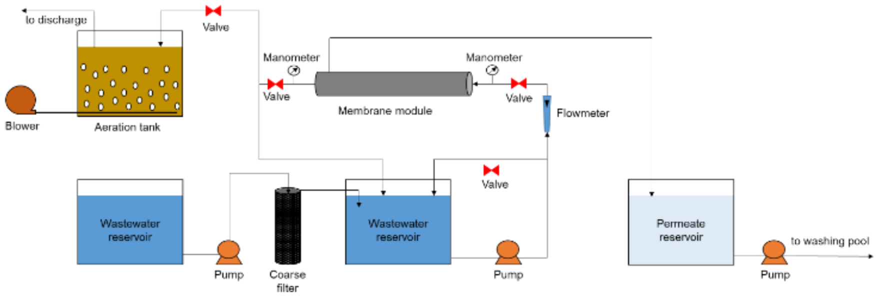

Fig. 2 Schematic diagram of the combined cross-flow/SBR experimental set up

Membrane concentrate was sent to a 5 L-SBR for the biological treatment. The SBR was inoculated with activated sludge obtained from the aeration tank of a municipal wastewater treatment plant. The SBR was operated with 10 days of sludge retention time (SRT) and 12 h hydraulic retention time (HRT) during the experiments. SBR was equipped with blower and tubing pumps and it was kept at room temperature of 25±1 °C. In SBR, two cycles were performed per day, in which the feeding time and settling time was 10 min and 1 h, respectively. The blower capacity was adjusted to maintain a dissolved oxygen concentration of 2-3 mg/L. The schematic diagram of the combined system is demonstrated in Fig. 2.

3. Results and discussion

3.1 Membrane selection

Selecting the appropriate membrane is very important in membrane studies. Therefore, we investigated appropriate membrane type and optimum conditions for this study. The permeate fluxes against time for UF and NF membranes are presented in Figs. 3A and 3B, respectively. Moreover, the characterization of the permeate is shown in Fig. 3C in which the transmembrane pressures were adjusted to 5 bar for UF type membranes and 20 bar for NF type membranes. Although the UP150 membrane had a higher MWCO than UP005 membrane, its permeate flux values were similar to the UP005 and decreased further towards at the end of the experiment. This behavior could be explained as UP150 membrane had a larger pore size than UP005 membrane, the contaminants in the wastewater were clogged the pores more quickly than UP005 membrane, which caused further flux reduction after 30 min. Cisse *et al*. (2011) was studied about the concentration of anthocyanins from roselle extract by UF and NF membranes and they observed that the membrane resistance of UF150 increased more than UF005 membrane with the increasing transmembrane pressure that confirmed the lower permeate flux of UP150 membrane in this study (Cissé *et al.* 2011).

The steady-state permeate flux was obtained 14.9 and 19.2 L/m^2.h at 5 bar for UP150 and UP005 membranes, respectively (Fig. 3A). NF membranes (NF270 and NF90) were also tested in order to compare the permeate quality properties of both UF and NF membranes. The highest steady-state permeate flux was obtained for NF270

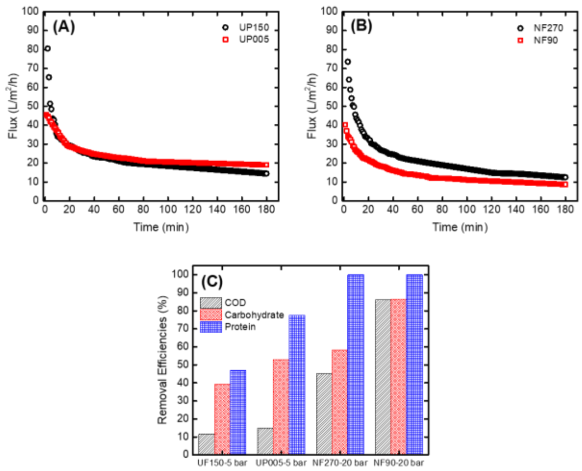

Fig. 3 Effect of membrane type on permeate flux for (A) UP150 and UP005 membranes, (B) NF270 and NF90 membranes, (C) COD, carbohydrate, and protein rejections for UF and NF membranes (ΔP=5 bar for UF membranes; ΔP=20 bar for NF membranes; wastewater pH=6.9)

