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# Performance evaluation of submerged membrane bioreactor for model textile wastewater treatment

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**Abstract.** Submerged Membrane bioreactor (SMBR) is one of the last techniques that allow a high quality of treated industrial effluents by coupling biological treatment and membrane separation. Thus, this research was an effort to evaluate performance of a SMBR treating a model textile wastewater (MTWW). Different SMBR operating parameters like mixed liquor suspended solids (MLSS) and Dissolved oxygen concentration, hydraulic retention time (HRT), and nutrients addition (N and P) have been investigated. MTWW (influent to the SMBR) was generated using the reactive azo-dye, Novacron blue FNG (100mg/L feed concentration). Results of MTWW treatment using SMBR under optimal operating conditions (MLSS, 4.2-13.3g/L; HRT, 4 days; pH, 6.9-7.2; conductivity, 400-900 µS/cm and temperature, 19.4-22.2 °C) showed that COD and blue colour treatment performances are between 94-98% and 30-80%, respectively. It is concluded that SMBR can be used in large scale textile wastewater treatment plants to improve effluent quality in order to meet effluent discharge standards.

**Keywords:** **d**ye removal; model textile dye wastewater; mixed liquor; submerged membrane bioreactor; wastewater treatment

## 1. Introduction

Huge quantities of chemicals (dyes, auxiliaries) are consumed during textile dyeing process, which strongly impact wastewater flow and composition. Textile wastewaters (TWW) are usually characterized by high pH value, and a low BOD/COD (biological oxygen demand/chemical oxygen demand) ratio. They contain large amounts of dissolved organic matter and inorganic substances. The residues of non-biodegradable dyes are kept mostly in the TWW, being the result of incomplete binding of dyes to textile fibres. The average rate of dye-fixating when dyed with reactive dyes is from 60 to 80%. The residues of non-fixed colours are washed from the textile and thus contaminating the wastewater. TWW are the major source of water pollution and represent a great threat to the environment and human health (Abiri *et al*. 2016).

Removal of dyes from wastewater has become a major concern in TWW treatment and has to be treated before returning to the natural environment (Konsowa *et al*. 2012; Yurtsever *et al*. 2015). Many studies have focused on physico/chemical and biological treatments of TWW. Therefore, it’s important to highlight the development of more efficient technologies for TWW treatment and reuse by for example, coupling advanced processes such as membrane bioreactor technology (Hoinkis *et al.* 2012). In a typical configuration, a membrane bioreactor (MBR) is composed of two unitary operation, the biological unit responsible for the soluble wastewater pollutant biodegradation and the membrane module for the filtration process of the treated water from mixed liquor (Couto *et al*. 2017). There are several advantages affiliated with the MBR, which makes it an important alternative over other industrial wastewater treatment techniques (Friha *et al*. 2015). MBR associates the benefits of high biomass levels with the possibility to run continued processes at controlled biomass retention. Since the membrane cost has decreased seriously over these last years and nowadays energy requirements the membrane aeration are also fast approaching the normal range of the common activated sludge process, this has now become an economically feasible solution even for a low-profit process such as TWW treatment (Jegatheesan *et al*. 2016). MBR technology will be a crucial part of advancing water sustainability because they stimulate opportunities for decentralized treatment and incite the reuse of biologically treated textile wastewater (Hoinkis *et al.* 2012, Kaykioglu *et al.* 2017, Al Sawaf and karaca 2018).

Presently, a wide range of MBR systems have been utilized including side-stream and submerged configurations. In side-stream or cross-flow systems, the use of recirculation loops leads to increased energy costs.

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Also, huge shear stresses and recirculation pumps for these first MBR generations have contributed to the destruction of bioflocs and this has been associated to a loss of biological activity Yamamoto (1989). To overcome these limits, the submerged MBR have developed an experimented growth in municipal and domestic wastewater treatment due to several advantages comprising low sludge production and excellent effluent quality (Fazal *et al*. 2015). The performance of submerged MBR has also gained more attention for industrial wastewater treatment owing to their cost-effective and high organic loadings. In this context, it is interesting to note that submerged MBR have been used in various industries including paper, chemical production, textile and pharmaceuticals. Recently, Rezakazemi *et al*. (2017) investigated the treatment of high loaded synthetic hazardous wastewater using micro filter ceramic submerged membrane bioreactor. The results showed a desired performance for the wastewater treatment with COD removals up to 90% after one week. Moreover, a study by Rezakazemi et *al*. (2018) evaluated an aerobic submerged hollow fiber made of polyvinylidene fluoride (PVDF) with 0.5 µm membrane pore size for treating wastewater containing phenol and ammonium. The results showed a performance of the membrane with phenol and COD removals of 99 and 95% respectively. In fact, the efficiency of PVDF hollow fiber as well as flat sheet membrane have been proved for wastewater treatment in several studies (Aryal *et al*. 2009, Johir *et al*. 2011; Rezakazemi *et al*. 2018). Thereby, it is important to evaluate the capacity of a microfiltration flat sheet membrane made of (PVDF) on textile wastewater treatment due to its higher water permeation rate and its tolerance to acid and base chemicals used for membrane cleaning (Johir *et al*. 2011).

