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Research Article

Application of an airlift internal circulation membrane bioreactor for the treatment of textile wastewater

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ABSTRACT

A large amount of water is used in the textile industry during the finishing and dyeing processes leading to the production of what is known as textile wastewaters. Textile wastewater is highly rich in COD and color and is characterized by relatively low biodegradability. This study aimed to investigate the treatability of reactive red dye-rich textile wastewater with the application of an airlift internal circulation membrane bioreactor (AIC-MBR). Experimental results demonstrated that high removal efficiencies of COD, NH_3-N, and reactive red up to 99.70%, 97.83%, and 97.23%, respectively, can be achieved using the AIC-MBR system. Besides, EPS and SMP analyses reflected an SMP polysaccharide (PS) and protein (PN) membrane rejection that reached 88% and 72.6%, respectively. Finally, the capillary suction time measurement highlighted a good dewatering capacity of the sludge with a low membrane fouling tendency at the end of the operating period.

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INTRODUCTION

Textile industry is recognized as one of the most wastewater producing manufacturing sectors. Textile wastewater is colored with a composition, which varies according to the type of fiber and chemicals used, the techniques applied, and the machines operated [1]. It is known that approximately 6900 additives and 8000 dying agents used in the sector increase the organic and inorganic pollution load of textile wastewater [2]. Therefore, it is characterized by a high total dissolved solid (TDS), COD/BOD5, salt and color. Color is usually studied as an important parameter in textile wastewater because if not removed effectively, it reduces light scarcity in the receiving water affecting in turn aquatic organisms [3]. Different types of dyes are used in the textile industry classified as reactive, dispersive, basic, acidic, azoic, direct and sulfuric dyes. Azoic, sulfuric and dispersive dyes are easily removed from effluents as they are insoluble in water. However, highly soluble dyes, namely direct, basic, acidic and reactive dyes, are hardly removed by conventional separation and treatment methods. Furthermore, the most problematic dye group used in the textile industry is reactive dyes. Because these dyes may form complexes with heavy metals such as nickel, copper and chromium, thus,

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posing a further risk when released into the environment [4]. Thus, different treatment processes including physical [5], chemical [6], electrochemical [7], and biological [8] are applied before its discharge to the environment.

Membrane bioreactors (MBRs) are proven to ensure higher effluent quality due to the complete retention of contaminants by the microbial community. However, membrane fouling is one of the limitations that affect MBR operation, increasing energy demand and reducing membrane permeability. Different reactor configurations have been developed to reduce this disadvantage of membrane bioreactors. An example of these is airlift membrane bioreactor which is characterized by a simple configuration [9]. The bioreactor is divided into a fully gassed riser and two ungassed downcomers. The density difference between them will result in the liquid circulation, which will positively affect the membrane fouling [10]. Airlift membrane bioreactors were successful in reducing the fouling processes by the rising air bubbles that generate shear stress and remove, in turn, the deposited particle along the membrane surface [11]. Besides, airlift membrane bioreactor was proved to reduce membrane fouling enhancing a better filtration performance with a lower energy consumption [12–14].

In this study, an airlift internal circulation membrane bioreactor (AIC-MBR) was tested for the treatability of Reactive Red dye-rich textile wastewater. Three different concentrations of the reactive dye were tested in the AIC-MBR. Operational parameters such as COD, NH_3-N and EPS/SMP were monitored throughout the study. Besides, the removal of reactive red dye was investigated.

