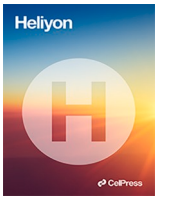
Heliyon 10 (2024) e40742

Contents lists available at ScienceDirect

Heliyon

journal homepage: www.cell.com/heliyon



Research article

Treatment of industrial textile wastewater by means of forward osmosis aiming to recover dyes and clean water

Carmen M. Sánchez-Arévalo [a,*], Laura García-Suarez [b], Maria Salud Camilleri-Rumbau [c], Jorg Vogel [d], Silvia Álvarez-Blanco [a,e], Beatriz Cuartas-Uribe [a,e], M. Cinta Vincent-Vela [a,e]

[a] Research Institute for Industrial, Radiophysical and Environmental Safety (ISIRYM), Universitat Politècnica de València, Camino de Vera, s/n, 46022, Valencia, Spain
[b] Jeanologia S.L., Ronda de Guglielmo Marconi, 12, 46980, Paterna, Valencia, Spain
[c] Eurecat, Centre Tecnològic de Catalunya, Water, Air and Soil Unit, P. de la Ciència, 2, 08243, Manresa, Spain
[d] Aquaporin, Nymøllevej 78, 2800 Kongens Lyngby, Denmark
[e] Department of Chemical and Nuclear Engineering, Universitat Politècnica de València, Camino de Vera s/n, 46022, Valencia, Spain

ARTICLE INFO

Keywords:
Forward osmosis
Textile wastewater
Draw solution
Dye concentration
Water recovery
Wastewater treatment

ABSTRACT

The textile industry is one of the largest water consumers, and, as a result of its activity, it generates tons of wastewater. In this research, forward osmosis has been employed to tackle the critical need of treating textile wastewater. The HFFO2 membrane (Aquaporin) was used to process large volumes of real cotton dyeing wastewater, wool dyeing wastewater, and several types of textile end-of-pipe wastewater. In all cases, the permeate flux was between 6 and 8 $L \cdot h^{-1} m^{-2}$ during the major part of the process. The recovery of clean water from each wastewater surpassed 90 %, whereas the membrane rejected more than 87 % of total dissolved solids. As a result, textile dyes were concentrated on the feed side of the membrane, which enables their recovery and potential reutilization in a subsequent dying process, along with the reclaimed water. The HFFO2 membrane was efficiently cleaned by a backwash process, restoring the initial water flux. These results indicate the suitability of forward osmosis to reuse dyes and water from textile wastewater, reducing the environmental impact of this industry and favoring its sustainability.

1. Introduction

Textile industry is a great water consumer. In the textile field, water is employed in numerous stages during the production of garments and other fabric items. Some of the most water-consuming stages are the textile printing and dyeing [1]. Furthermore, as a result of these processes, the vast amount of employed fresh water is transformed into enormous volumes of wastewater [2,3]. Textile wastewaters are problematic, since they are related to high values of chemical oxygen demand (COD), pH, color, and turbidity [4]. Moreover, some of the compounds present in these wastewaters, such as dyes or silicones, can be difficult to degrade, which further compromises their treatment and disposal.

In order to process textile wastewaters, several strategies have been proposed. One of them is the physicochemical treatment of the

* Corresponding author.
E-mail address: carsana5@upv.es (C.M. Sánchez-Arévalo).

https://doi.org/10.1016/j.heliyon.2024.e40742
Received 30 September 2024; Received in revised form 26 November 2024; Accepted 26 November 2024
Available online 28 November 2024

effluents. It includes coagulation/flocculation methods and adsorption processes [5,6]. The biological treatment of textile wastewater has also been explored. Several authors have proposed the implementation of sequencing batch reactors in order to remove the color and reduce the COD and the total suspended solids of textile effluents [7–9]. Oxidation processes, such as photo-Fenton, ozonation, and photo-catalytic treatments can also be applied to textile wastewater, aiming to degrade dyes and dissolved solids [10]. Even though these methodologies have resulted to be useful for withdrawing some dyes from the wastewaters, they are not applicable to all types of dyes. Furthermore, they can be expensive, tedious, and energy-consuming.

