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### **Research Article**

# Treatment of textile wastewater in combined granular activated carbonmembrane bioreactor (GAC-MBR)

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#### ABSTRACT

In recent years, the membrane bioreactor (MBR) process has been seen as a promising technology for the treatment of both municipal and industrial wastewater, including textile wastewater which has the potential to generate high levels of pollution in the receiving environment. However, membrane fouling during MBR operation is seen as one of the most important drawbacks due to the reduction of membrane flux. In this study, granular activated carbon MBR (GAC-MBR) technology was investigated to treat real textile wastewater. In this context, conventional MBR (R1) and GAC-MBR (R2) with GAC (300 mg) were operated for 48 days. A flat-plate ceramic membrane module was used in both reactors. The chemical oxygen demand (COD) and color removal efficiencies were found to be  $87\pm3\%$  and  $73\pm7\%$  in conventional MBR, whereas these pollutant removal efficiencies were determined as  $89\pm6.4\%$  and  $78\pm4.8\%$ , respectively, in the GAC-MBR process. According to the results obtained, while conventional MBR required physical cleaning every other day, GAC-MBR did not require any cleaning after the addition of GAC. It was also observed that GAC had no direct effects on the excretion of soluble and loosely-bound or tightly-bound extracellular polymeric substances, however, reduced the transmembrane pressure, capillary suction time, and membrane fouling propensity.

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### INTRODUCTION

The textile industry is of great importance for economic development all over the world [1]. This industry consumes high amounts of water during the production processes, therefore, appropriate treatment processes should be applied before its discharge to receiving media [2]. An average textile factory producing 8,000 kg of fabric per day consumes approximately 1.6 million liters of water, of which 24% is due to these processes [3]. Textile wastewater contains various pollutants such as dyes, alkalis, acids, hydrogen peroxides, surfactants, and dispersants. These contaminants are usually of complex chemical origin and originate mainly from dyeing and finishing processes [4,5]. Dyes are mostly aromatic and heterocyclic compounds

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and make treatment difficult due to their stable and complex structures [2]. Textile wastewater, which is generally rich in color, has a high pH due to various chemical species. Due to these adverse properties, many countries have strict discharge standards for textile wastewater [5]. Textile dyes have a significant impact on the quality of water bodies where they are released, resulting in increased oxygen demand, impaired photosynthesis, inhibited plant growth, and toxic effects. The dyes can also cause mutagenicity and cancer through the food chain and bioaccumulation [6]. Discharging untreated colored textile wastewater can harm the ecosystem by blocking sunlight penetration in natural water bodies. The key pollutants in textile wastewater include color, COD, sulfates, chlorides, and NH<sub>3</sub>-N [7].

The character of textile wastewater is very different due to the differences in the chemical structure of the dyes and the diversity of the dyeing process. The adsorption of dyestuffs on yarn varies depending on the textile yarn and the type of dye. The combination of dyes of different chemical classes for a single dyeing process further complicates the composition of the effluent. Reactive and acid dyes, which are brightly colored and soluble in water, are the most environmentally problematic as they cannot be removed by conventional treatment systems [8].

Various treatment methods are available to treat textile wastewater and meet the required discharge standards. The selection of an appropriate treatment technology depends on the specific discharge criteria and the characteristics of the wastewater. Generally, three treatment methods are used for textile wastewater: physical, chemical, and biological treatment. Often, a combination of these methods is necessary to achieve the desired treatment efficiency. [9]. Physical methods commonly used in the treatment of dye-containing textile wastewater are adsorption, ion exchange, and membrane filtration, which can achieve high color removal efficiencies of 85-99% [10, 11]. Physical methods have many advantages such as requiring a simple design, ease of use, low cost, and fewer chemical requirements [12]. On the other hand, these methods have several disadvantages such as toxic by-products and sludge production and limited applicability areas [11]. Common chemical treatment methods used in the treatment of textile wastewater are chemical precipitation, flocculation, and oxidation [11, 13]. Apart from electrochemical treatment technology, these chemical methods typically cost more than physical and biological methods [11, 12]. The major disadvantages of using chemical methods for highly efficient dye removal from textile wastewater are high energy requirements, large quantities of chemicals used, and the need for appropriate equipment. It also creates additional challenges due to toxic metabolites and by-products during chemical treatment methods [11]. Biological treatment processes are commonly used in treating textile wastewater due to their low removal and operating costs, non-toxic product formation, and less sludge production. However, they are less effective in removing color from wastewater.