MATERIALS AND METHODS

Airlift Membrane Bioreactor System and Operating Conditions

An airlift membrane bioreactor was used in this study for the treatment of synthetic textile wastewater. The reactor design is shown in Figure 1. The reactor was made of plexiglass (32 cm x 9 cm) with an effective volume of 1.5 L. Two vertical plates, having a height of 27.7 cm and placed 4.8 cm apart, were mounted inside reactor creating a riser and two downcomers. Each plate was perforated creating a hole opening of 1.8 cm. The air diffuser is located at the bottom of the reactor supplying air inside the riser. The holes on each plate ensured the circulation of air bubbles between the riser and downcomers. A hollow fiber membrane module having a pore size of 0.2 µm and an effective area of 0.0170 m^2 was used in the AIC-MBR system. The membrane was made of polyvinylidene fluoride (PVDF)-based microporous membrane containing a small quantity of polyethersulfone (PES). The system was operated continuously based on the Archimedes theory where the same volume filtered from the membrane module was fed with synthetic wastewater to the reactor.

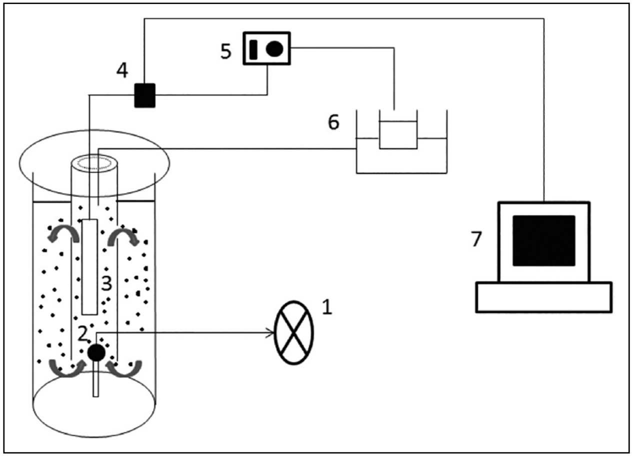

Figure 1. Schematic representation of the AIC-MBR system, air compressor (1), air diffusor (2), hollow fibre membrane module (3), manometer (4), peristaltic pump (5), feed tank (6), permeate tank, computer (7).

Table 1. Operating conditions of the AIC-MBR

Operating parameter	Value
pH	8.48
Dissolved oxygen, mg/L	8.65
Temperature, °C	18.28
SRT, days	20
HRT, hours	24
F/M (mg COD/mg MLSS.day)	0.26
Lorg, mg COD/L.day	1.095
Effective membrane area (m^2)	0.017
Net flux (LMH)	3.70

Table 2. Operational Periods in the AIC-MBR

Periods	Concentration of reactive red in feed (mg/L)	Duration (day)
1	0	1–13
2	10	14–34
3	20	35–66
4	40	67–95

AIC-MBR was operated at a steady-state condition under a sludge retention time of 20 days and an organic loading rate of 1.095 L/m^2.d. The operating conditions are summarized in Table 1. Oxygen was provided continuously through an air diffuser. The trans-membrane pressure (TMP) was measured using a pressure gauge.

The concentration of the reactive red dye was increased every 30 days as 10, 20 and 40 mg/L, in order to monitor the treatability of three different concentrations of the colorant. The operational periods are shown in Table 2.

Characterization of Textile Synthetic Wastewater and Inoculum

Real textile wastewater is characterized by a high concentration of biochemical oxygen demand (BOD) and chemical oxygen demand (COD), in addition to high values of chlorides, nitrates, suspended solids and metals [15]. However, synthetic wastewater is usually preferred because of its simplicity in evaluating results. The recipe used in this study was previously used by Yurtsever et al. (2016) as shown in Table 3 [16]. About 1000 mg/L of glucose as a source of biodegradable carbon source and different inorganics were added to meet the characteristics of real textile wastewater. Activated sludge seeded to the reactor was taken from a municipal wastewater treatment plant in İstanbul having an MLSS concentration of 5800 mg/L.

