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Cake layer reformation rates on self forming dynamic membranes and performance comparison with microfiltration membranes

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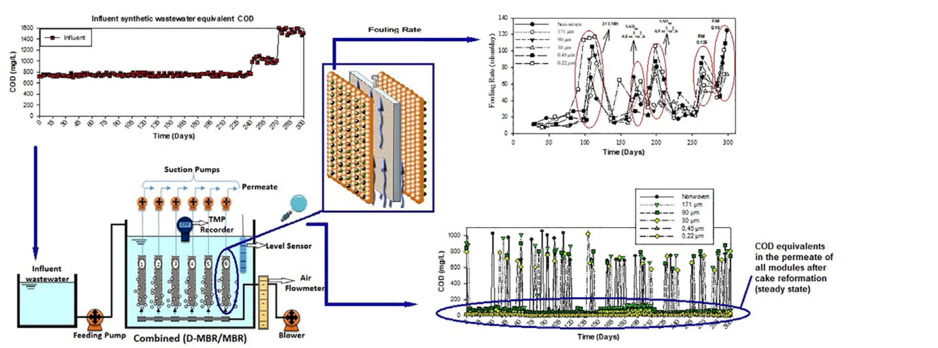
HIGHLIGHTS

- The study focused on cake re-formation rate of Dynamic Membrane Bioreactor (DMBR).
- Six different membranes were operated simultaneously in a single reactor.
- DMBR (30 μm) results obtained in the study were quite similar to MFs results.
- Increasing F/M and decreasing the SAD_m value caused an increase in fouling rates.
- As the cake formation rate increases, the fouling rate also increases.

GRAPHICAL ABSTRACT



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ABSTRACT

Dynamic membranes (DMs) keep on attracting attention progressively as an alternative to conventional membranes because they can be operated with relatively higher fluxes and lower fouling rates. However, there are many factors affecting the performance of DMs, such as DM pore size, structure, and operating conditions. In this study, mainly focused on the investigation of cake formation rates both in initial formation and reformation rates after physical/chemical cleaning. In this context, it has been evaluated the performances of DMs with different pore sizes (171 μm, 90 μm, and 30 μm) and different structures under the same conditions and compared their performances with microfiltration (MF) membranes (0.45 μm and 0.22 μm) in a single reactor. In the study, the effects of different fluxes (15-, 20-, 25 L/m^2·h (LMH), SAD_m (1-, 0.8-, 0.5 m^3-air /m^2·h) and F/M (0.095, 0.125, 0.19 g-COD/g-MLSS·day) conditions on the treatment and filtration performance of DMs were investigated. High COD (>95%) and turbidity (<10 NTU) removals were obtained in this study. In particular, the 30 μm DM (0.65 ± 0.47 NTU) produced quite close effluent turbidity compared to MFs (0.12 ± 0.05 NTU). Low SAD_m and high F/M values resulted in increased effluent COD concentrations and turbidity values. By decreasing the SAD_m, the cake formation rate and the fouling rate increased, which showed that there is a definite relationship between the cake formation rates and the fouling rates. Additionally, considering all the results, the most stable operation was obtained in the 30 μm DM, although it has been occurred the least fouling in the 90 μm membrane in the study. This study, focused on cake reformation rates, attempts to show that DMs can be used as an alternative to MBRs. Especially, when taking into account the results of the reformation rate of 30 μm DM (6.09 NTU/h) and other high filterability features.

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1. Introduction

In recent years, membrane bioreactors (MBRs) have been used in the laboratory and real-scale applications in the treatment of domestic and industrial wastewater due to their specific advantages such as high effluent quality, low sludge production, low footprint, high shock load resistance, and good disinfection capability (Khan et al., 2011; Melin et al., 2006). However, the fouling problem limits the widespread use of these systems as it causes low flux and consequently high cost (Meng et al., 2020; Cai et al., 2018). There are many studies in the literature to mitigate the membrane fouling problem (Zhang and Jiang, 2018; Zhang et al., 2021). Also, many studies on good disinfection capability and advanced treatment using carbon nanotubes (CNTs) and laccases make the use of membranes attractive (Chen et al., 2018). Although cake formation, which is the biggest cause of fouling, is reported as the key factor limiting the flux, this cake layer actually acts as a secondary membrane (Dynamic Membrane: DM) (Ersahin et al., 2013). In this context, dynamic membrane bioreactors (DMBRs) have been studied in recent years based on the concept of performing filtration by forming a cake layer on a support material such as mesh, filter cloth, non-woven (NW) fabric, or textile product, which is significantly cheaper than conventional MBR (Yurtsever et al., 2021). Thus, the aim is to tackle the disadvantages of MBRs.

An effective cake formation on the support material is an important parameter, and it is affected by many factors such as sludge properties, operating conditions, bioreactor design, as well as support layer properties in the DMBR (Isik et al., 2020). Other important considerations when choosing the support materials are resistance to high/low pH and temperature, as well as durability to pressure caused by suction applied during operation. Among these, pore size and structural properties of the support material are important factors since the selection of a suitable support material with appropriate pore size plays a major role for the stable operation of DMBRs (Yurtsever et al., 2020). In a study of Kiso et al. (2000), three different mesh support materials with large pore sizes (100 μm, 200 μm and 500 μm) were tested. In the study, although bacteria in the reactor can be highly retained even by larger meshes, it has been reported that the support material with a pore size of 100 μm provides better effluent quality and retains sludge flocs better. In another study, 20 μm, 40 μm and 60 μm monofilament woven filter cloths made of polypropylene were used in high strength food wastewater treatment, and it has been reported that the most suitable support layer for cake layer development was a 20 μm pore size monofilament filter cloth. Additionally, in the same study it has been reported that the cake layer formed in the DM with a smaller pore size provides higher purification efficiency compared to larger pore size (Mahat et al., 2020). In a study by Cai et al. (2018), smaller pore size (about 1, 5, 10, 25 and 50 μm) support materials were used. For the 10, 5 and 1 μm filters, the initial effluent turbidity was measured as very low (less than 10 Nephelometric Turbidity Unit (NTU)), but when the pore size decreased from 10 to 5 μm, filtration resistance has been reported to increase significantly. In another study, a nylon mesh with 20, 53 and 100 μm pore sizes was used for optimized filtration and removal performances (Yurtsever et al., 2020). In the study, although very low effluent suspended solids (SS) concentrations were obtained in a very short time in the 20 μm support layer, it has been reported that a proper cake layer formation took longer in the 53 and 100 μm support layers. In addition to that, the stable flux for the 20 μm and 53 μm support layers was determined as 8 $L/m^2 \cdot h$ (Liter per square Meter per Hour ((LMH)) and 15 LMH, respectively. In a study of Zhi-Guo et al. (2005), the filtration and treatment performances of a non-woven support layer (3 μm and 5 μm) and conventional hollow fiber MBR were compared. It has been reported that the treatment performances for both membranes show very little difference under the same operating conditions. Based on differences in the filtration resistance, it was deduced that the fouling in the non-woven filter was mostly caused by internal contamination. Considering the effluent turbidity values, the 3 μm non-woven filter performed better than the 5 μm filter. Consequently, the studies indicate that properties such as pore size and support layer characteristics have important effects on treatment and filtration performance.

Accordingly, in this study a non-woven support layer, support layers with different pore sizes and conventional Microfiltration (MF) membranes were all operated under the same conditions, and the differences in both filtration and treatment performances between these membranes were evaluated. Unlike other studies, this study mainly focused on the filtering properties and re-formation properties of the cake layer of non-woven and other support materials. Cake formation properties after chemical or physical cleaning of the membranes were investigated under different fluxes and Specific Air Demand per unit membrane area (SAD_m) conditions. Thus, the optimum operating conditions for effective cake layer development were determined for each support layer.