Although some basic, direct, and azo dyestuffs are not biodegradable, they can be adsorbed and removed from the wastewater by microorganisms. [14]. Textile wastewater can be effectively treated using aerobic or anaerobic biological processes following physical or chemical pre-treatment. Activated sludge processes can be used to treat textile wastewater to meet local discharge criteria, but the resulting water may not be suitable for reuse in the textile production process. Therefore, advanced treatment alternatives should be evaluated and compared to conventional methods to ensure that the water is of sufficient quality for reuse [15]. Membrane bioreactor technology (MBR), one of the innovative technologies, has wide application possibilities in the treatment of textile wastewater [16].

Membrane bioreactor (MBR) is widely used for the treatment of various wastewaters as it can be operated at high biomass concentrations and produces high quality treated wastewater [17]. Today, MBRs have become an attractive alternative for the treatment and reuse/recycling of municipal and industrial wastewater compared to the traditional activated sludge process. The MBR technology combines membrane filtration with direct solid-liquid separation with biodegradation in a bioreactor [18]. The MBR process has several advantages such as small footprint, low maintenance cost, consistency in the final treated water quality, lower amount of sludge production, and higher efficiency removal of nutrients, organic and persistent organic pollutants compared to conventional activated sludge processes. On the other hand, fouling of the membrane leads to a decrease in permeate flow or an increase in trans-membrane pressure (TMP) during the operation. This is the main disadvantage of the MBR process as it leads to higher operating costs for membrane cleaning and consequently shortens the lifetime of the membrane [5]. To overcome membrane fouling phenomena as well as improvement of treatment performance, granular or powdered activated carbon (AC) may be added to the MBR [19].

AC addition to the membrane bioreactor has resulted in significant improvements in decreasing membrane fouling [20]. In the literature, it is concluded that the important when adding AC to a membrane bioreactor is the size. In the study conducted by Johir et al. [21], the effect of granular AC applied in variable sizes on MBR was investigated. With the addition of different particle sizes used, 95% efficiency was achieved in COD and dissolved organic carbon removal, while it was concluded that 75% of humic substances were removed. Similarly, another important point is that AC positively affects membrane fouling [22]. Johir et al. [23] extensively studied the effect of AC on membrane fouling. Unlike the previous study they carried out, in the current study, biopolymer formation was prevented by using granular activated carbon (GAC) and the process efficiency was increased. Pradhan et al. [24] also concluded that GAC filtration reduces membrane fouling by 85% in membrane bioreactor studies.

In this study, conventional MBR (R1) and GAC-MBR (R2) were investigated to better understand the effects of AC on the process performance and membrane fouling during the treatment of real textile wastewater. In this context, COD and color removal efficiencies were monitored to determine the process performance differences between MBR and GAC-MBR. On the other hand, transmembrane pressure (TMP) and extracellular polymeric substances (EPSs) and their species namely soluble EPS (S-EPS), loosely-bound EPS (LB-EPS) and tightly-bound EPS (TB-EPS), and capillary suction time (CST) were monitored to determine membrane fouling propensity and filtration characteristics.

#### MATERIALS AND METHODS

#### **Experimental Set-up and Operational Conditions**

In this study, the treatment of textile wastewater with a membrane bioreactor was investigated and the treatment of a classical membrane bioreactor and a membrane bioreactor with added AC was compared. The study was carried out on a laboratory scale and two MBR systems were installed in parallel. The systems were operated for 48 days and the first reactor (R1) was used as a control reactor. The washed and dried AC was added to the second reactor (R2) on the 27th operating day. The reactors were cylindrical with an internal diameter of 17 cm and an effective volume of 5 L. The ceramic membranes with a nominal pore size of 0,1 µm obtained from (Cembrane, Denmark) were submerged in the reactors. The effective filtration area of ceramic membranes was 0,057 m<sup>2</sup>. Water was circulated with a water bath through the outer jacket of the reactors so that the temperature of the reactors was kept at 20±2°C. A stainless-steel air diffuser was placed at the bottom of the reactors, and an air pump was used to supply dissolved oxygen (DO) to the system at a range of 3.5 to 5.4 mg/L. Wastewater fed to the reactors was provided from a textile factory in Halkali, Istanbul. The initial inoculation sludge was obtained from the aeration tank of the Istanbul Atakoy Advanced Biological Wastewater Treatment Plant. The properties of textile wastewater used in the study are shown in Table 1.