Sampling and Analyses Procedure

Chemical oxygen demand (COD), ammonium-nitrate NH_3-N, mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS) were measured three days per week based on the Standard Methods [17]. COD was analyzed in both the filtrate and the supernatant taken from the activated sludge. The dissolved oxygen (DO) and pH were monitored daily in the bioreactors using a WTW Multiline P4 multimeter (CellOx 325 DO probe and SenTix 41 pH probe). Extracellular polymeric substances (EPS) and soluble microbial products (SMP) with their protein and polysaccharide fractions were analyzed once per week from both the activated sludge and filtrate samples. EPS and SMP analyses were performed following the formaldehyde extraction method [18] where the protein (PN) and polysaccharide (PS) fractions were tested using the Lowry [19] and phenol-sulphuric acid methods [20]. The capillary suction time (CST) and sludge volume index (SVI) were analyzed every 15 days. CST was measured using a capillary suction timer (Triton type 304M) and a standard filter paper obtained from Triton. EPS, SMP and CST analysis were measured at steady-state conditions. Reactive red color treatability was monitored by analyzing its concentration in both the filtrate and the supernatant from the activated sludge. Reactive red was measured at a wavelength of 455 nm using Hach Lange DR 5000 spectrophotometer.

Table 3. Composition of the synthetic textile wastewater used in this study [16]

Added chemical	mg/L
$C_6H_{12}O_6.H_2O$	1000
$NaHCO_3$	1000
NH_4Cl	230
K_2HPO_4	37
KH_2PO_4	67
$CaCl_2.2H_2O$	4
$MgCl_2.H_2O$	3.4
$FeSO_4.7H_2O$	5.92
$MnSO_4.2H_2O$	0.4289
$ZnSO\pm.7H_2O$	0.1053
Na_2SO_3	0.2811
$NiSO_4.6H_2O$	0.1
$CoCl_2$	0.5457
Reactive red	10, 20 and 40

RESULTS AND DISCUSSION

Evaluation of AIC-MBR System Performance

Figure 2 shows the variations in the MLSS and MLVSS concentrations in addition to MLVSS/MLSS ratio. The

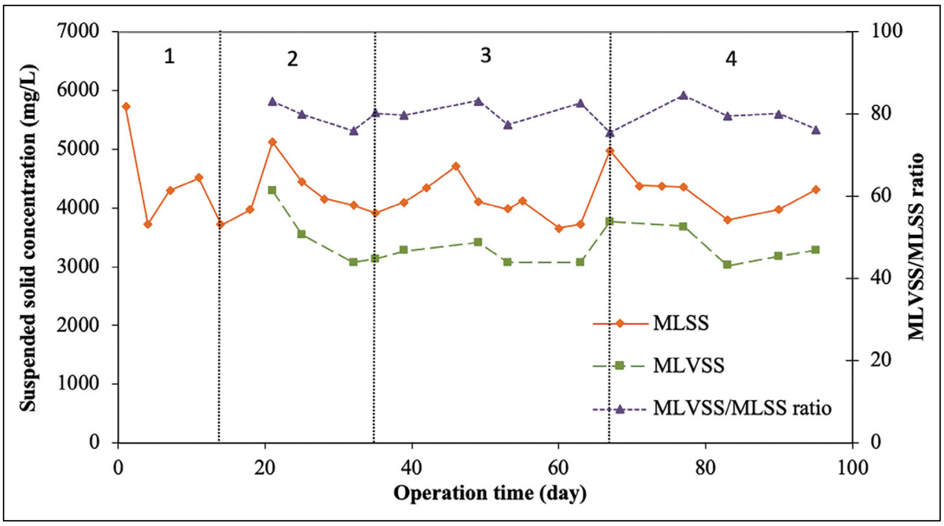

Figure 2. Variation of MLSS and MLVSS concentration and ratio.