2. Material and methods

2.1. Bioreactor setup

This study was performed on a lab-scale aerobic bioreactor featuring six membrane modules (Fig. 1). The bioreactor was made of plexiglass and featured an active volume of 7 L. It was completely mixed by means of aeration (Dissolved Oxygen (DO) > 2 $mg \cdot O_2/L$). In DMBR studies, a non-woven (pore size non-uniform) and support materials with three different pore sizes (171 μm, 90 μm and 30 μm) were used for cake formation comparison. The MF-MBR membranes installed had 0.45 μm and 0.22 μm pore sizes, respectively, and were used for performance comparison with the DMBRs. Each membrane had an active surface area of 112.5 cm^2 bipartitely.

The feed was provided with a peristaltic pump (Baoding City, China). The water level was constantly maintained with the use of a Siemens 5SQ21 C6 (Munich, Germany) liquid level control device, and the permeate was drawn using a Filtec peristaltic pump (FPP-1/PH-II 3 / Australia) for each module separately. The system was operated with a constant flux for 4.5 min suction followed by 0.5 min relaxation which was controlled with a Siemens 5SQ21 C20 (Munich, Germany) circuit breaker. In all pump lines, pressure gauges were used to monitor the membrane fouling.

2.2. Materials structures and properties of DMs and MBRs

Preliminary tests were performed to determine the optimum support layer materials structures and properties of DMs and MBRs. In this context, performance tests of NW materials with different viscosities and durability and the other DMs materials with nylon mesh support materials with different pore size were carried out (data not shown). Based on the obtained results, the support materials used in the study were decided by considering some criteria like price, durability at low/high pH and temperatures, resistance to applied suction force, permeate NTU. NW was specially manufactured to durability relatively low/high pH and temperatures with 60% strength and 40% viscosity made up Polipropilen (PP). DMRs support materials, 30 μm (about 160 node/cm^2), 90 μm (about 90 node/cm^2), 171 μm (about 24 node/cm^2) made up Polyethylene (PE) Nylon Mesh, were selected from cheap materials used in silk screen printing also used in the printing press and textile industry. The preliminary studies were carried out before operating in the same way. MF MBRs, formed Polyvinylidene Fluoride (PVDF), is the commonly used Flat Sheet Type MF Membrane (0.45 μm and 0.22 μm) (Kubota Mark, Japan).

2.3. Operating conditions

The bioreactor was inoculated with sludge from return activated sludge (RAS) of Ambarli Advanced Biological Wastewater Treatment Plant in Istanbul, with an initial reactor biomass concentration of 8000 ± 200 mg/L. It was determined that this corresponds to a biomass concentration as volatile suspended solids (VSS) of 7500 ± 200 mg/L. The system was operated on a continuous basis, whereby the feed (reactor influent) was synthetic municipal wastewater having initially a chemical oxygen demand (COD) equivalent value of 750 ± 50 mg COD/L. The synthetic domestic

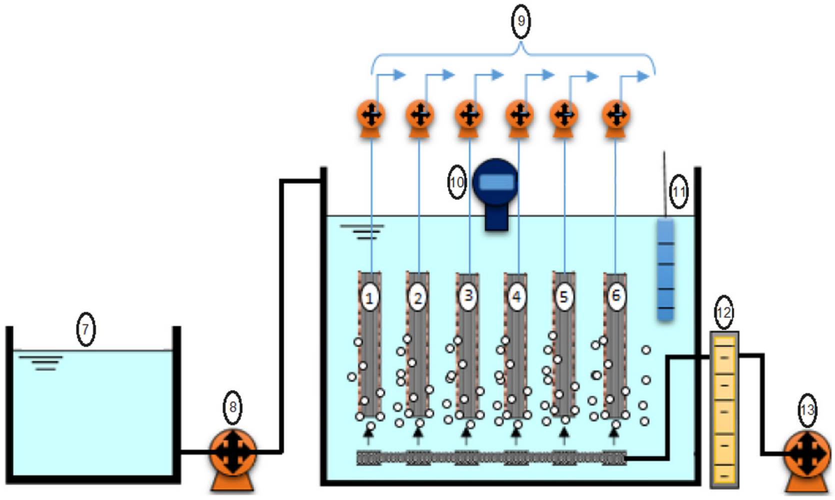

Fig. 1. Combined (DMBR/MBR) aerobic reactor: (1) Non-woven, (2) 171 μm DMBR, (3) (2) 90 μm DMBR, (4) 30 μm DMBR, (5) 0.45 μm MF, (6) 0.22 μm MF, (7) Influent wastewater, (8) Feeding Pump, (9) Suction Pumps, (10) TMP Recorder, (11) Level Sensor, (12) Air Flowmeter, (13) Blower.

wastewater was used to supply a source of carbon, nitrogen, phosphorus and trace metals required for microorganism growth. Based on the ISO 8192-E (2007) method, the content of synthetic domestic wastewater was modified by adding micro and macro elements, including the information in the study conducted by Cinar et al. (2008) as tabulated in Table 1. In the study, tap water was used for feed preparation.

The reactor was operated at three different fluxes of 16.66, 22.22 and 27.77 LMH. Since the reactor operated on the principle of suction for 4.5 min and relaxation for 0.5 min, these values are equal to 15, 20 and 25 net LMH, respectively. Subsequently, the reactor was operated at three different SAD_m values (1, 0.8 and 0.5 m^3-air/m^2·h) in order to evaluate the effect of different operating conditions on cake formation rates. In addition, to demonstrate whether DMs can also be applied in wastewater with higher COD content other than domestic wastewater, three different Food/Microorganism (F/M) ratios (0.095-, 0.125- and 0.19 g-COD/g-Mixed Liquor Suspended Solids (MLSS)·day) were applied. Thus, each DMBR and MBR was subjected to the cake formation process for 300 days in 9 different operating conditions in total. In order to provide steady-state conditions for microorganisms, the time between all periods is a minimum of three solids retention time (SRT). This period was not taken into account in the aforementioned 300 days. The operating conditions of the study were shown in Table 2. At first, each membrane was operated with a flux of net 15 LMH, and the cake formation and pressure relations were examined in this process. Then, the flux was increased to 20 LMH (net) and 25 LMH (net) on days 61 and 91, respectively. SAD_m was kept at 1 m^3-air/m^2·h at these stages. In these periods, three different fluxes (15, 20 and 25 LMH) were analyzed and the optimum flux was determined for each membrane.

Subsequently, optimum SAD_m values were determined by gradually decreasing them (1, 0.8 and finally 0.5 m^3-air/m^2·h), taking into account aeration, which creates one of the highest costs in the operation of wastewater treatment plants. In the different periods, the SAD_m values were reduced to 0.8 and 0.5 m^3-air/m^2·h on the 180 and 210 days, respectively. Finally, the effect on the filtration and treatment performance was investigated by increasing the F/M ratio at the flux at which optimum performance was achieved for each membrane and at the SAD_m value of 1 m^3-air/m^2·h.

In the study, constant flux, variable TMP principle was applied. In the first three periods, the same fluxes were used for all membranes and the fluxes were increased sequentially according to the study (15 LMH between 1 and 60 days, 20 LMH between 61 and 90 days, 25 LMH between 91 and 120 days). After optimum fluxes were determined for each membrane in these periods and it was carried out at optimum fluxes for each membrane in the continuation of the study. The optimum fluxes obtained in the flux optimization studies (periods 1–3) were 20 LMH, 15 LMH, 20 LMH and 15 LMH for 30 μm, 90 μm, 171 μm, and for the NW membrane, respectively. In the continuation of the study (4–9 periods), the operation was continued with these fluxes determined as optimum, and these fluxes were used in the reformation rates calculations.

Throughout the study, sludge was not drawn from the reactor except for the analyses. An average of 350 ml of sludge was taken daily for analyses, which corresponds to 20 days of SRT for the 7-liter reactor. The hydraulic retention time (HRT) was adjusted with the effluent of the 0.22 μm MF membrane, and the effluent of the other membranes (DMBRs and MF-MBR, except the 0.22 μm MF-MBR) were internally returned to the reactor, thus, the escape of biomass was also prevented. When the reactor was operated at different fluxes (15, 20 and 25 net LMH), the HRT values were

Table 1
Modified synthetic domestic wastewater.