Table 1. Textile wastewater ch	aracteristics
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Parameter	Value	Standard Deviation
pН	7.5	0.4
Conductivity (mS/cm)	2.6	0.2
COD (mg/L)	1680	435
$BOD_5 (mg/L)$	590	20.5
$\rm NH_4$ -N (mg/L)	12.7	1.1
Orthophosphate (mg/L)	0.8	0.02
Colour (Pt-Co)	930	115
Chloride (mg/L)	420	1.2

Period	<b>Operational Conditions</b>	R1	R2
1	SRT (day)	30	30
	HRT (h)	26.43±11.2	25.21±6.65
	Net flux (L/m2.h)	1.19±0.31	$1.20 \pm 0.31$
2*	SRT (day)	30	30
	HRT (h)	18.05±2.06	17.92±2.29
	Net flux (L/m2.h)	1.60±0.19	$1.61 \pm 0.17$

Table 2. Operational conditions of MBRs

\*GAC was added to R2 reactor on the 27th day of operation.

For the adaptation of the inoculum to the textile wastewater, the reactors were operated for a while without throwing excess sludge from the reactors. Afterwards, the reactors were operated at 30 days sludge retention time (STR) and constant flux. An intermittent filtration cycle of 30-minute suction and 30-minute relaxation was applied to control and reduce the fouling of the membranes in the reactors. GAC (Jacobi Carbons branded Aquasorb 1000 series) having 0.595-2.38 mm diameter was used in the study after being downsized to a 300-600µm diameter range in size, then washed with distilled water and kept in the drying oven for 1 hour followed by desiccation for 30 minutes. Starting from the 27th day of the system, 300 mg of AC corresponding to the volume of excess sludge removed from the reactor was added to the R2 reactor every day. After adding AC to the R2 reactor, both reactors were operated for another 21 days. The operating conditions of MBRs are given in Table 2. The HRT differences between R1 and R2 reactors was the operational.

#### Membrane Cleaning Procedure

The transmembrane (TMP) pressure values of the ceramic membranes in the reactors were monitored online by a negative pressure gauge manometer integrated with an automation system. When the TMP values of the membranes are above 0.6 bar, the targeted hourly flow rate during the operation could not be achieved and the permeate volumes at the outlet of the MBR systems decreased. For this reason, when the TMP values of the membranes reached 0.6 bar pressure, the membranes were physically washed with tap water and a sponge followed by a backwash with 2 bar pressured air [25].

#### Analytical Methods

The dissolved oxygen (DO), temperature, pH, and electrical conductivity (EC) values of the raw textile wastewater used in the study and the permeate water from the reactors were measured daily by WTW Multiline P4 Multimeter with CellOx 325 DO probe, SenTix 41 pH probe, and EC probe. Mixed liquor suspended solids (MLSS) analysis of activated sludge 4 times a week and mixed liquor volatile suspended solids (MLVSS) analysis of activated sludge once a week were carried out according to the Standard Methods [26].

To determine the removal efficiency in the reactors, COD and color analyzes were carried out on the raw wastewater and permeate wastewater at certain periods. The COD analyzes were performed 4 times a week according to the 5520 C Closed Reflux Titrimetric method by the Standard Methods [26]. The color was analyzed at a wavelength of 455 nm using the Hach Lange DR 5000 spectrophotometer. All other analyses such as ammonia nitrogen, orthophosphate, chloride, and 5-day biochemical oxygen demand (BOD<sub>5</sub>) were carried out according to the Standard Methods [26]. All the analyses were repeated in duplicate and the mean values were used.