In this context, membrane technology can provide an efficient alternative to remove dyes and purify water. The main membrane processes that have been proposed with this objective include nanofiltration and reverse osmosis [11,12]. Despite its numerous advantages, these membrane processes have some weak aspects, such as membrane fouling and the associated energy requirements [13,14]. To avoid these problems, the application of emergent membrane technologies, such as forward osmosis (FO), can be an excellent approach [15,16]. FO operates at atmospheric pressure [17], which contrasts with the application of 10–35 bar that corresponds to nanofiltration and reverse osmosis. As a result, FO membranes do not suffer from a severe fouling and, in consequence, it determines lower energy requirements and less waste of cleaning agents and time. The application of FO to treat textile wastewater is not abundant in the literature [18–20]. Moreover, most of the research in this field has dealt with low volumes of textile effluent. However, the potential scaling of this technique determines the necessity of studying the process with larger volumes of the implied solutions, even at a laboratory scale.

In this work, FO was employed to process textile wastewaters, aiming to concentrate the dyes (in order to later reuse them) and, at the same time, obtain clean water. Several valuable contributions have explored the concentration of dyes by means of laboratory-scale FO. These contributions include the processing of low volumes of effluents, in the range of 2–10 L. Normally, flat sheet membranes of reduced area are employed [18,21], corresponding to a maximum 42 cm^2 in some cases [15,20,22] and 10 cm^2 in other cases [5,23,24]. One of the innovations of this work's approach is that large volumes of wastewater were treated (50 L) by means of a large membrane area (2.3 m^2), maintaining a large-scale perspective. Furthermore, the feasibility of FO to treat a wide range of textile effluents (whose composition may differ a lot), was demonstrated by treating different wastewaters, including the textile wastewater coming from the dyeing of different types of fabrics (wool and cotton) and from the textile equalization tank. As it will be presented in this paper, FO was a successful strategy even during the treatment of challenging textile wastewater, with higher values of conductivity and total solids.

2. Materials and methods

2.1. Textile wastewater

The employed wastewaters were provided by a textile industry, which is one of the main underwear producers in Denmark. The main stages of a dyeing process in a textile industry are reflected in Fig. 1.

Three types of wastewater were treated, resulting from the dyeing of cotton textiles, the dyeing of wool textiles, and the equalization tank, which is employed during the conventional treatment of textile wastewaters and contains a mixture of cotton dyeing wastewater and wool dyeing wastewater. In this case, the wastewater corresponded to the effluent at the end of the pipe. To perform a representative evaluation of the latter wastewater, two different streams (from two different working days) were collected from the equalization tank. Furthermore, the cotton dyeing wastewater was diluted five times (due to its high values of pH and total dissolved