Chemical	Concentration (mg/L)
$C_6H_{12}O_6.H_2O$	750 (mg-COD)/L equivalent
Peptone	(200 mg/L of glucose remaining from others)
Meat extract	
NaHCO3	275.4
NH_4CI	230
K_2HPO_4	37
KH_2PO_4	67
$MnSO_4.H_2O$	0.042
$ZnSO_4.7H_2O$	0.010
Na_2SO_3	0.028
$CuSO_4.5H_2O$	0.005
$FeSO_4.7H_2O$	0.592
$NiSO_4.6H_2O$	0.01
$CoCl_2.6H_2O$	0.1

Table 2
Operating conditions of the bioreactor.

Periods	Time (day)	Flux (L/m^2·h (LMH))	SAD_m (m^3-air/m^2·h)	F/M (g-COD/g-MLSS·day)
1	1–60	15	1	
2	61–90	20		0.095
3	91–120	25		
4	121–150	20 LMH for 30 and 90 μm,	1	0.095
5	151–180	15 LMH for 171 μm and	0.8	
6	181–210	NW	0.5	
7	211–240	20 LMH for 30 and 90 μm,	Optimum	0.095
8	241–270	15 LMH for 171 μm and	SAD_m	0.125
9	271–300	NW		0.19

calculated as 41.5 h, 31.1 h and 24.9 h, respectively. The study was carried out at room temperature of 23 ± 2 °C. Theoretically, 11.25 L/h of air (for SAD_m: 1 m^3-air/m^2·h) and the DO level was tried to be kept above 2 mg/L for aerobic conditions. Throughout the study, the SS concentration was kept stable at 8000 ± 200 mg/L. The bioreactor was sampled three times a week for turbidity, COD, pH, and suspended solid (SS) and volatile suspended solid (VSS) analyses. In addition, soluble microbial product (SMP) and extracellular polymeric substances (EPS) were measured at regular intervals.

In addition to in-situ physical cleaning such as intermittent operation (4.5/0.5 min - on/off) and stripping the cake layer with the aeration, ex-situ physical and chemical cleaning have also been applied to recover the flux. When the pressure in the membranes increased above 300 mbar, the system was stopped and the membranes were removed out. First of all, the removal of the cake layer on the membrane surface was done physically using a sponge under tap water. Then, it was kept in sulfuric acid solutions with pH: 3 and hypochlorite solutions with pH: 9.5 for 1 h, respectively. After chemical cleaning, rinsing was done and the membranes were immersed in the reactor. In order to demonstrate the efficiency of both chemical cleaning and physical cleaning, only physical cleaning was applied at different times in the study. Depending on the fouling frequency of the membranes and the pressure increase, both physical and chemical cleaning were applied.

2.4. Analyses

Samples were collected from the reactor feed and effluent and were analyzed directly. Unfiltered COD for effluent and filtered COD for influent measurements were performed as specified in the standard method (Standard Methods, 2011; SM 5220B). Transmembrane pressure (TMP) was measured daily using a programmable digital manometer (Keller-Leo record, 119 8404 Winterthur, Switzerland) and Logger5 software. Turbidity measurements were carried out by WTW TURB 550 IR (Wissenschaftlich-Technische Werkstatten, Weilheim, Germany) turbidity meter. DO measurements were carried out with the use of Hach HQ40D portable multimeter (Loveland, USA). Biomass concentration in the reactor was determined according to Standard Methods (APHA, 2005).

There are various methods for EPS and SMP extraction from different microbial cultures and different types of wastewater sludge (aerobic/anaerobic; activated/granular). In this study, the mixed liquor was centrifuged for 10 min at 4000 rpm and the supernatant filtered through a 0.45 μm membrane. The filtrate was used for the measurement of SMP content as protein and carbohydrate according to the methods of Bradford (1976) and Dubois et al. (1956), respectively. In order to perform EPS extraction, the remaining pellet from centrifugation was washed two times with distilled water and suspended in saline water (0.5% NaCl). The mixed liquor was then subjected to heat treatment (80 °C for 1 h) and afterward centrifuged once more. The supernatant was filtered through a filter with 0.45 μm pore size and EPS concentration of the filtrate was measured in terms of protein and carbohydrate content (Yurtsever et al., 2015).

3. Results and discussions

3.1. Treatment performances and comparison of DMBRs and MBRs

Unfiltered COD and turbidity results from MF and dynamic membrane permeate throughout the study are shown in Fig. 2. In this study, the concentration of influent COD was 750 mg/L until the 240 days and increased to 1000 mg/L and 1500 mg/L on the 240 and 270 days, respectively. Throughout the study, similar effluent concentrations were obtained from all membranes in steady-state conditions (the removal efficiency >90%, except 4–6 Periods). Between days of 150 and 210, when SAD_m values were evaluated, the removal efficiencies of dynamic membranes decreased slightly. In this study, COD analysis was performed unfiltered the samples taken from the permeate. It was observed that the permeate COD values were directly related to the permeate SS values in all periods of the study. The reason for this is that inorganic colloidal substances and microorganisms are also sources of COD in the permeate. In this context, when the operating conditions were kept constant, the effect of the support materials used on COD removal performance may be evaluated. While the COD removal efficiencies of MF membranes were around 99%, removal efficiencies of 30 μm, 90 μm, 171 μm and non-woven dynamic membranes were around 92%, 90%, 83% and 88%, respectively, under constant conditions (Table 3). The aeration rates especially affect the flocculation behavior significantly. Smaller flocs are formed at higher aeration rates due to the shear force effect created by the aeration rate (Etemadi and Yegani, 2019). It has been reported that greater aeration rates cause floc breakage and the formation of smaller particles (De Temmerman et al., 2015). It was considered that the structure of the dynamic layer was negatively disturbed, due to the change in particle size associated with the decrease in the SAD_m value. Although there was a decrease in COD removals due to size disturbance, high removal efficiencies were still obtained. In this study, the closest COD removal to the high efficiency MF membranes was obtained in the 30 μm dynamic membrane. In particular, in the last two periods when the F/M ratio was increased, the removed COD amount also increased by increasing the influent COD concentration. At this stage, COD removal efficiencies were obtained above 98% in the 30 μm dynamic membrane. This result was almost the same as the removal efficiencies of MF membranes, meaning that it can be concluded that the 30 μm dynamic membrane can be an alternative to conventional MF membranes. In addition, the lowest efficiency was obtained in the 171 μm dynamic membrane, and better COD removal efficiencies were obtained in the non-woven membrane compared to the 171 μm membrane in all periods except the 3rd period (Table 3). This result means that the filtration of dissolved organics by the non-woven dynamic membrane is better than that of the 171 μm dynamic membrane. A similar result was achieved in the study conducted by Seo et al. (2002). It is expected that the dynamic cake layer will be more compact and dense in non-woven membranes in long-term operation and it is possible to obtain better quality filtrate (Seo et al., 2002). Despite the fact that the best results were obtained in 30 μm and 90 μm dynamic membranes, the results obtained in the non-woven dynamic membrane were better compared to 171 μm dynamic membrane. Although all dynamic membranes were contained within one reactor, different COD removal efficiencies were observed in the permeate. The reason for this difference is that the cake layer formed on dynamic membranes is different and accordingly has different filtering properties. As described in the next sections, a faster and more compact cake formed on the 30 μm dynamic membrane as compared to the other membranes. The results revealed that the pore size significantly affects the treatment performance and better cake layer was formed in smaller pore size dynamic membranes. Previous studies in the literature also proven this (Cai et al., 2018; Chang et al., 2007). Additionally, it has been reported that more efficient dynamic cake layer formation was achieved with a smaller sized support layer by Mahat et al. (2020). In the study, while it was obtained 95% COD removal with 20 μm DM, they were obtained 70% and 83% removal with 40 and 60 μm DM, respectively. This explains why similar results were obtained with the 30 μm dynamic membrane and microfiltration membranes.