On this basis, this study aims to evaluate the applicability of a pilot-scale aerobic MBR system for the biological treatment of textile dye wastewater. The aerobic system comprised a submerged microfiltration module based on flat sheet membrane made of PVDF, in order to investigate the efficiency of this membrane module in reducing COD, colour and thus to improve the effluents quality for water reuse. The azo-dye reactive blue novacron FNG was used as a model textile dye wastewater (MTWW), provided from a Tunisian Textile industry widely used to colour cotton and cellulose fibers.

## 2. Material and methods

### *2.1 MTWW characteristics*

Since the quality of TWW changes due to the use of several colouring matters, dyestuffs, accompanying chemicals and processes, the model textile dye wastewater (MTWW) was developed based on literature studies to design a MTWW benchmark (Deowan *et al*. 2013). The MTWW was also based on a Blue reactive dye which is widely applied in Tunisian textile industry. Glucose, Ammonium chloride and Potassium dihydrogen phosphate were added as carbone, nitrogen and phosphore sources in order to keep the ratio C:N:P equal to 100:5:1, a common ratio used in aerobic activated sludge for a better biomass development (Khouni *et al*. 2012). The initial pH was adjusted to 7.2 with 0.1 M HCl or 0.1 M NaOH addition. Table 1 summarized the characteristics and composition of the MTWW used in this study.

Table 1 Composition and characteristics of Model Textile Wastewater

| Parameters | Measured values |
|---|---|
| Reactive Blue (mg/L) | 100 |
| Glucose (g/L) | 1.00 |
| Ammonium chloride (g/L) | 0.19 |
| Monopotassium phosphate (g/L) | 0.04 |
| COD (mg/L) | 1290 |
| $NH_4$-N (g/L) | 0.05 |
| Conductivity (µS/cm) | 50.30 |
| pH | 7.20 |

Table 2 Characteristics of the mixed liquor

| Parameters | Unit | Measured values |
|---|---|---|
| MLSS | g/L | 13.30 ± 1.30 |
| MLVSS | g/L | 2.95 ± 0.22 |
| Dissolved COD | mg/L | 27.20 ± 1.36 |
| $NH_4$-N | mg/L | 0.19 ± 0.02 |
| $NO_3$-N | mg/L | 0.37 ± 0.02 |
| $NO_2$-N | mg/L | 0.10 ± 0.01 |

### *2.2 Mixed liquor characteristics*

Mixed liquor was obtained from municipal wastewater treatment plant, Grande Motte, Hérault, Montpellier. This wastewater treatment plant is dealing with a daily pollution load of 65000 population equivalents. Other characteristics of the mixed liquor were shown in Table 2.

### *2.3 Membrane bioreactor unit*

The diagram of SMBR unit operating at the laboratory scale was presented in Fig. 1

The study was conducted using a laboratory scale SMBR (Fig. 1). The reactor had a total volume of 10L, a working volume of 6L and maintained for a period of 47 days at room temperature 21±1°C. At the beginning, the SMBR was seeded with 4L of mixed liquor obtained from municipal wastewater treatment plant and continuously fed with a MTWW pumped with a diaphragm metering pump (ProMinent® gamma G/4b) at a flow rate of 1.5 L/day (Table 3). A flat sheet membrane module made of PVDF obtained from the A3 company was submerged in the membrane tank. The pore size of the PVDF membrane was 0.14 µm, the pore size typically represents microfiltration, and can retain almost of the biomass including the isolated microorganisms. The membrane module contained 8 flat