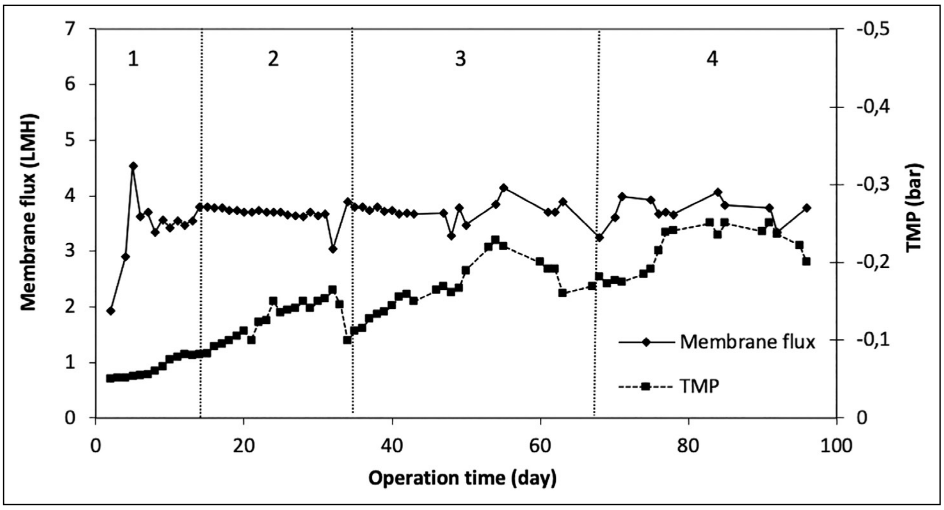

Figure 3. Variation of membrane flux and TMP values throughout AIC-MBR operation.

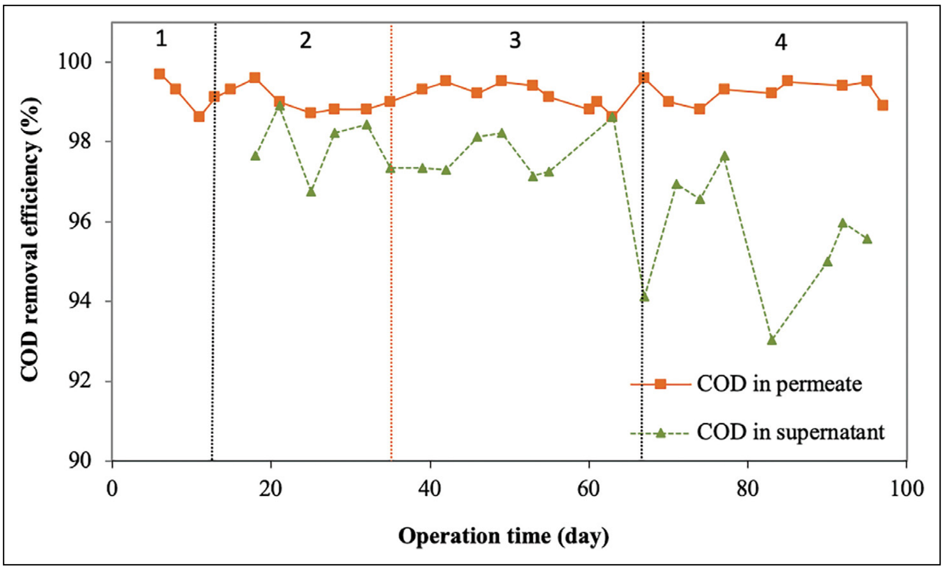

Figure 4. COD removal efficiencies in the supernatant and permeate over time.

initial MLSS concentration was 5800 mg/L which fluctuated until it reached its steady-state conditions after about 20 days. MLSS was then kept stable between 4000 to 5000 mg/L for the remaining 80 days of operation. Similarly, the MLVSS and MLVSS/MLSS ratio changed in the range of 4000 to 3000 mg/L and 75.5 to 85.5%, respectively. This high MLVSS/MLSS ratio reflected the microbial activity in the reactor. An SRT value of 20 days was kept by drawing a daily sludge volume of 75 mL from the AIC-MBR. The SRT in aerobic MBRs is usually preferred to be between 20 and 50 days, which confirms the suitability of the operating SRT in this study [21].