MBRs are operated at lower F/M ratios than conventional activated sludge systems in order to both reduce membrane fouling and provide high oxygen transfer efficiency. Generally, the F/M ratio in MBRs is chosen as one-third of the value of conventional activated sludge systems (0.08–0.24 g-COD/g-MLSS·day) (Yoon, 2015). In this study, the initial F/M ratio was 0.095 g-COD/g-MLSS·day, and then increased gradually to 0.125 and 0.19 g-COD/g-MLSS·day on the 240 and 270 days, respectively. Despite increasing the influent COD concentration, there was no significant change in the effluent COD concentrations.

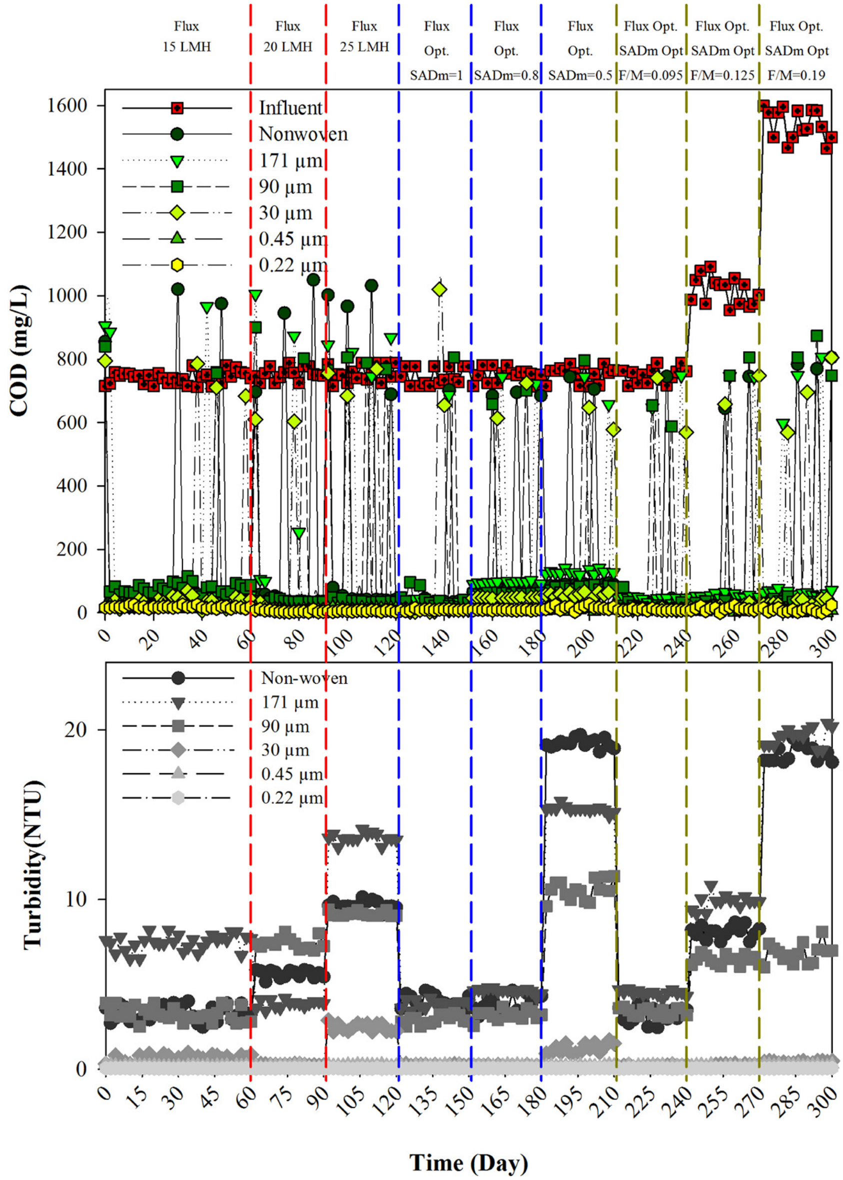

Fig. 2. Obtained COD concentrations (top) and turbidity results (bottom).

Thus, the COD removal efficiency increased due to the increase in the amount of COD removed. At this stage, removal efficiencies of over 95% were obtained in 90 μm, 171 μm and non-woven dynamic membranes. Better removal efficiencies were observed in MFs and 30 μm dynamic membranes. COD removal achieved with around 99% in these membranes. Khoshfetrat et al. (2011) found that increasing the organic loading rate from 1 to 2.5 kg-COD/m^3·day reduced the COD removal efficiency from 90% to 74%. However, in this study, although the loading rate was increased, no change was observed in the effluent COD concentration and accordingly removal efficiencies were obtained at high levels. These results are thought to be related to the good adaptation of microorganisms.

Table 3
COD removal efficiencies obtained from all membranes in different periods.

COD removal efficiencies (%)						
P.	Nonwoven	171 μm	90 μm	30 μm	0.45 μm	0.22 μm
1	94.20 ± 2.65	93.36 ± 2.35	89.66 ± 1.99	95.63 ± 2	97.78 ± 0.56	97.52 ± 0.52
2	94.45 ± 1.27	94.58 ± 3.85	95.02 ± 0.60	98.57 ± 1.07	98.63 ± 0.63	99.30 ± 0.42
3	93.91 ± 1.67	95.51 ± 0.18	95.06 ± 0.74	98.66 ± 0.20	98.11 ± 0.26	99.25 ± 0.21
4	95.22 ± 0.42	95.12 ± 0.46	94.75 ± 3.4	98.92 ± 0.46	98.69 ± 0.19	99.73 ± 0.28
5	90.39 ± 0.54	87.26 ± 0.52	91.81 ± 0.39	93.68 ± 0.29	98.83 ± 0.20	98.80 ± 0.17
6	88.37 ± 0.48	82.83 ± 1.13	89.87 ± 1.26	92.30 ± 0.84	98.82 ± 0.48	97.81 ± 0.78
7	95.31 ± 0.75	94.05 ± 0.79	96.68 ± 2.29	98.38 ± 0.31	98.83 ± 0.21	98.76 ± 0.30
8	96.82 ± 0.28	94.80 ± 0.59	97.61 ± 0.69	98.29 ± 0.63	99.22 ± 0.37	98.75 ± 0.54
9	97.22 ± 0.19	95.82 ± 0.42	97.64 ± 0.50	98.14 ± 0.80	99.90 ± 0.54	98.40 ± 0.41