membrane (12.9 L/m^2.h) due to its looser membrane properties compared to NF90 membrane, which enhanced lower steady-state permeate flux (8.9 L/m^2.h) at 20 bar (Fig. 3B). The increment in permeate water flux due to the looser membrane properties is caused by the larger nanosize pore diameters in the looser activate layer of the membrane that allows more permeate water passage (Kang *et al.* 2019). The permeate fluxes of the NF membranes were similar to another study in which the water permeability was higher for the NF90 membrane compared to NF270 membrane used for the treatment of bisphenol A (BPA) (Yüksel *et al.* 2013).

When the characterization of the permeate water was examined, the highest COD (86.0%) and carbohydrate (86.4%) removal efficiencies were obtained for NF90 membrane due to its tighter membrane properties compared to other membranes. The protein removal efficiency was 100% for NF90 and NF270 membranes; however, UP150 and UP005 membranes were supplied 46.9% and 77.5% protein removal efficiency, respectively. Similar results were obtained in a study by Yüksel *et al.* (2013) in which the BPA removal efficiency of NF270 was lower than NF90 membrane due to its larger pore size (Yüksel *et al.* 2013). Besides, the reason for the unchanged rejection value of

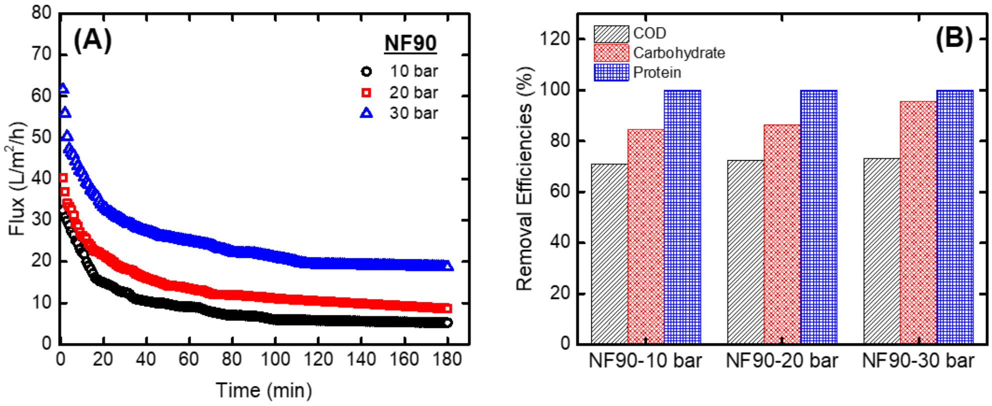

Fig. 4. The effect of transmembrane pressure on permeate flux and rejection (wastewater pH=6.9)

protein was the molecular size of it that could be easily removed by both NF90 and NF270 membranes. As a result, the optimum membrane was determined as NF90 for the next experimental studies due to its highest rejection and acceptable permeate flux values.

3.2 Optimization of the operating conditions

The effect of transmembrane pressure on permeate flux and rejection efficiencies are given in Fig 4. Because the NF membrane is a pressure-driven membrane, the permeate flux was enhanced by increasing transmembrane pressure. The highest permeate flux was obtained as 18.8 L/m^2.h at 30 bar after 180 min while it was 9.0 L/m^2.h and 5.3 L/m^2.h for 20 bar and 10 bar, respectively (Fig. 4A). Besides, the maximum protein (100%) rejection, carbohydrate rejection (95.5%), and COD rejection (73.3%) were obtained at 30 bar due to an increased permeability of solute at high pressure, which caused an increase of rejection (Fig. 4B). The other reason for improved permeate flux and rejection could be explained by the classical solution-diffusion model in which the solute transport is not affected by applied transmembrane pressure while the permeate flux is proportional to net pressure which is the difference between applied pressure and osmotic pressure. The permeate flux was folded double at 30 bar compared to 20 bar which were 18.8 L/m^2.h and 9.0 L/m^2.h, respectively which means a quite high value and makes operation at 30 bar pressure acceptable. Also in our study, the permeate flux decline trend at 30 bar was similar to permeate fluxes at 20 bar and 10 bar. As a result, the optimum operating pressure was determined as 30 bar considering both the permeate flux and carbohydrate rejection.