The average membrane flux was about 3.70 L/m^2.h (LMH) where the transmembrane pressure (TMP) decreased from zero to -0.2 bar at the end of the operating period (Fig. 3). Membrane flux and TMP were directly affected by the aeration rate influencing in turn membrane fouling. In this study, no back-washing or chemical washing was applied to the membrane. Therefore, the minimal reduction occurred in TMP is acceptable.

COD Removal Efficiency by AIC-MBR

COD removal efficiency was measured in both the permeate and the supernatant during the operating period of the reactor. As can be seen from Figure 4, the COD removal efficiency in the supernatant fluctuates in the range of 93.02–98.91% while in the permeate it registered a removal efficiency of about 98.62 to 99.70%. Thus, COD removal efficiency was higher in the permeate then in the supernatant noting that both demonstrated great removal efficiencies. This is in correspondence with Lee

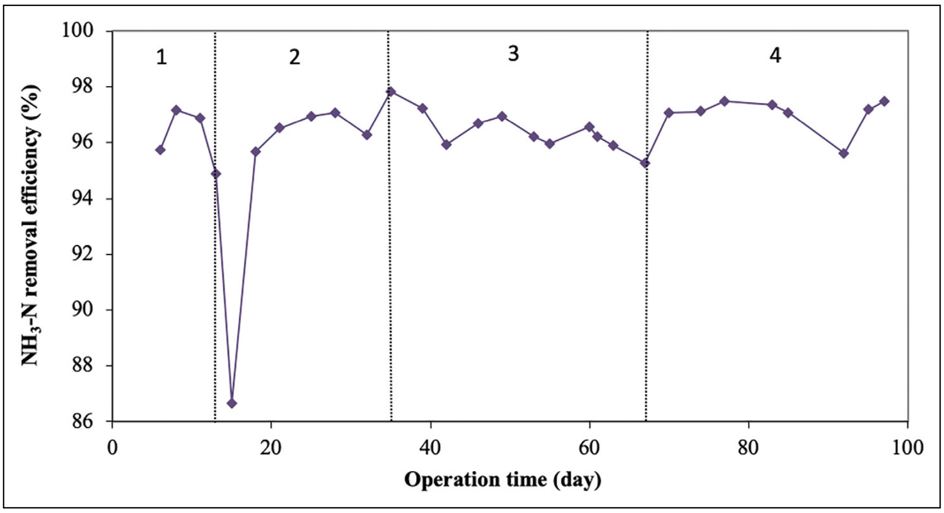

Figure 5. NH_3-N removal efficiencies over time.

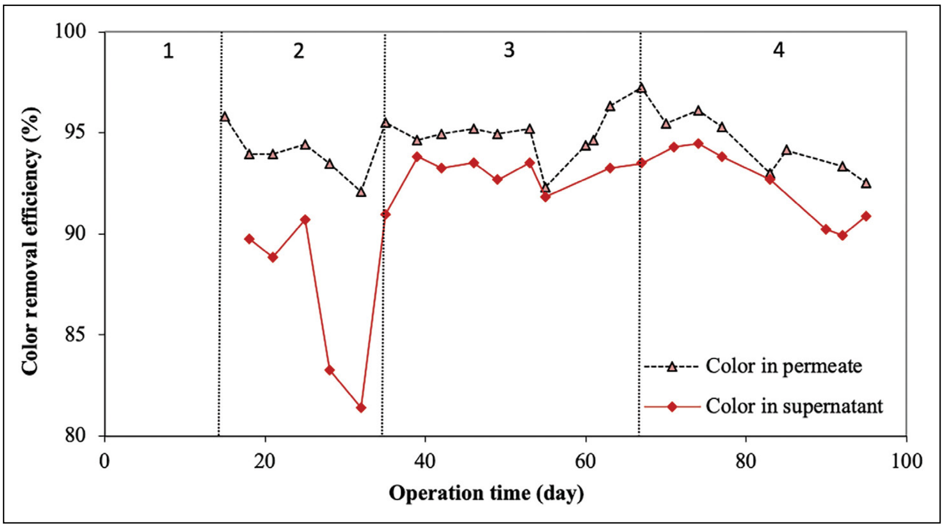

Figure 6. Color removal efficiencies over time.