Although the effluent COD concentrations of all dynamic membranes were similar to those of the MF membranes under steady-state conditions, high effluent COD concentrations (low efficiency) were observed for a short time on the days of physical and chemical cleaning, due to biomass escape until the dynamic layer was re-formed. However, as discussed in more detail in the next section, the effluent COD concentrations decrease to the MF level in a short time after the formation of the cake layer. In this study, 4 different dynamic membranes and 2 different MF membranes were used in a single reactor to ensure exactly the same conditions. Depending on the different operating conditions and the membranes of different structures, differences in turbidity values were also presented in Fig. 2. In addition, filtration performances vary depending on many parameters and factors such as type of support layer, flux, SAD_m, TMP, etc. in the dynamic membranes (Mohan and Nagalakshmi, 2020; Siddiqui et al., 2021). However, it can be stated that very high turbidity removals were observed in all dynamic membranes in this study. The effluent turbidity values were obtained below 10 NTU in all dynamic membranes through the study except for a few periods. The highest turbidity removal was obtained in the 30 μm dynamic membrane as expected among the DMBRs. Throughout the study, the average effluent turbidity value of the 30 μm dynamic membrane was observed as 0.65 ± 0.47 NTU at steady-state conditions, which was almost the same as for the 0.22 μm MF membrane (0.12 ± 0.05 NTU). Despite the fact that the 171 μm dynamic membrane had a low filtering performance (7.46 ± 0.46 NTU) at 15 LMH flux, it performed comparatively better performance (3.85 ± 0.21 NTU) at 20 LMHs. However, when the flux was increased to 25 LMH, the effluent turbidity increased significantly (13.63 ± 0.28 NTU). Similar to what was observed with COD removal, the non-woven dynamic membrane had a better performance compared to the 171 μm dynamic membrane in turbidity removal, and lower performance than other dynamic membranes. Although flux changes did not significantly affect the performances of 30 and 90 μm dynamic membranes, they did affect the performance of non-woven and 171 μm dynamic membranes. SAD_m evaluation was started on the 120 days of this study, and turbidity values were investigated by gradually decreasing the SAD_m values. Although similar turbidity results were obtained in the periods when the SAD_m values were 1 and 0.8 m^3-air/m^2·h, an increase in the effluent turbidity values of all dynamic membranes were observed when the SAD_m value reduced to 0.5 m^3-air/m^2·h. In particular, the effluent turbidity values of the non-woven membrane increased to 20 NTU levels. On the other hand, the lowest change was observed in the 30 μm dynamic membrane, and the effluent turbidity value of this membrane increased to 1.22 ± 0.27 NTU. In previous studies, it was emphasized that the SAD_m (aeration rate) value also significantly affected the particle size. Namely, at higher SAD_m values, the particle size of the biomass in the reactor decreases, and therefore a denser and more compact cake layer is formed on the support layer (Ding et al., 2016; Etemadi and Yegani, 2019). Thus, it is possible to retain small particle size structures and colloidal molecules on the membrane surface. For this reason, it is expected that the effluent turbidity values decrease with increasing SAD_m values. In this study, results were obtained in accordance with this mechanism, and better effluent turbidity values were obtained at higher SAD_m values.

In the final stage of this study, the F/M ratio was increased and the effect of organic loading on the performance was studied. With the increase of F/M rates, it was observed that the effluent turbidity values increased in all membranes except the 30 μm dynamic membrane. Especially, non-woven and 171 μm dynamic membrane effluent turbidity values increased to 20 NTU in the last period. Turbidity in the 90 μm dynamic membrane increased up to 6 NTU, while in the 30 μm dynamic membrane it did not increase similarly to the MF membranes. It has been reported in previous studies that increasing the F/M ratio causes lower consumption of biodegradable organic and colloidal matter (Johir et al., 2012). In this study, although high turbidity removals were achieved in the previous periods, increasing the F/M ratio also increased the amount of substances escaping through the membrane. This caused the turbidity values to increase. However, since the 30 μm dynamic membrane performed similarly to MFs, particle escape in this membrane did not change despite all the variability in the reactor.

3.2. Cake formation on DMBRs

Cake formation can be qualified as the formation of a flock of microorganisms thanks to SMP and EPS structures, and their attachment to the support layer thanks to these structures. While the cake layer formed by microorganisms continues their microbiological activities, they simultaneously act as a secondary layer and perform the filtering process successfully. The schematic representation of the cake layer formed on the flat sheet DMBRs used in this study was demonstrated in Fig. 3.

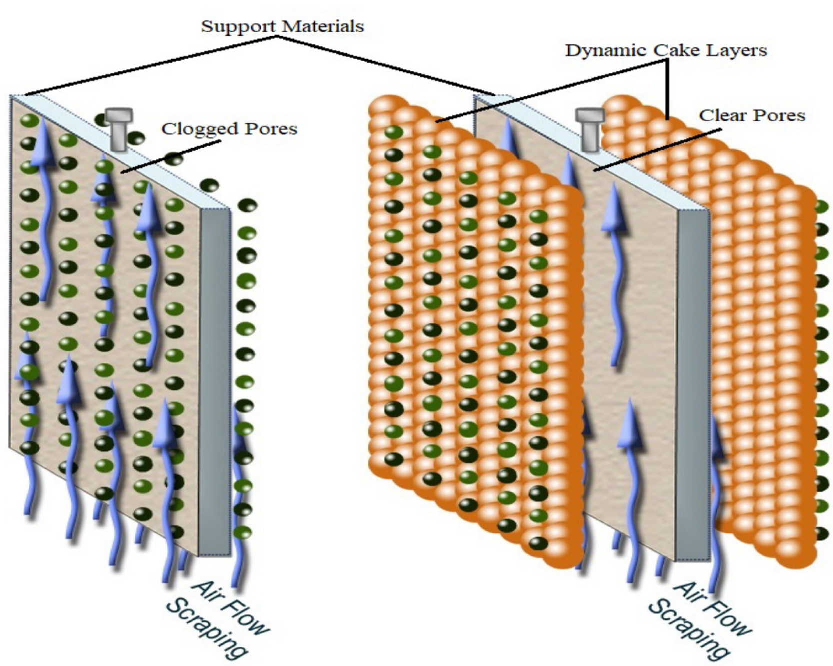

Fig. 3. Cake layer formed on the flat sheet DMBRs schematic representation.

Dynamic membrane formation is a complex structure that involves many and microbiological mechanisms such as internal precipitation, pore-clogging, and cake formation, and the cake layer can act as a protective barrier (Bae and Tak, 2005). Cake formation depends on three main factors: the properties of the support material (type, pore size, structure, etc.), permeate flow rate, and the friction force on the material surface due to the air shear effect (Rezvani et al., 2014). In this study, the changes of cake formation tendency in dynamic membranes were analyzed depending on these main factors. In the first stage, with the gradual increase of flux (15 LMH, 20 LMH and 25 LMH), cake formation on the support materials with different properties was observed. The membranes were started to operate with 15 LMH flux and the first cake formation on the membranes was obtained in this flux. For all membranes, the first cake formation was achieved in the range of 6–8 days, and the effluent turbidity value decreased below 5 NTU after cake formation. As explained in more detail in the previous section, the effluent turbidity remained below 10 NTU in steady-state conditions until the pressure increased in all dynamic membranes in the flux tests periods. Based on the obtained NTU findings, the effect of the support layers used on the filtration removal performance of DM can be deduced as the pore size increased (for 30 μm, 90 μm, 171 μm) the permeate NTU values increased. In this context, when 15 LMH flux was applied, results were obtained as 54.76 NTU, 92.49 NTU, 153.61 NTU for 30 μm, 90 μm, and 171 μm, respectively. In addition, it was obtained 115.29 NTU of turbidity for non-woven (these results included unstable conditions). Similar results were obtained for support materials when LMH, SADm and F/M ratios were changed throughout the study. Particularly, the effluent turbidity value of 30 μm dynamic membrane was below 1 NTU, which was very close to the effluent turbidity value of MF membranes. In the first cake formation process, the highest cake formation rate was obtained at 30 μm dynamic membrane (6.37 NTU/h), as expected. In other membranes, the initial cake formation rates were 5.58 NTU/h, 3.38 NTU/h and 2.80 NTU/h for non-woven, 171 μm and 90 μm, respectively (Fig. 4). In the studies, it is reported that the pore size of the support material is one of the important factors affecting the cake layer formation (Ersahin et al., 2013; Hu et al., 2016; Mahat et al., 2020). Ersahin et al. (2013) stated that small pore size has a higher potential for cake layer formation on the support material. In another study, dynamic membrane formations on 75, 48 and 38 μm pore sized filter cloth were completed in 90, 30 and 10 min, respectively (Hu et al., 2016). In the study, it has been reported that it is required a longer time for the formation of the dynamic cake layer in the membrane with higher pore size. According to Mahat et al. (2020), on the other hand, it was found that 20 μm pore size (lowest pore size) monofilament filter cloth is the most suitable to develop of cake layer. However, cake formation rates increased in all membranes