After the optimization of transmembrane pressure, the feed wastewater pH value was studied for pH 5, 7, and 9. The effect of pH on permeate flux and rejection efficiencies are given in Figs. 5A and 5B, respectively. Moreover, the zeta potential of the NF90 membrane and the wastewater are shown in Fig. 5C. As it can be seen in Fig. 5A, the permeate flux was close to each other in pH 5 and pH 7 while it was improved slightly in pH 9. Additionally, the carbohydrate rejections (≥93.7%) and protein rejections (100%) were similar for all pH values and the COD

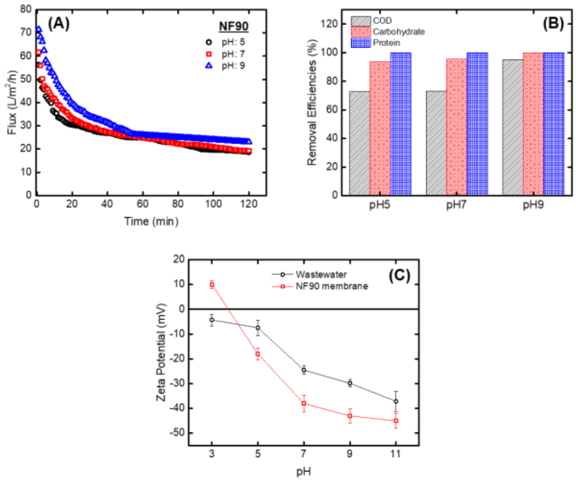

Fig. 5 The effect of pH on (A) permeate flux and (B) rejection efficiencies. (C) The zeta potential of the NF90 membrane and wastewater (ΔP=30 bar)

rejection was increased from 72.7% to 95.1% when the pH value was increased from pH 5 to pH 9 because of the zeta potential of the membrane was more negative at pH 9, which improved the carbohydrate and COD rejections due to electrostatic repulsion between membrane surface and negatively charged colloids in wastewater (Fig. 5C). Besides, similar results were obtained by Nghiem and Hawkes (2007) that the NF90 membrane was not affected too much by the pH of wastewater due to the size exclusion mechanism was predominant than the electrostatic repulsion for the rejection mechanism in NF90 membranes (Nghiem and Hawkes, 2007). However, since there was not much improvement in both permeate flux and solute rejections, it was considered appropriate to work at the original pH (6.9) in order to minimize operating costs.

After the optimization of the membrane type and the operating conditions, the wastewater was treated at a 70-75% recovery rate and obtained concentrated stream was treated with a SBR to carry out the biological treatment. The characterization of the concentrated wastewater is shown in Table 3 in which the COD value was increased from 2928 mg/L to 8320 mg/L while the carbohydrate and

Table 3 The characterization of the concentrated wastewater

Parameter	Unit	Inlet	Permeate	Concentrate
COD	mg/L	2928	144	8320
TN	mg/L	31.2	0.3	88.9
TP	mg/L	13.6	0.1	38.8
Carbohydrate	mg/L	30.9	1.4	137.8
Protein	mg/L	83.3	0	196.7