The extracellular polymeric substances secreted from microorganisms in the mixed liquor were analyzed according to the procedure adopted by Zhou et al. [27] and Liu et al. [28] as S-EPS, LB-EPS, and TB-EPS with two main fractions (protein and polysaccharide). First of all, in order to extract dissolved EPS (SB-EPS), activated sludge samples were centrifuged at 8000 rpm at 4°C for 15 minutes, and the supernatant formed was filtered using a 0.45 µm syringe. Up to 50 mL 0.05% NaCl solution was added to the sludge pellet remaining in the tube and the resulting sludge suspension was kept in a vortex device until homogeneous and then kept in a water bath set at 70°C for 1 minute. Later on, it was centrifuged at 8000 rpm for 10 min at 4°C. The formed supernatant was filtered with a 0.45 µm syringe and loosely bound EPS (LB-EPS) was extracted. For the tightly bound EPS (TB-EPS) extraction, up to 50 mL of NaCl solution (0.05%) was added to the last sludge pellet remaining in the tube, and a vortex was applied to homogenize the sludge suspension. Afterwards, it was kept in a water bath set at 60 °C for 30 min. Then, it was centrifuged at 8000 rpm at 4°C for 15 minutes and the resulting supernatant parts were filtered with a 0.45 µm syringe. Protein (PN) analysis by Lowry Method [29] and polysaccharide (PS) analysis by modified phenol-sulfuric acid method [30] were performed on supernatant samples collected and filtered after EPS extractions.

Capillary suction time (CST) analysis was measured using Triton CST instrument according to the Standard Methods [31] to determine the filterability of the activated sludge in the reactors.

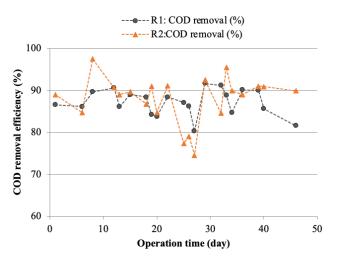
#### **RESULTS AND DISCUSSION**

#### **Treatment Performances of MBR and GAC-MBR**

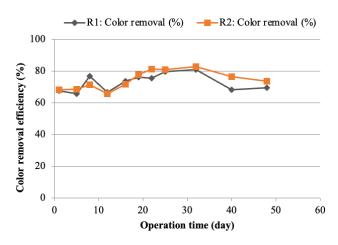
In the study, the performances of both reactors were determined by monitoring COD and color removals. The reactors (R1: Control and R2: Test) were operated as conventional MBR for the first 27 days after the start-up of the operation for acclimation of inoculum to real textile wastewater. GAC (300 mg) was added to the R2 reactor daily and was operated as GAC-MBR from day 27. Figure 1 shows the COD concentrations and removal efficiencies in raw and MBR effluent. The COD removal efficiency was found as  $87\% (\pm 3\%)$  for the R1 reactor throughout the operation. For the R2 reactor, the average COD removal was 88%±5.5% during the 1st period when the reactor was operated as a conventional MBR. The average COD removal was also found to be 89%±6.4% during the 2nd period when it was operated as GAC-MBR. Figure 2 demonstrates color removal efficiencies and color concentration in the effluent. The color removal efficiencies in the 1st period were found as 72.7±5.3% and 73±5.6%, respectively for the R1 and R2 reactors. The average color removal efficiency of the R2 reactor was determined as 78±4.8% after GAC was added, while color removal efficiency was found still at 73±7% in the R1 reactor. Considering these results, it was observed that COD and color removal slightly increased after adding GAC to the MBR reactor.

The mean COD concentration in the effluent of the control reactor (R1) operated as conventional MBR throughout the study was determined as 208±55 mg/L. The average COD concentration in the effluent was 190±64 mg/L in the period when the R2 reactor was operated as a conventional MBR, and the average COD concentration in the effluent was 177±71 mg/L in the 2nd period when the reactor was operated as GAC-MBR. The COD concentration of treated textile wastewater according to the Turkish discharge standards should be lower than 240 mg/L (for 24-h composite sample for fiber and yarn production and finishing), and when the effluent COD values obtained from the study with this discharge standard are compared, it was observed that both MBR and GAC-MBR system was sufficient to meet the standards.