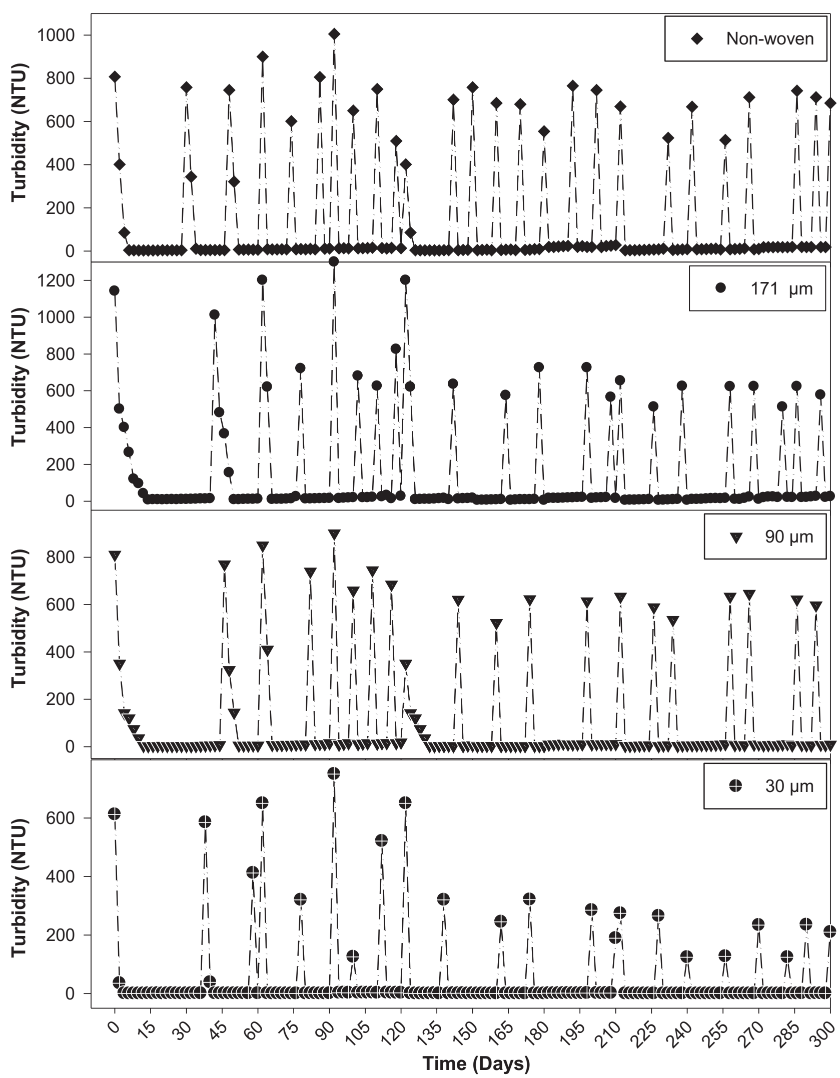

Fig. 4. Turbidity alteration and cake formation rates throughout the study.

after physical cleaning due to the increase in pressure in each membrane. The reason for this is that fouling, which cannot be removed by physical cleaning, plays an active role in cake formation. An important point that draws attention is that, after physical cleaning, the cake formation rate in the non-woven membrane (7.78 NTU/h) was higher than the 30 μm dynamic membrane (6.09 NTU/h). This result actually means that the physical cleaning is performed better on the 30 μm membrane, and in a study it was stated that the pore blocking is higher in non-woven membranes (Zhi-Guo et al., 2005). These results obtained in this study are also in agreement with the literature. After physical cleaning, cake formation rates increased in other membranes as well, resulting in 5.22 NTU/h and 5.31 NTU/h for 171 μm and 90 μm dynamic membranes, respectively (Fig. 4).

Although cake formation rates increased due to the increase in flux, as explained in the next section. Pressure increases also occurred frequently at high fluxes. Especially when the flux was 25 LMH, cake formation accelerated, and frequent fouling was occurred. For all dynamic membranes, while the average fouling frequency at 15 LMH flux was 18.5 ± 1.65 days, when the flux was increased to 20 LMH and 25 LMH, the average fouling frequency increased to 16 ± 2.82 days and 8.5 ± 0.87 days, respectively (Fig. 6). Fouling frequency can be defined as the time elapsed between two cleanings. By increasing the flux, the pressure increases faster and the time between cleanings decreases. This trend was the same for all support material (Fig. 6). For these reasons, the effect of chemical cleaning on the cake formation rate was investigated by applying chemical cleaning to the membranes in addition to physical cleaning in this period. Although there was a decrease in the cake formation rate when chemical cleaning was applied, no significant change was observed in the fouling frequency.

Low aeration rates provide more stable cake layer formation and thus lower effluent SS and filtrate quality. However, at low aeration rates, the cake thickness may increase and the pressure increase may occur more rapidly. Moreover, in a study, it was found that protein and carbohydrate concentrations increased at high aeration rate. Also, it was reported that a net decrease in floc size with an increase in aeration rates. In the study, the particle size decreased from 43 μm to 23 μm by increasing the aeration rate (Etemadi and Yegani, 2019). In this study, the effect of SAD_m on cake formation at the optimum flux (non-woven and 90 μm membrane at 15 LMH, 171 μm and 30 μm membrane at 20 LMH) for each dynamic membrane was investigated after 120 days. First of all, studies were started for the optimum flux for each membrane with a SAD_m value of 1 m^3-air /m^2·h. In this period, the frequency of fouling was found as 19.5 ± 2.18 days on average of all membranes. By decreasing the SAD_m value by 0.8 m^3-air/m^2·h and 0.5 m^3-air /m^2·h, the cake formation rate increased, and the fouling frequency also increased (12.5 ± 1.66 days). Although the higher air applied to the membranes slows down cake formation, it also delays fouling due to causing physical cleaning. For this reason, although faster cake formation rate is desired in dynamic membranes, slower fouling rates are preferred as high fouling rates also affect system stability. For this reason, it is important to determine the optimum SAD_m values and to operate the dynamic membranes at those values. In the study, the most stable operation was obtained at 1 m^3-air/m^2·h.

Aeration rate has a bidirectional effect on fouling in membrane bioreactors. At higher aeration rates, while the fouling accumulation on the membrane surface decreases due to the shear effect of the bubble air (Etemadi and Yegani, 2019), at the same time it causes the formation of smaller sized particles. According to the literature, smaller sized particles decrease the cake permeability and cause the pressure to increase faster (Bai and Leow, 2002).

It is known that SMP and EPS are important factors controlling fouling in MBRs, and their main fouling mechanisms include the deposition and consolidation of SMP and EPS on membrane surfaces (Nagaoka et al., 1996). The increase of SMP and EPS in the reactor causes faster and thicker cake formation. In addition, F/M ratio causes changes in colloidal substances and SMP-EPS concentration in the reactor and can affect membrane fouling rates (Johir et al., 2012). In the study, this situation was clearly observed, especially in the non-woven dynamic membrane. As the F/M ratio increased gradually, the SMP and EPS values increased both on the membrane surface and in the permeate (data not shown). Since the non-woven membrane is more suitable for pore-blocking due to its structure, the increase in colloidal material in the reactor caused the non-woven membrane to block more frequently. A more stable operation was obtained in other mesh membranes compared to the non-woven membrane.

3.3. Filtration performances of DMs and MBRs

Critical flux is a commonly used fouling control method to prevent and predict fouling in membrane operation (Matošić et al., 2008). In this study, using the short-term flux step method, the instantaneous pressure increase point ($\frac{\Delta P}{\Delta t}$) was determined by monitoring the trans-filter pressure (TFP) development. For this purpose, the flux was increased gradually at certain intervals and the point at which the pressure suddenly increased was determined. The instantaneous pressure increase points of the dynamic membranes were given in Fig. 5.