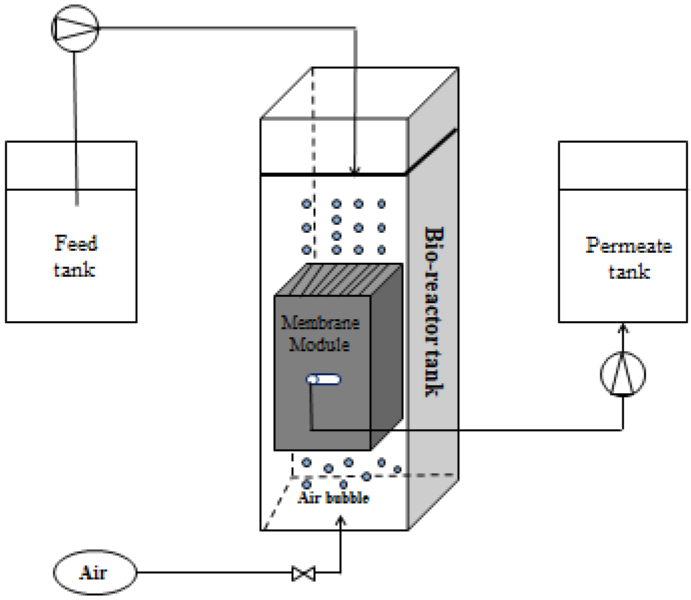


Fig. 1 Schematic representation of laboratory Submerged Membrane Bioreactor (SMBR) Influent pump, Air flow meter, Effluent (permeate) pump

Table 3 Operating conditions of membrane bioreactor

| Operating conditions | |
|---|---|
| Working volume (L) | 6 |
| Hydraulic flow rate (L/d) | 1.5 |
| Volumetric organic load (g/L.d) | 0.25 |
| Hydraulic retention time (d) | 4 |
| Temperature (°C) | 22 |
| pH | 7 |
| Dissolved oxygen | 6.78 |

sheets at an interval of 12 mm; the sheets were connected together and acted as a simple membrane module with an effective membrane area of 0.2 $m^2$. Air flow rate in the range of 0.5-1.5 $m^3$/ ($m^2$ membrane area h) was utilized to create a shear stress on the membrane surface. This range of aeration flux rate was used to minimize sludge accumulation and to reduce fouling. A study by Meng et *al.* (2008) confirmed that in submerged MBR systems, the shear stress is created by aeration which not only provides oxygen to the biomass, but also maintains the solids in suspension and scours the membrane surface to alleviate membrane fouling. The membrane filtration was from outside to inside and permeate was pumped out using a peristaltic pump (Watson Marlow/505S) at a flow rate of 1.5 L/day. Mixed liquor suspended solids in the bioreactor were maintained to a value between 4.2-13.3g/L and the hydraulic retention time (HRT) was 4 days. Table 3 summarized the operating conditions of membrane bioreactor.

### *2.4 Analytical methods*

Samples were collected from the bioreactor twice a week and filtered by glass fiber filters in order to analyze soluble compounds. Permeate rate (flux), sludge production, conductivity, turbidity, dissolved oxygen, pollutants removal efficiency and effluent quality were measured with respects to operation time and operating conditions. Samples of influent and effluent were analyzed based on colorimetric tests (Hach Lange, Germany) for chemical oxygen demand (COD; Micro-Method “LCK414” 5-60 mg/l), ammonium (NH4-N; Micro-Method “LCK303” 1-12mg/l), nitrate (NO3-N; Micro-Method “LCK340” 5-35 mg/l), and nitrite (NO2-N; Micro-Method “LCK342” 0,6-6 mg/l). Mixed Liquor Suspended Solids (MLSS) was determined by drying samples at 105°C for 24h and the Mixed Liquor Volatile Suspended Solids (MLVSS) was measured after two hours of furnace operations at 550°C. The laboratory analyses were determined according to the standard methods for the examination of water and wastewater (APHA, AWWA, WEF 2012). Dissolved oxygen and pH were measured using an oxygen probe Oxi 340WTW and pH meter pHenomenal 111 (662-1157) VWR pH1000H respectively.

Colour was measured by spectrophotometric absorbance at 609 nm where the dye presents its pick of absorbance using UV/VIS spectrometer (Shimadzu, Japan).

Hach’s definition of sensitivity for the DR/2500 is the change in concentration, (ΔConcentration) for a 0.010 change in absorbance (ΔAbs). Thus the guaranteed measurement error is 20 $mg_{COD}$/L for COD high range, 3 $mg_{COD}$/L for LCK 414 and 0.2 $mg_{COD}$/L for LCK 340.