The color concentrations in effluents were found to be 244±52 Pt-Co and 241±61 Pt-Co during the 1st period, respectively for the R1 and R2 reactors. In the R1 reactor, the color concentration was measured as 278±99 Pt-Co in



**Figure 1.** COD concentrations and removal efficiencies for effluent in MBR and GAC-MBR throughout the study.



**Figure 2.** Color concentrations and removal efficiencies for raw wastewater and effluent in MBR and GAC-MBR throughout the study.

2<sup>nd</sup> period, while it was found as 229±72 Pt-Co for the R2 reactor. When these values are compared with the effluent color concentration in local discharge standards (260 Pt-Co), it has been observed that MBR and GAC-MBR systems also met this parameter.

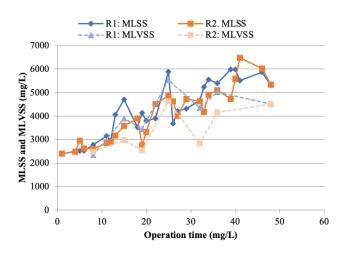
Several studies in the literature focused on the MBR combined with PAC or GAC in textile wastewater treatment [2, 19, 32]. In the study conducted by Kozak et al. (2021) sequencing anaerobic moving bed biofilm reactor (AnMBBR) and powder activated carbon-aerobic membrane bioreactor (PAC-AeMBR) were investigated for the treatment of textile wastewater [2]. In the study, the AnMBBR effluent was fed to the PAC-AeMBR and 89% dissolved organic carbon (DOC) removal was obtained only with AnMBBR process, while total removal efficiency reached 96% after PAC-AeMBR treatment. The PAC-AeMBR was found successful in color removal with a filtration effect of over 65% and the total removal efficiency of over 90% corresponding to 30 Pt-Co color values in the effluent. Fouling in AeMBR increased when HRT was reduced, but improved by 90% under PAC supplementation conditions. The MLSS concentration, which was  $5000 \pm 500$ mg/L at the beginning of the AnMBR treatment, increased to 7500  $\pm$  500 mg/L with the addition of PAC (0.26 g/g MLSS). The obtained results show that it is an important option for the treatment of real textile wastewater that meets the discharge standard quality for the textile industry in Turkey (260 Pt-Co). The hybrid system has greater stability at a short HRT (18 hours) and better removal of both color (95%) and DOC (96%) compared to conventional systems, with both biofilm and membrane advantages.

In another study carried out by Hai et al. (2012) in which adding GAC on the membrane module in a wholecell fungal membrane bioreactor (MBR) was set up for color removal from dye wastewater [19]. Depending on the initial conditions, up to 50% enzymatic dye degradation was achieved by the co-adsorption of the enzyme and dye on the GAC. In a control MBR, the dye removal efficiency was variable; in contrast, 85% of dye removal was achieved by using membrane-coupled MBRs with additional layers of GAC. In the study, a combined critical effect on the dye removal efficiency of the hydraulic retention time (HRT), instantaneous membrane flow, and amount of GAC coating was observed. Although the dye loading under different HRTs was kept the same by varying its concentration in the synthetic wastewater, the better removal efficiency was obtained in the case of longer HRT. As a result of the evaluation of MBR performance enhancement due to the use of a GAC-coated membrane, which is the main objective of the study, it was reported that additional GAC layers on the membrane module were effective in preventing fungal enzyme washout and improving discoloration.