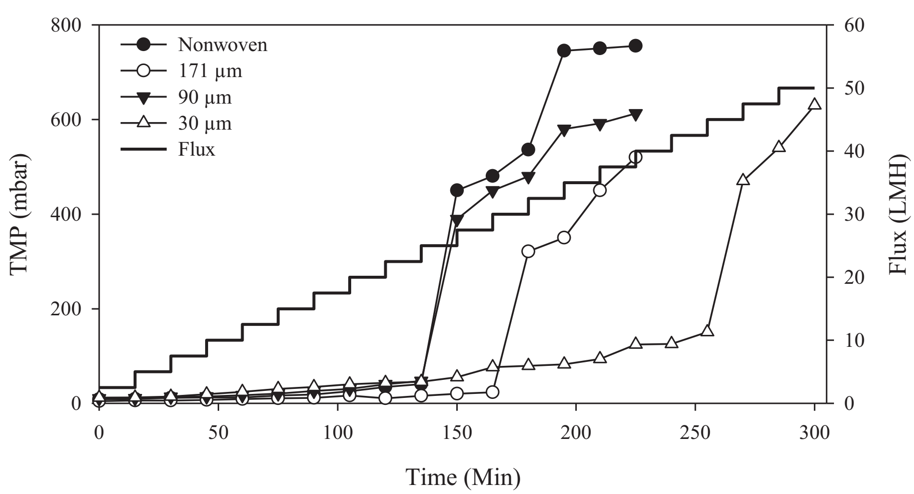

Fig. 5. The instantaneous pressure increase point obtained in dynamic membranes.

Critical fluxes were obtained in non-woven, 171 μm, 90 μm and 30 μm dynamic membranes as 25 LMH, 30 LMH, 25 LMH, and 45 LMH, respectively. Parameters such as reactor operating conditions, SS-VSS concentration, sludge characteristic, wastewater characteristic, etc. affect the critical flux value (Le Clech et al., 2003), and different critical fluxes have been reported for dynamic membranes in different studies in the literature. While the critical flux was obtained as 3.9 LMH in the study performed with 30 μm nylon mesh by Satyawali and Balakrishnan (2008), the critical flux for 38 μm stainless steel mesh was determined as 75 LMH in the study of Chu et al. (2014). In this study, the instantaneous pressure increase point values for non-woven and 90 μm dynamic membranes were obtained at lower levels for 171 μm and 30 μm dynamic membranes. In parallel with this result, the optimum flux was determined as 15 LMH for non-woven and 90 μm dynamic membranes, while the optimum flux was determined as 20 LMH for 171 μm and 30 μm dynamic membranes. When the flux values were increased gradually, it was also observed the increases in the TMP values. Similarly, when the flux values were increased, the permeate NTU values increased as expected. However, when the filtrate NTU values were taken into account, it was exceptionally observed as the optimum flux of 20 LMH on 171 μm instead of 15 LMH. This demonstrated that when the pore size exceeds a certain level, the flux must be at a certain speed (20 LHM) for cake formation. This was one of the important results obtained in the study. These results clearly demonstrate that there is an exact relation between critical flux and optimum flux.

TMP values obtained daily in DM and MBR modules were automatically recorded every minute. TMP values were monitored both daily and instantaneously to obtain information about membrane fouling under constant flux. The TMP results obtained in the study were presented in Fig. 6, green and red color indicates physical cleaning and chemical washing, respectively. Although chemical cleaning was carried out especially in cases where the pressure increased very rapidly and the TMP exceeded 500 mbar, chemical cleaning was generally applied in the last stages of all dynamic membranes. The reason for this is that irreversible fouling has increased considerably from the last stages of the study. However, the

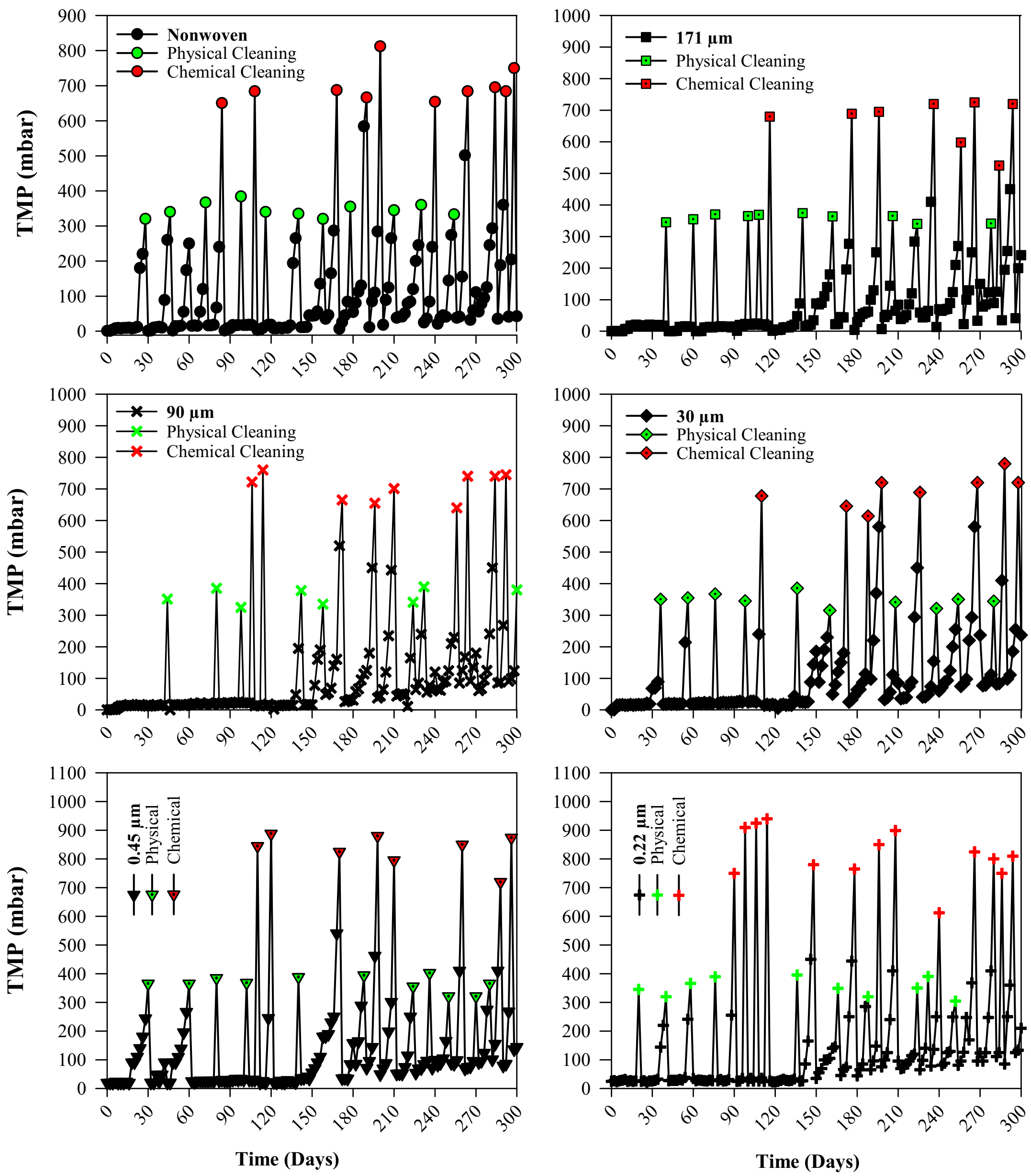

Fig. 6. TMP changes in all membranes.