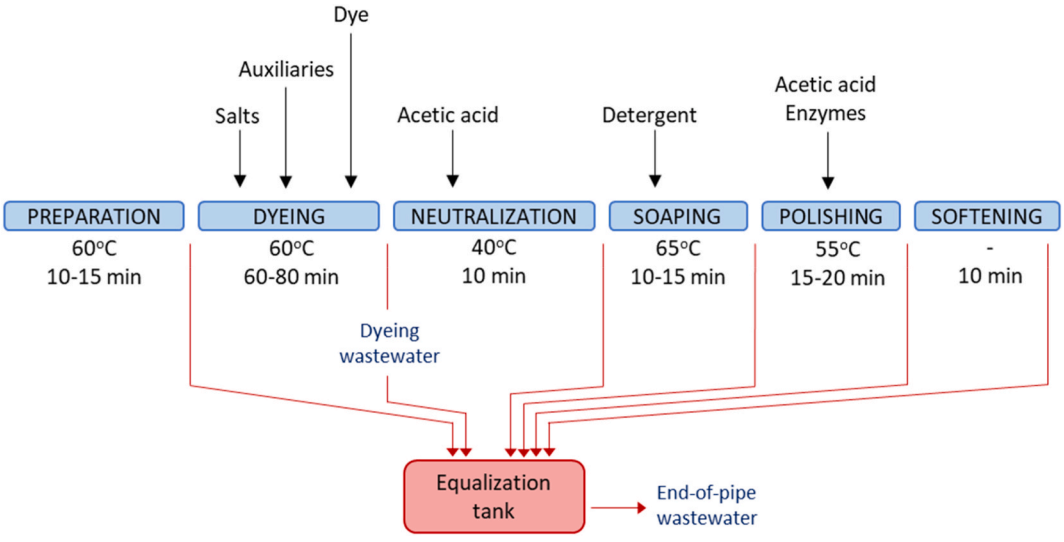


Fig. 1. Stages implied in the textile dyeing process, including the collection points for the wastewater studied in this work.

solids), and the treatment of both the original stream and the diluted stream was evaluated in the membrane plant. Therefore, five streams (cotton dyeing, diluted cotton dyeing, wool dying, end-of-pipe 1, and end-of-pipe 2) were studied.

The company that provided the wastewaters employs reactive dyes to manufacture cotton and wool textile. Additionally, emulsifiers, surfactants, enzymes, and salts are used during the process.

2.2. Membranes and experimental setup

2.2.1. Membrane plant setup

The plant (Fig. 2) was equipped with a 100 L tank to contain the draw solution, and a 50 L tank to contain the feed solution.

Mass data were obtained with a Kern FKB 36K0.1 balance (Kern & Sohn, Germany) connected to a laptop for an automatic data recording. Water flux (J_w) was calculated according to equation (1):

$$J_w = \frac{\Delta V}{A \cdot \Delta t} \tag{1}$$

where ΔV indicates the volume increment in liters, A indicates the membrane area in m^2, and Δt indicates the time increment in hours. The reverse flux of solutes (J_s) was calculated according to equation (2):

$$J_s = \frac{m_s}{A \cdot \Delta t} \tag{2}$$

where m_s refers to the mass of solutes in grams.

The HFFO2 hollow-fiber membrane (Aquaporin Inside®) was employed. Aquaporin Inside® membranes are designed to achieve a very high water flux, while allowing a high salt rejection [25]. This membrane has been previously tested with feed solutions containing caffeine, niacin, urea, and olive mill wastewater [26,27]. However, its performance during the treatment of textile effluent has not been explored before. The characteristics of the membrane are indicated in Table 1. As can be seen in the table, the maximum operating temperature of the HFFO2 membrane is 30 °C. The temperature of textile wastewater can be above this value. Therefore, if FO is applied, the temperature of the dyebaths should be reduced. In this context, heat exchangers can contribute to the sustainability of the textile industry by recovering the heat from the wastewater. This heat can be employed to increase the temperature of dyebaths in subsequent dyeing stages. Also, a suitable alternative to avoid damaging the membrane (because of a high temperature) is placing the FO plant after the equalization tank (where the end-of-pipe effluents of this work were collected), which homogenizes the composition of the textile effluent as well as the temperature. The temperature of the effluents in the equalization tank is room temperature, which cannot compromise the membrane integrity.

In this work, the membrane plant was always operated with the membrane active layer facing the feed stream. After wetting the membrane module (lumen and shell sides) with deionized water for 30 min (in single-pass mode), the J_w and the J_s of the pristine membrane was tested, employing deionized water as feed and 0.5 M NaCl as draw solution. The process was performed in single-pass mode, during 5 min. To that end, the flow rate was set at 60 L h^{-1} and 25 L h^{-1} (as recommended by the manufacturer) for the feed and draw streams, respectively. A transmembrane pressure of 0.2 bar (feed to draw) was applied. These conditions were also used to test J_w and J_s after the treatment of the wastewater and after the membrane cleaning.