# Variation of MLSS and EPS Concentrations Throughout the Operation

The variations of MLSS and MLVSS concentrations throughout the operation in conventional MBR and GAC-MBR systems with added AC are given in Figure 3. At the beginning of the study, the MLSS concentrations of the two reactors operated simultaneously as conventional MBRs were 2390 mg/L and 2400 mg/L for R1 and R2, respectively. It took 27 operating days to reach the steady-state conditions. The average MLSS concentrations in reactors R1 and R2 were, then, found around 3490 and 2970 mg/L. After the addition of AC to the R2 reactor, significant fluctuations were observed in the MLSS concentration. Although fluctuations were observed in both reactors during the operation, in general, the MLSS concentration values measured in the R1 reactor were higher than the values measured in the R2 reactor. The average MLSS concentrations in the reactors were obtained as 5281mg/L and 5154mg/L for reactors R1 and R2, respectively. MLVSS analyses of the sludge were made during the entire operation and are shown in Figure 3. The MLVSS/MLSS ratio for the R1 reactor varied between 83% to 94% (average value 88%), while the MLVSS/MLSS ratio for the reactor R2 ranges from 81% to 92% (average value 85%). The addition of GAC to the MBR may have led to a slight decrease in MLSS and MLVSS concentration. When GAC is added to the MBR system, it can provide additional surface area for microbial growth and adsorption of organic matter. As a result, the microorganisms may consume more organic matter, leading to a slight decrease in MLSS and MLVSS concentrations. This can have both positive and negative effects on the system, as lower concentrations of organic matter and microorganisms can reduce fouling and improve membrane performance, but may also result in reduced treatment efficiency and higher membrane blockage or abrasion due to the higher turbidity in supernatant especially added PAC in MBR [33].

Extracellular polymeric substances (EPSs) secreted by microorganisms in mixed liquor have an important role in MBR operation because they play an active role in



**Figure 3.** MLSS (Suspended Solid) and MLVSS (Volatile Suspended Solids) concentrations of mixed liquor throughout the operation.

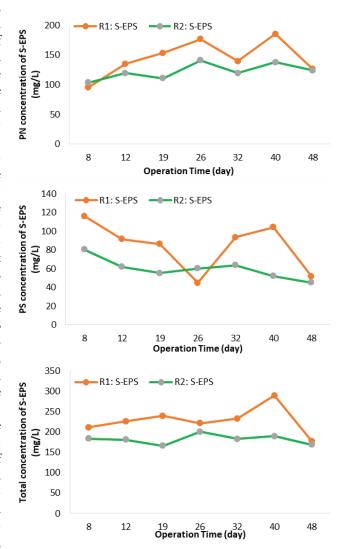
membrane obstruction and fouling. As seen in the literature, it is one of the analyses used to create a control mechanism [34]. On the other hand, the most important fractions of EPS containing soluble, loosely-bound and tightly-bound polymeric matters are protein (PN) and polysaccharide (PS), which are the main components of EPS, which are secreted up to 90% in activated sludge. Apart from the main components, DNA, RNA and phospholipids are also present in EPS components, albeit at lower rates [35].

Figure 4 shows the variation of PN, PS and total concentrations of S-EPS. Initial PN concentrations of S-EPS were measured as 95.3 and 103.6 mg/L, after that PN concentration of S-EPS increased to 176.5 and 140.8 mg/L for the R1 and R2 reactors. Then, the PN fraction of S-EPS suddenly decreased to 139.7 mg/L on day 32 in the R1 reactor which was operated as conventional MBR throughout the operation. Similarly, the concentration of PN of S-EPS decreased to 119.4 mg/L on the day 32 in the R2 reactor and then increased to 185.2 mg/L and 137.75 mg/L for both the R1 and R2 reactors, respectively. On the other hand, the PS concentrations of S-EPS were determined as 116 and 80.5 mg/L for the R1 and R2 reactors on the operation day 8, then the values decreased to 44.9 and 60.15 mg/L for both reactors on day 27 at which 300 mg AC were dosed to the R2 reactor. After that, the PS fraction of S-EPS in the R1 reactor increased on the day 40, and then a sudden decrease was observed for this reactor. Conversely, no sudden variations were observed in the R2 reactor for PS fraction of SB-EPS. After the addition of GAC, the values decreased slightly. Moreover, the PS values of S-EPS decreased similarly to the PN fraction of S-EPS in the R1 reactor. As seen from the last plot of Figure 4, the total S-EPS concentrations remained almost stable until the 40<sup>th</sup> operating day, after that a sudden increase was observed for the R1 reactor. The reason why no significant change was observed in the

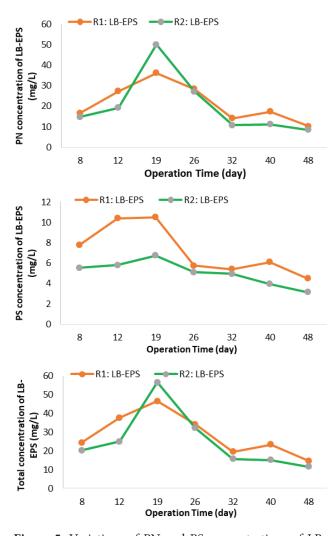
total S-EPS concentration is thought to be due to the addition of AC to the R2 reactor.