et al. (2003) who found out that the COD removal in the permeate was higher than the supernatant [22]. Besides, Yurtsever et al. (2016) reported in their study that COD concentrations of 54±14 mg/L and 47±12 mg/L were recorded for the supernatant and permeate, respectively [16]. Berube et al., (2010) reported that the COD removal efficiency in a conventional activated sludge process may typically reach about 95%, however, this value may increase to reach a range of 96 to 99% in a membrane bioreactor process [23]. This can be explained by the fact the colloids and soluble compounds can be attached to the suspended solids which can be retained in MBR system giving a better COD removal and particle-free effluent [24, 25]. The average removal efficiency of 90% was obtained in another study that used airlift external circulation membrane bioreactor (AEC-MBR) for the treatment of toilet wastewater [26].

NH_3-N Removal Efficiency by AIC-MBR

The laboratory prepared synthetic textile wastewater had a NH_4^+ concentration of 77.4 mg/L. The removal efficiency of NH_3-N in the permeate from an AIC-MBR was studied and the experimental results are presented in Figure 5. NH_3-N removal efficiency changed between 95.73 and 97.83% during the operating period. However, an important decrease in NH_3-N removal was observed after the addition of reactive red color to the reactor which affected the performance of the microorganisms. The adaptation time of microorganisms to any new component entering the reactor medium may differ in COD values, which may explain the reason behind the sudden decrease observed on the day of dye addition [27]. The removal efficiency of 96.6% was obtained using a draft tube MBR without carriers for the treatment of mixed wastewater [28]. The COD/TN ratio with the dissolved

Table 4. Typical characteristics of textile effluents

	Ghaly et al. (2014) [32]	Kehinde and Aziz (2014) [33]	Tavangar et al. (2019) [34]	Bhuvaneswari et al. (2016) [35]	Yurtsever et al. (2020) [36]
pH	6–10	6.95–11.8	7.03±0.01	8.6–9.2	8–9.5
Color (Pt-Co)	50–2500	50–2500	2100±5	N/A	500–1250
COD (mg/L)	150–12000	150–30000	2690±10	3880–4400	700–1250
TSS (mg/L)	15–8000	15–8000	280±2	550–650	200–450