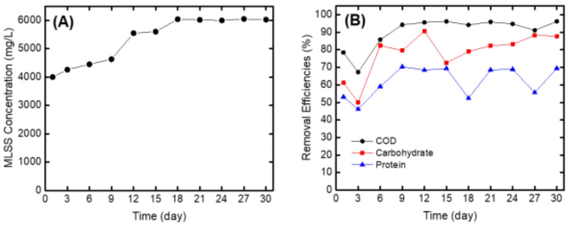

Fig. 6 NF90 membrane concentrate treatment by SBR. (A) MLSS concentration versus time (B) efficiency in terms of COD, carbohydrate, and protein removal (pH: 7).

protein values were increased from 30.9 mg/L to 137.8 mg/L and 83.3 mg/L to 196.7 mg/L, respectively. Moreover, TN and TP values were increased from 31.2 to 88.9 and from 13.6 to 38.8, respectively, which was good for biological treatment.

The SBR was operated at 10 days of SRT and 12 h of HRT for about 1 month. The obtained result was shown in Fig. 6. The biological system reached equilibrium conditions after half of the operation month. By the end of 15 days, the mixed liquor suspended solids (MLSS) and effluent COD concentrations reached a constant value. The average MLSS concentration of the bioreactor was 5956±175 mg/L under steady-state conditions (Fig. 6A). Effluent COD concentrations were observed between 740–316 mg/L for the last 15 days (Fig. 6B). The maximum COD removal efficiency was 96.2% under steady-state conditions. The obtained results demonstrated that NF90 membrane concentrate from household plastic garbage bags recycling industry wastewater could be treated biologically.

The protein rejection was higher than that of the carbohydrate rejection in the NF process but lower in the SBR process. It can be explained as most of the proteins have positive or negative charges depends on solution pH and isoelectric point. Moreover, NF membrane has also charges mostly negatively between pH 5-9. However, carbohydrate has a neutral charge. It could be explained why protein was rejected better than carbohydrate. In the SBR process, the opposite result was obtained. The reason could be explained as soluble microbial products as protein and carbohydrate fractions were different in the SBR process. Carbohydrate fraction might be used higher than protein by microorganisms and it caused better removal efficiency for carbohydrates.

It is known that SBRs can be used effectively as a biological treatment process for various industrial wastewaters due to the high removal efficiency of COD, TN, and TP (Suresh *et al.* 2011). For example, SBR was integrated with the membrane for the treatment of oil and gas field wastewater. COD and oil/grease removal efficiencies were 90.9% and 91.5% for an HRT of 20 h, respectively (Fakhru'l-Razi *et al.* 2010). In another study, the SBR process followed by chemical oxidation was investigated for the treatment of textile industry wastewater. It was found that COD, total Kjeldahl nitrogen (TKN), and TP removal was 91.1%, 91.6%, and 80.6%, respectively (Fongsatitkul *et al.* 2004). It was also reported that COD removal efficiency was 75% in Fenton process followed by SBR operated at 10 days of SRT and 12 h of HRT for textile membrane concentrate (Balcik-Canbolat *et al.* 2019).

4. Conclusion

The obtained results have stated that pressure-driven membrane process coupled with biological treatment are suitable treatment options to recover/reuse water and treatment the NF90 membrane concentrate, according to Turkish Regulations.

The permeate quality of UF membranes was detrimental to the reuse of the water. However, NF90 membrane enhanced the desired permeate water quality. Maximum COD removal efficiency was found to be 95.1% at 30 bar transmembrane pressure and pH 9 for 180 min filtration when NF90 membrane was used. The change in wastewater pH did not affect the carbohydrate and protein rejections. Therefore, the original wastewater pH value was determined as optimum operating pH due to less operating cost and acceptable rejection values. The maximum COD removal efficiency in biological treatment for NF90 concentrated stream was 96.2% under steady-state condition using a SBR operated at 10 days of SRT and 12 h of HRT.

Consequently, the proposed combined process including membrane and biological treatment processes for the treatment of membrane concentrate should be considered as an environmentally and economical friendly process to recover/reuse wastewater from household waste plastic garbage bag recycling industry.

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