As seen from Figure 5, total LB-EPS concentration and their PN and PS fractions dramatically decreased after day 19. PN concentration of LB-EPS increased from 16.6 mg/L to 36.1 mg/L on the days between the 8<sup>th</sup> and 19<sup>th</sup> in the R1 reactor. A similar trend was also observed in the R2 reactor and the PN concentration of LB-EPS increased from 14.75 mg/L to 49.95 mg/L on the days between the 8th and 19th. As can be seen from Figure 5, dosing GAC to the R2 did not significantly affect the concentration of LB-EPS and their fractions.

The TB-EPS concentrations of each reactor are shown in Figure 6. As shown in Figure 6, the as seen in Figure 6, the PN fraction of TB-EPS was stable until the 27<sup>th</sup> day of operation, after which it decreased from 106.9 to 72.3 mg/L in the R1 reactor. In the R2 reactor, the PN concentration of TB-EPS remained almost stable during the operation. In



**Figure 4.** Variations of PN and PS concentrations of S-EPS throughout the MBR and GAC-MBR operation.



R2: TB-EPS R1: TB-FPS 120 PN concentration of TB-EPS 100 80 (1/8m) 40 40 20 0 40 8 12 19 26 32 Operation Time (day) 48 R2: TB-EPS R1: TB-EPS 50 PS concentration of TB-EPS 40 30 (mg/L) 20 10 0 19 26 32 Operation Time (day) 40 8 12 48 R2: TB-FPS R1: TB-EPS 160 Total concentration of TB-140 120 EPS (mg/L) 100 80 60 40 20 0 8 12 19 26 40 48 Operation Time (day)

**Figure 5.** Variations of PN and PS concentrations of LB-EPS throughout the MBR and GAC-MBR operation.

terms of PS concentrations of TB-EPS, no significant values could be obtained for the R1 reactor as the observed PS concentration varied from day to day of EPS analysis. In terms of PS concentrations of TB-EPS, no significant values could be obtained for the R1 reactor as the observed PS concentration varied from day to day of EPS analysis. On the other hand, in the R2 reactor, the PS of TB-EPS varied between 36.1 and 41.2 mg/L and the average PS of TB-EPS concentration was 39.1 mg/L from the 8<sup>th</sup> day to the 48<sup>th</sup> day. It may be said that GAC equilibrated the PS fraction of TB-EPS according to obtained data.

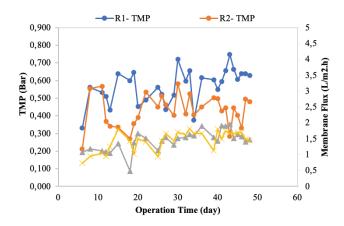
As it was reported that the addition of powdered activated carbon (PAC) to an MBR decreases the viscosity of mixed liquor and increases the sludge floc size, resulting in lower TMP values [36]. The addition of AC to the MBR also reduces the EPS concentration, slows the development of cake resistance and mitigates membrane fouling [36, 37]. In addition, it also reduces the SMP concentration, slows the concentration polarization layer resistance and prevents further membrane fouling [38]. The studies also show that

**Figure 6.** Variations of PN and PS concentrations of TB-EPS throughout the MBR and GAC-MBR operation.

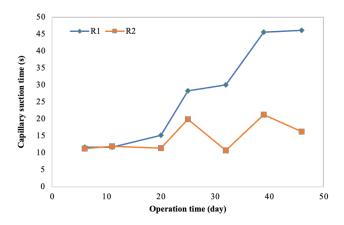
GAC addition (between 0.5 to 2g/L) into MBR prevents the sudden rise of TMP which is an indication of membrane fouling [23]. Similar to PAC, this can be attributed to GAC's ability to prevent blocking agents from accumulating on the membrane or to clear blockages on the membrane surface due to extra shear stress. It showed that the addition of GAC maintained the pore blockage at a low level and also reduced cake deposition on the membrane surface [18].