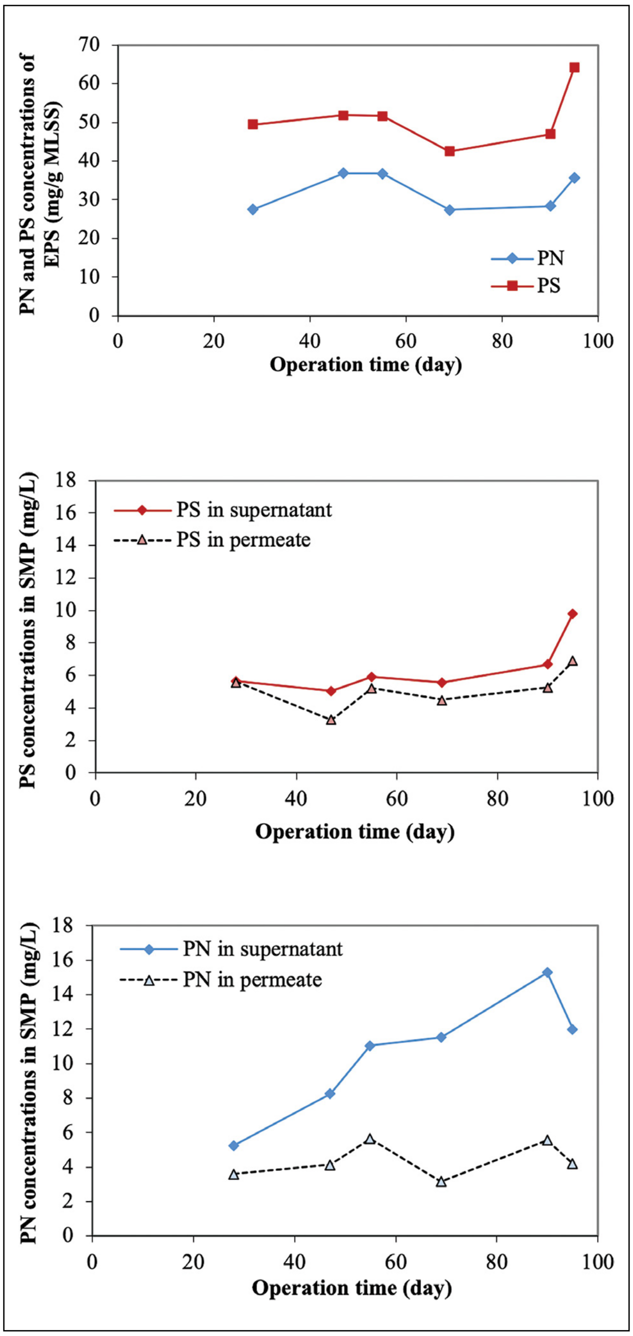

Figure 7. Variance in the protein and polysaccharide concentrations of EPS **(a)**, polysaccharide **(b)** and protein **(c)** concentrations in SMP in both supernatant and permeate.

oxygen level was found to affect the simultaneous nitrification and denitrification in an airlift membrane bioreactor. A COD/TN level in the range of 4.77 to 10.04 leads to a nitrogen removal that exceeds 70% which is comparable to the results in this study taking into consideration that COD/TN is about 12.92 [29].

Reactive Red Removal Efficiency by AIC-MBR

Reactive red was used in this study as the type of colorant to be removed. As mentioned earlier, three different concentrations of reactive red were added subsequently to the reactor. The color removal efficiency was monitored in both permeate and supernatant and the obtained results are shown in Figure 6. The removal efficiency in the supernatant varied between 83.25 and 94.47% where it increased to 89.29 and 97.23% in the permeate. A slight decrease occurred after the addition of the third concentration of reactive red of 40 mg/L. Color removal was performed using different methods in the literature. The removal efficiency varied according to the initial concentration and the treatment process applied [30]. In an anaerobic/aerobic sequential batch reactor system, 20 mg/L of reactive black 5, reactive blue 19 and reactive blue 5 was treated and the removal efficiencies were found to be 63, 64 and 66%, respectively [31]. Color may be removed through different processes through cleavage of the chemical bonds or adsorption to the microbial flocs. The removal occurred in supernatant reflects the role of the microbial activity in reducing or adsorbing the dye molecules [30]. Besides, the higher removal efficiencies obtained in the permeate is in close relationship with the membrane used which blocks the passage of any particle, as mentioned earlier.

The composition of textile industry wastewater varies from factory to factory and from country to country, depending on the process, the equipment used, the type of fabric produced and the chemicals applied [30]. Table 4 shows the real textile wastewater characteristics reported from different sources and countries. As seen from Table 4, the pH value of real textile wastewater varies in a wide range between 5.5–11.8. This wide pH or COD variations may lead to negative effects in MBR operation, especially on active biomass, making it impossible to achieve a stable biological treatment in terms of pollutant removal and membrane fouling.