### *2.5 Statistical analysis*

The data presented are the average of the results of two replicates with a standard error of less than 5%.

## 3. Results and discussion

The SMBR technology has been proven to be a single step process in efficient treatment of industrial and

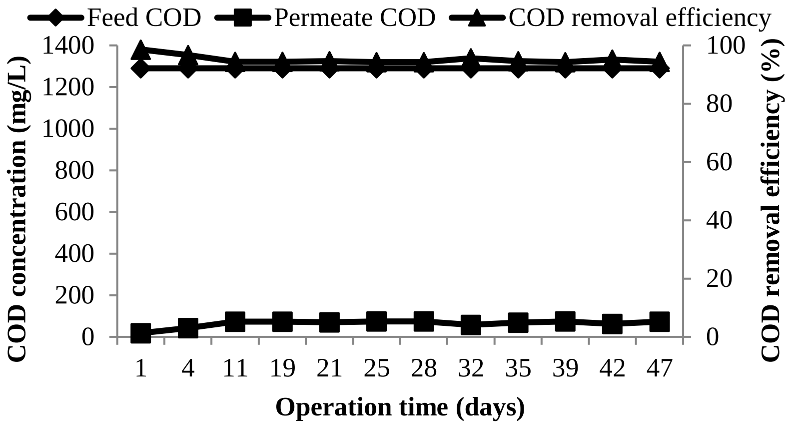


Fig. 2 COD removal efficiency during SMBR experiment

municipal wastewaters (Naghizadeh *et al*. 2008). In this study, a laboratory scale experiment was studied to treat a MTWW. The aerobic reactor with a submerged membrane used in this work was regularly aerated for organic matter oxidation, nitrification and phosphorous uptake as well as for fouling control. The SMBR experiments were started only after having the microorganisms acclimated with the feed conditions. Subsequently, tests were carried out continuously investigating a variety of parameters mainly COD, colour, pH, MLSS, MLVSS, conductivity, $O_2$ consumption and N-compounds.

### *3.1 COD and colour removal performance of the SMBR system*

COD is considered as one of the main parameters to define the performance of a SMBR system in terms of organic pollutant removal (Deowan *et al*. 2013). It is used to indirectly measure the amount of organic compounds in the MTWW and permeate. Several studies investigated the efficiency of the MBR module on COD and color removal. Spagni et *al*. (2010) confirmed the high stability of membrane filtration and biological processes combination on COD removal for a model textile wastewater containing an azo-dye reactive orange 16 in an anaerobic-biofilm anoxic aerobic bioreactor. Also, a study by Spagni et *al*. (2012) revealed the importance of a submerged MBR as a suitable system for the decolourization of textile wastewater containing azo-dye orange 16 under anaerobic conditions with values up to 99.2% of color removal. Moreover, Grilli et *al*. (2015) demonstrated the performance of the MBR as an effective system for the treatment of dyeing wastewater on COD (50-80%) and color (70-90%) removals. Recently, Li *et al*. (2018) proved the feasibility and the performance of utilizing the membrane module for the treatment of industrial dyeing and their characteristic pollutants. The relevant COD and colour removals efficiency was 90% and 94% respectively over 48h continuous operation conditions. Fig. 2 showed the evolution of COD in permMeate during the MTWW treatment by SMBR technology. The COD removal efficiency of the SMBR system is steady around 94±4% for 1290mg/L inlet COD fed to the membrane bioreactor. Similar results were reported by Saha *et al*. (2014), revealing 90% of COD removal efficiency in a bench scale membrane bioreactor (MBR) unit and Luong *et al*. (2016), revealing up to 95% of COD removal efficiency in a pilot-scale membrane bioreactor for the treatment of a MTWW. Such findings underlined the performance of membrane bioreactor system for sustainable development contribution in the textile industry to improve water quality of treated dyeing wastewater (Friha *et al*. 2015, Luong *et al*. 2016).