main difference between DM and MF membranes is that the pressure increases after cake formation in dynamic membranes occur much faster than in MF membranes. In the studies, it has been reported that the TMP change characterized in 3 stages in MBRs operated with the constant flux mode. These stages follow each other as a short-term rapid TMP increase (stage 1), long-term slow TMP increase (stage 2) and again a short-term rapid TMP increase (stage 3, jumping stage) (Lin et al., 2013). However, in dynamic membranes, TMP at low levels until cake formation suddenly increases to 300–500 mbar levels immediately after cake formation. The second and third stages in conventional MBRs are passed quickly in dynamic membranes. Thus, stage 3 pressure increase in conventional membranes was observed in dynamic membranes in stage 2 (long-term slow pressure increase and short-term rapid pressure increase (Jumping stage)). NW support layer material was specially manufactured to considering viscosities and durability and DMs with pore size of 30 μm, 90 μm and 171 μm had the same material (screen printing silk), the only difference being the pore sizes in this study. Some studies were reported that DMs with smaller pore sizes have higher cake formation rates and the TMP increased as the pore size decreased in the support layers made with similar material (Cai et al., 2018). In addition, it was reported that NW membranes were more tend to fouling due to the fact that internal fouling occurs in (Zhi-Guo et al., 2005). Accordingly, similar results were obtained in this study. It was observed that the frequency of fouling of the NW was more than the others due to the non-uniform distribution of the pores. In this context, it was determined that the NW membrane fouled more frequently than the others and required 21 washings during the study, especially when the TMP changes were examined. According to the TMP changes, while the membrane fouling occurred 21 times (maximum) in the non-woven dynamic membrane, it has occurred 17 times (minimum) fouling in the 90 μm dynamic membrane during the study (300 days). The fouling occurred 18 times in the other dynamic membranes. Moreover, while it has occurred relatively high fouling in the MF membranes, it has especially occurred the fouling 23 times in 0.22 μm membrane. As it can be seen in Fig. 6, the highest fouling rates were obtained in the period 3 at 25 LMH flux. Especially, 0.22 μm MF membrane was the most fouled membrane at this stage. Dynamic membranes are developed as an alternative to conventional membranes, the most important purpose of which is to prevent rapid fouling and decrease the frequency of fouling (Zhang et al., 2010). The results obtained in this study also show that dynamic membranes will reduce the frequency of fouling compared to conventional membranes. At the same flux, 0.22 μm and 0.45 μm MF membranes were fouled 23 and 21 times, respectively, while the 30 μm dynamic membrane was fouled 18 times. Despite less fouling, as mentioned in the previous section, it was obtained similar effluent turbidity values in 30 μm dynamic membrane and MF membranes under steady-state conditions. Therefore, as a result of this study, it can be concluded that dynamic membranes can be an alternative to MF membranes.

It has been shown the calculated fouling rates based on the TMP changes for each fouling cycle in Fig. 7. In general, while the fouling rates increase as a result of successive physical cleaning in all membranes, after chemical cleaning fouling rates decrease slightly.

In the study, on average, the lowest fouling rate was obtained in 30 μm dynamic membrane (36 ± 20.65 mbar/day), while the highest fouling rate was obtained in 0.22 μm MF membrane (56 ± 35.24 mbar/day). For other dynamic membranes, it was calculated as 43.88 ± 29.31 mbar/day, 36.45 ± 21.93 mbar/day and 45.52 ± 31 mbar/day for non-woven, 171 μm and 90 μm dynamic membranes, respectively. In addition, the effects of flux, SAD_m and F/M conditions on filtration performance were investigated in the study. The fouling rate, which was at very low levels in the first fouling cycle in virgin membranes, increased both with the extended operation and with changing operating conditions. As seen in Fig. 7, while the fouling rate was around 15 mbar/day for all membranes in the period when the flux was 15 LMH, it increased to 20 mbar/day with a very small increase when the flux was increased to 20 LMH. However, the fouling rate in all membranes increased significantly and reached 80–100 mbar/day by increasing the flux to 25 LMH. In this period, the maximum fouling rate was obtained at 0.22 μm MF membrane (average 115.63 ± 1.53 mbar/day). In addition, there was an increase in the fouling rates and deterioration in the stability of the reactor depending on the decrease of SAD_m value. This situation causes the turbidity to increase as mentioned in the previous section. Increasing the F/M also had reverse effects in the reactor. When F/M was increased from 0.095 g-COD/g-MLSS·day to 0.125- and 0.19 g-COD/g-MLSS·day, the fouling rate increased from 20 to 40 mbar/day to 40–60 mbar/day and 80–120 mbar/day, respectively (Fig. 7). Trussell et al. (2006) stated that membrane fouling increased at high organic loading rates and they reported that the membrane fouling rate increased 20 times when the F/M ratio was increased 4 times. In another study, it was emphasized that the accumulation of hydrophilic compounds on the membrane surface at higher OLR may cause membrane fouling and faster TMP increase (Pan et al., 2010).

4. Conclusions

DMs do not meet the discharge standards during the period until cake formation. For this reason, in this study mainly focused on the investigation of cake formation /reformation rates. Besides, unlike similar studies, six

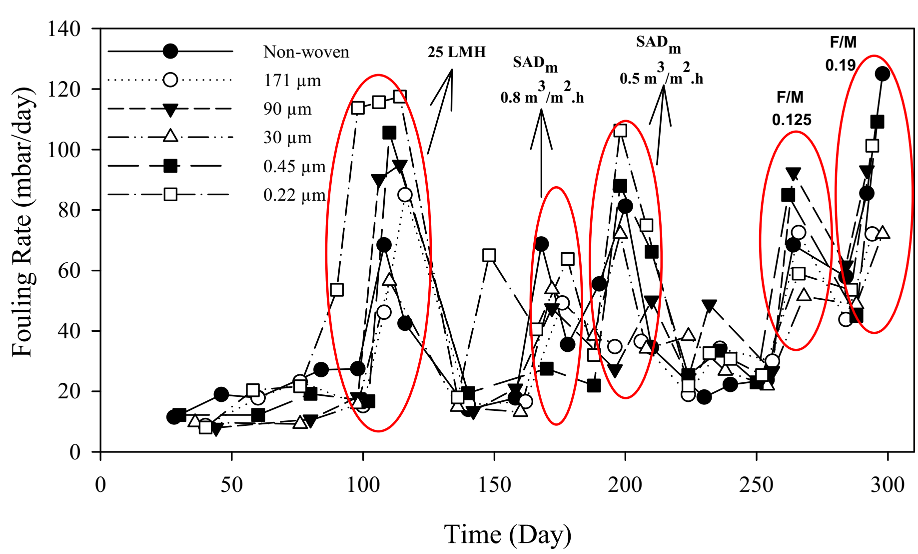

Fig. 7. The calculated fouling rates based on the TMP changes for each fouling cycle.

different modules were operated simultaneously in a single reactor in order to observe the DMs and MFs operations under exactly the same conditions. This study aimed to evaluate the filtration performance of different pore sized dynamic membranes (NW, 171 μm, 90 μm and 30 μm) and to compare their performances with MF membranes (0.45 μm and 0.22 μm). In the study, it was obtained 99% removal efficiency for MF membranes, while COD removal efficiencies between 90% and 99% were obtained in DMs in steady-state conditions. When the SAD_m value was decreased, the COD removal also decreased and it decreased to 83% at 171 μm dynamic membrane. Turbidity removal was performed successfully, and turbidity values were obtained below 10 NTU throughout the study. In particular, the 30 μm DM performance was observed very close to the MF membranes in all periods due to the high cake formation rate (6.37 NTU/h for a clean membrane). Despite the highest cake formation rate, the lowest fouling rate (average 36 ± 20.65 mbar/day) was obtained at 30 μm DM. Increasing the flux and F/M and decreasing the SAD_m value caused an increase in fouling rates in all membranes. In the study, the reformation rates were investigated, it was obtained very close results to MF membranes, especially at 30 μm DM, in terms of filtration and treatment performance, and it was shown that DMs can be used as an alternative to traditional MF membranes. It is thought that the results obtained from DMs, which used much cheaper materials compared to MFs, can guide future full-scale and lab-scale studies and provide an insight to researchers. Besides, the reformation rates findings obtained in the study can shed light on DMBR studies using different support materials and different pore sizes on cake reformation/formation rates.

CRediT authorship contribution statement

Abdullah Kizilet: Investigation, Formal analysis, Data Curation, Methodology, Conceptualization. **Adem Yurtsever:** Data Curation, Methodology, Writing - Review & Editing. **Kevser Cirik:** Supervision, Conceptualization, Methodology, Resources. **Ozer Cinar:** Conceptualization, Resources, Writing - Review & Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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