# Performances of Membrane Filtration for MBR and GAC-MBR

The variations of membrane fluxes and TMPs of conventional MBR and GAC-MBR processes are shown in Figure 6. The ceramic membrane fluxes were kept at 1.4 L/m<sup>2</sup>.h throughout the operations of both reactors, however, average membrane flux values were determined as  $1.44\pm0.31$  and  $1.38\pm0.25$  L/m<sup>2</sup>.h, for the R1 and R2 reactors, respectively. The TMP value in the R1 reactor increased from 0.330 bar to 0.640 bar in the first 15 days of operation, and a visible



**Figure 7.** Variations of TMP and membrane fluxes of each MBR throughout the operation.



**Figure 8.** Variations of TMP and membrane fluxes of each MBR throughout the operation.

cake layer was observed on the membrane surface. When the TMP value exceeded 0.6 bar, the membranes were physically backwashed with pressured air at 2 bars. Although the TMP value decreased after the applied backwashing process, it was observed that the membranes could not reach the initial state in the R1 reactor in terms of TMP value. However, only physical cleaning was carried out in the R1 reactor every other day throughout the study. In the R2 reactor, which was initially operated as MBR, the TMP value increased from 0.211 bar to 0.535 bar on the 22nd day, similar to the R1 reactor. After the addition of GAC on the 27th day of operation, there was a noticeable decrease in TMP values in the R2 reactor. As a result of GAC addition, the formation of the cake layer on the membrane surface was reduced and there was almost no need for physical backwashing. The average TMP value was determined as 0.448 bar after the addition of AC. Figure 7 shows the CST values of the sludge samples taken from the R1 and R2 reactors. It was observed that the CST values in the R1 reactor operated as a conventional MBR were higher compared to GAC-MBR. Thus, it was concluded that the addition of AC was effective in reducing the capillary absorption time. According to the results obtained, it can be seen that the MBR process operated by adding GAC has superior membrane filtration performance compared to conventional MBR.

Several studies have shown that addition of AC to MBRs can improve membrane performance in terms of both filtration resistance and sustainable operation [23, 38]. During the monitoring membrane fouling of a conventional MBR and a PAC-integrated MBR, the TMP profiles for both MBRs showed an initial slow TMP increase followed by a rapid TMP increase. However, when the two TMP profiles were compared, the initial "slow TMP rise period" for the PAC-supported MBR was found to be twice as long. This observation practically means that the addition of PAC significantly delays the need for membrane cleaning [38, 39].

The addition of AC in MBRs also improves the CST values. The improvement of CST when AC is added to an MBR tank can be attributed to both the removal of high amounts of dissolved organic carbon (DOC) and the alteration of flocs due to the incorporation of AC particles [40].

#### CONCLUSION

This study used conventional MBR (R1) and GAC-MBR (R2) technology to investigate the treatment performance of real textile wastewater and membrane characteristics. According to the data obtained, the COD removal efficiencies were determined as 87±3% and 89±6.4%, respectively, while the color removal efficiencies were 73±7% and 78±4.8%, for the R1 and R2 reactors, respectively. In addition, when extracellular polymeric substances were examined, it was observed that the addition of GAC to the R2 reactor did not have a direct effect, but the TMP pressure in the membranes, the CST of the sludge and the fouling tendency of the ceramic membrane in the reactors were affected positively. In the light of all these inferences, it is concluded that the GAC-MBR process increases both the COD and color removal efficiency and the filtration performance of the membranes compared to the classical MBR system.

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#### **AUTHORSHIP CONTRIBUTIONS**

Authors equally contributed to this work.

#### DATA AVAILABILITY STATEMENT

The authors confirm that the data that supports the findings of this study are available within the article. Raw data that support the finding of this study are available from the corresponding author, upon reasonable request.

#### **CONFLICT OF INTEREST**

The author declared no potential conflicts of interest with respect to the research, authorship, and/or publication of this article.

#### **ETHICS**

There are no ethical issues with the publication of this manuscript.

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