The colour from the TWW is mainly due to the intense dyeing process, which involves a wide range of different types of recalcitrant dyes. The colour removal efficiency from MTWW is one of the most critical aspects (Deowan *et al.* 2013). Regularly during the SMBR process, samples from the MTWW and permeate were taken for UV-Vis measurements in order to more clearly illustrate the changes in absorbance during the MBR process. According to Fig.3, as well as visible observation, a clear reduction in colour was obtained from MBR treatment. The highest blue colour removal efficiency was observed at the beginning of the wastewater treatment by SMBR technology which was more than 80%. The colour removal yields registered during the MBR under an HRT of 4 days substantiated the significant contribution of the membrane to the overall dye removal (biosorption, cake layer filtration, biodegradation). Since the colour causing dye has smaller molecular weight than the pore size of the applied membrane, the dye is supposed to be passed through the submerged bioreactor membrane. Similar behaviors were also reported by Deowan *et al*. (2012 and 2013) and (Bouhadjar *et al*. 2016).

### *3.2 Nutrient (nitrogen and phosphorous) removals*

To obtain the nitrogen (N) balance, the contents of total nitrogen, ammonium, nitrate and nitrite in MTWW and permeate were measured as described in Fig. 4. The main sources of nitrogen (N) were mainly $NH_4Cl$ as well as to smaller extend the blue dye used in MTWW.

From the Fig. 4, it is observed that the removal efficiency of nitrogen content is fairly variable, since quite significant changes in the content of ammonium and nitrate

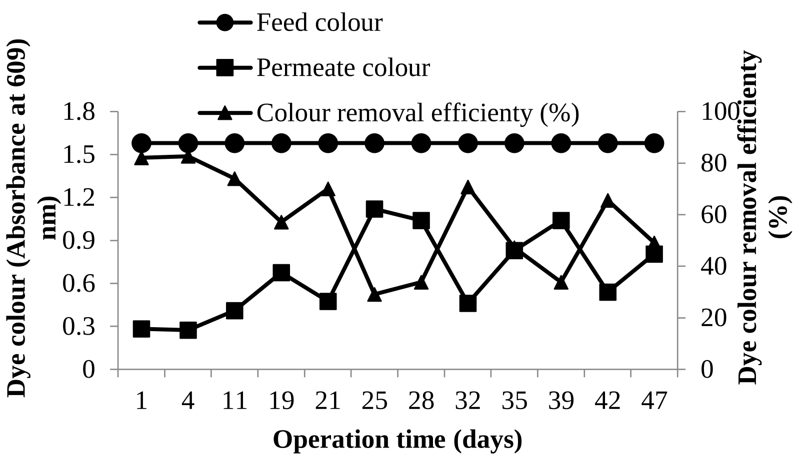


Fig. 3 Colour removal efficiency during SMBR experiment

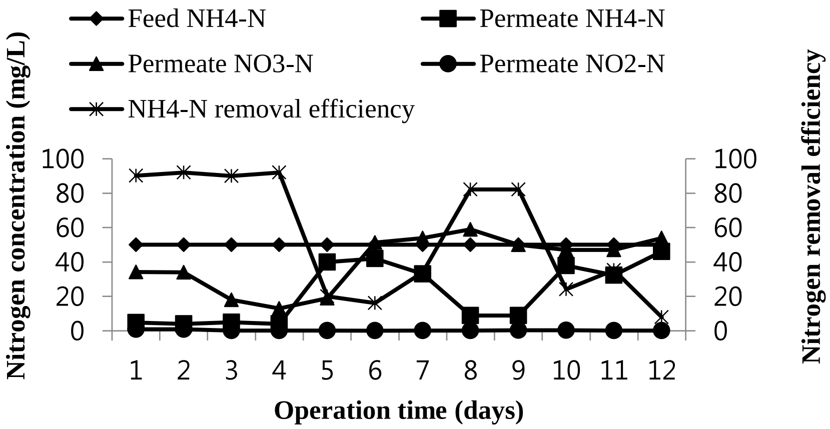


Fig. 4 Nitrogen removal efficiency during SMBR experiment

of the MBR process are shown. Generally, the contents of ammonium, nitrate and nitrite are at reasonable low values, indicating efficient nitrification and denitrification. The ammonia removal efficiency was higher than 50% because in aerobic condition, the ammonia was converted during nitrification cycle. The results were in line to Blstakova *et al*. (2012) and Rondon *et al*. (2015).

### *3.3 Effect of some operating conditions on Submerged MBR performance*

The MLSS concentration varied from 4.2 to 13.3 g/L over the operation period of 45 days. The MLSS values indicated that bacterial population has been adapted to the system environment. During the period of operation, no excess sludge was wasted from the system except for sludge withdrawn for analytical purposes. MLVSS to MLSS ratio was currently used as an indicator of the amount of organic sludge, and it was found to be relatively constant (data not shown). Several experimental studies have also reported a constant performance of MBR, with an effective balance of active biomass and inorganic fraction during long-term operating periods (Pollice *et al*. 2005). Pollice *et al*. (2005) explained this phenomenon by a possible hydrolysis and/or enzymatic solubilization of inert matter. As well, Abiri *et al*. (2016) have shown a direct correlation between the initial MLSS concentration of the dyeing factory wastewater and COD and colour removal.