Variation in EPS and SMP Components

Extracellular polymeric substances (EPS) are divided into bound EPS and soluble EPS [mainly named soluble microbial products (SMP)]. Major constituents of EPS consist mainly of proteins and polysaccharides [37]. The results regarding the variation in EPS and SMP concentrations are presented in Figures 7, respectively. An overall evaluation of both plots in Figure 7a shows a general increasing trend in EPS and SMP level with time noting that EPS values were higher than SMP ones. The increase in these values may be attributed to the increase in MLSS concentration in the reactor. Besides, the increasing trend may be related to the increase in the TMP levels discussed above and thus to membrane biofouling [38]. It was stated by researchers that EPS concentration and characteristics are directly affected by multiple parameters such as the sludge composition, sludge retention time (SRT) and aeration [39, 40]. The comparison between the polysaccharide and protein fractions of EPS and SMP reflected a superiority of polysaccharide levels in both EPS and SMP. Polysaccharide fractions usually affect fouling in membrane bioreactors, as reported by Yigit et al. [41]. SMP polysaccharide (PS) and protein fractions (PN) were measured in both the supernatant and permeate. The corresponding results are presented in Figure 7b and 7c. It can be seen clearly than PS and PN of SMP shows higher and increasing values in the supernatant than in the permeate. The PS rejection of SMP by the hollow membrane was found to be 0.88% at 28th day, while the PS rejections by the membrane increased in the next operation period and were determined between 64 and 88%. On the other hand, PN rejection of SMP was found 31.4% at 28th day, while after this point, the rejection rates were found to vary between 49.7 and 72.6%. The obtained results can be explained by the membrane filterability which blocks the soluble part of EPS that may be adhered to microbial flocs or other particles in the sludge [38]. However, the minimal part of the SMP appearing in the permeate is caused by the membrane permeability that may enhance the passage of the soluble and non-adhesive part of the SMP [42].

Variation in CST Levels

The capillary suction time (CST) of the sludge was measured in order to test the dewatering properties of the activated sludge. As can be seen from Figure 8, CST values showed fluctuations between 5.5 and 9.6 s with a minimal increase at the end of the operating period. Additionally, an increase observed in the CST may be related to the increase occurred in the polysaccharide values of EPS. However, CST values were still in an acceptable range reflecting a good dewatering capacity of the sludge [43].

On the 69th day of operation, CST value decreased from 6.9 s to 5.5 s and then increased to 9.6 s on the 90th day. These results could be related to the concentrations of PN and PS of SMP, as the CST values presented a similar trend with SMP concentrations of supernatant. It was reported by Zhang et al. (2015) [44] that there is a significant relationship between SMP and membrane fouling and SMPs are the main soluble components in a gel layer and cake layer on the membrane surface and pores. Higher specific filtration resistance occurred in the gel layer due to the adsorption of SMP [45].

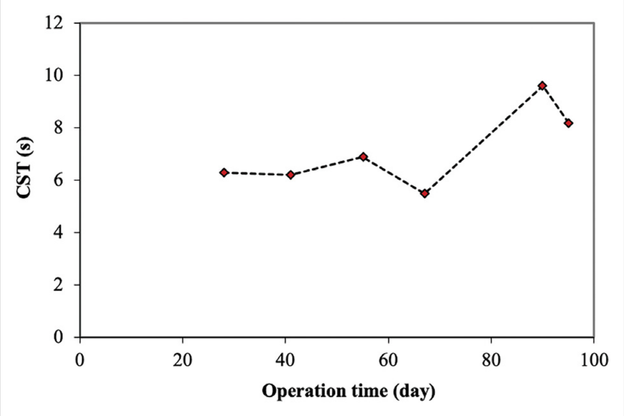

Figure 8. CST values obtained from activated sludge samples over time.

CONCLUSION

The application of an AIC-MBR demonstrated quite high COD, NH_3-N and reactive red removal efficiencies up to 99.70%, 97.83%, and 97.23%, respectively. Moreover, the proposed system ensures satisfactory results even with increased initial concentrations of the reactive dye. Additionally, a relatively low membrane fouling tendency was observed in this technology based on the results from EPS, SMP and CST. The overall results indicated that the AIC-MBR can be applied for the treatment of real textile wastewater, after necessary preliminary tests were carried out.

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 authors 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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