In general, the performance of SMBR used in this study for the treatment of MTWW was evaluated and compared to other research works (Table 4). COD and colour removal efficiencies showed similar trends than those obtained by Deowan *et al*. (2013) and Bouhadjar *et al*. (2016). Deowan *et al*. (2013) reported the application of a submerged membrane bioreactor (SMBR) with commercial membrane module and novel MBR module for the treatment of model textile dye wastewater (MTWW). under the operating conditions of 12 g/L of MLSS, 40-80 h of HRT, 1.0 m3/h of air supply to MBR reactor, pH of 8.2±0.2- 10.5±0.2 and temperature of 18±2 °C, the COD removal efficiency was around 90% for 2450 mg/L inlet COD fed to the membrane bioreactor and Red and Blue colour removal efficiencies were 25-70% and 20-50% respectively. Bouhadjar *et al*. (2016) studied the performance of commercial membranes in a side stream and submerged membrane bioreactor for model textile wastewater treatment. The COD removal efficiency was varied between 90 and 97%, respectively,

Table 4 MTWW treatment performance in MBR processes and comparison with previous studies

| Type of wastewater | Application level | Operation conditions | COD removal (%) | Colour removal (%) | Membrane characteristics | References |
|---|---|---|---|---|---|---|
| MTWW<br>Remazol Brillant Blue R (50 mg/L)<br><br>Acid Red 4 (50 mg/L) | Pilot scale SMBR | HRT:40-80hours<br>MLSS: 12g | 90% | 20-50%<br><br>25-70% | Commercial membrane module<br>Flat sheet<br>Pore size : 0.050 µm | Deowan *et al*. (2013) |
| MTWW<br>Remazol Brillant Blue R (50mg/L)<br><br>Acid Red 4 (50mg/L) | Pilot scale SMBR | HRT: 40-90<br>MLSS: 8-12 | 90-97% | 50-90%<br><br>20-40% | flat sheet membrane module<br>UF and NF<br>Pore size UF: 0.04 µm | Bouhadjar *et al*. (2016) |
| Indigo blue dye | Pilot scale SMBR | HRT: 8 hours | 73% | 100% | hollow fiber polyetherimide-based membrane module | Couto *et al*. (2017) |
| MTWW<br>Blue novacron FNG (100mg/L) | Pilot scale SMBR | HRT 4days<br>4.2-13.3 MLSS | 94-98% | 30-80% | Submerged MBR<br>Flat sheet (PVDF)<br>Pore size0.14µm | This work |

and colour rejection was found in the range of 20–40% for red dye and 50–90% for blue dye in both units. Couto *et al*. (2017) have also studied the performance of microfiltration membrane in SMBR for the treatment of indigo blue dye, indicating a COD reduction by about 65% and 100% of colour removal. In order to improve the wastewater quality, a nanofiltration membrane (NF) was tested in the SSMBR unit and still has to be tested in the SMBR.

## 4. Conclusion

In this study, a MBR bioreactor combined to a submerged flat sheet membrane module made of PVDF was operated for more than 45 days to treat a MTWW provided from a Tunisian Textile Industry. The SMBR was operated to investigate the performance of a PVDF flat sheet membrane for COD and color removal under the operating conditions of 4.2-13.3 g/L MLSS, 4 days HRT, pH 6.9-7.2, conductivity 400-900 µS/cm and temperature 19-22°C. The membrane module was found to be effective for COD removal efficiency 94% for 1290 mg/L inlet COD fed to the membrane bioreactor and Blue colour removal efficiencies varied between 30 and 80%. The developed laboratory scale SMBR utilizing an available membrane module significantly improved the MTWW quality. On the basis of the results obtained in this work, it can be concluded that the PVDF flat sheet submerged MBR could be considered as useful system in the large scale textile industrial wastewater treatment plants to improve effluents quality while following discharge standards. However, in order to upgrade SMBR effluent for water reuse, ultra- or nano-filtration as alternatives treatments may be needed.

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## Conflicts of interest

Authors disclose that there are no conflicts of interest.

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