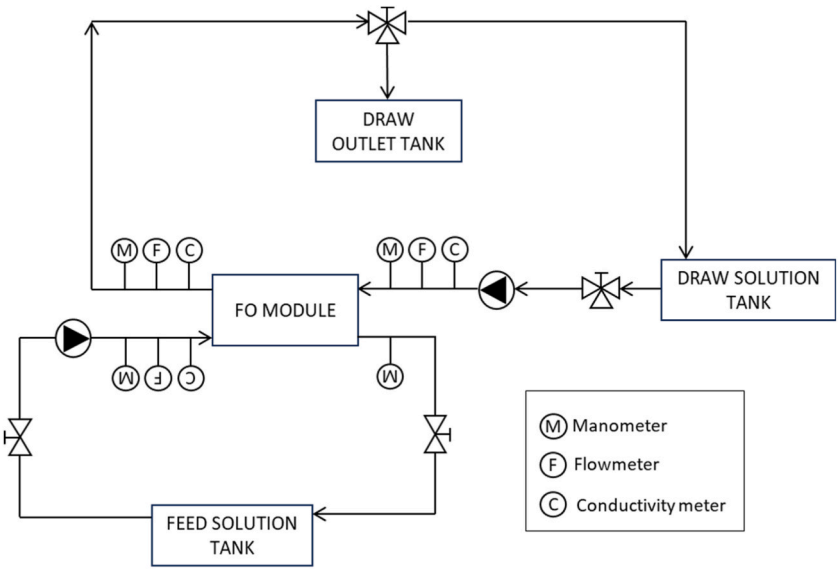


Fig. 2. Scheme of the employed forward osmosis (FO) plant.

Table 1
Characteristics of the membrane employed in this work.

PARAMETER	HFFO2 MEMBRANE
FIBER INNER DIAMETER (MM)	0.2
NUMBER OF FIBERS	30800 [26]
MATERIAL	Polyamide thin film composite with integrated water channels
MEMBRANE AREA (M^2)	2.3
WATER FLUX ($L{\cdot}H^{-1}{\cdot}M^{-2}$)	11.0 ± 1.5[a]
SPECIFIC REVERSE SALT FLUX ($G{\cdot}L^{-1}$)	0.15 ± 0.05[b]
TEMPERATURE (^{O}C) AND pH RANGE	5-30 (pH 3–10)

[a] Testing conditions: 0.5 M NaCl (2.9 %) draw vs deionized water (forward osmosis), 25 °C, single pass mode, counter-current flow, 60 L h^{-1} feed flow rate, 25 L h^{-1} draw flow rate, 0.2 bar.
[b] Testing conditions: 0.5 M NaCl (2.9 %) draw vs. deionized water (forward osmosis), 25 °C, single pass mode, counter-current flow, 400 L h^{-1} feed flow rate, 200 L h^{-1} draw flow rate, 0.2 bar.

When textile wastewaters were treated, 50 L of each effluent was employed. A solution of NaCl was used as the draw solution for the FO process. To define the concentration of NaCl in the draw solution, the following equation was applied:

$$TDS_{draw} = 1.2 \cdot \ TDS_{concentrated\ feed} = TDS_{initial\ feed} \cdot VCF \tag{3}$$

where TDS_{draw}, $TDS_{concentrated\ feed}$, and $TDS_{initial\ feed}$ correspond to the total dissolved solids of the draw solution, the final concentrated feed and the initial feed, respectively. *VCF* corresponds to the volume concentration factor. This reasoning was applied in order to ensure a sufficient osmotic pressure in the draw solution, even when the FO process was advanced and a high percentage of water was recovered. Therefore, the selected concentration for each draw solution can be found in Table 2.

The flow rates corresponding to the feed and the draw solutions were 60 L h^{-1} and 25 L h^{-1}, respectively.

The percentage of recovered water (*recovery*) in a determined time (t) during the FO process was calculated according to the following equation:

$$Recovery_t = \frac{Feed\ mass_0 - Feed\ mass_t + Feed\ sample\ mass}{Feed\ mass_0} \cdot 100 \tag{4}$$

2.2.2. Dyeing test

In order to study the possible adsorption of the dyes on the membrane surface, several flat sheet FO membranes prepared with the same active layer as the hollow-fiber HFFO2 membrane were dyed with the concentrated stream obtained during each FO experiment. To that end, the obtained concentrated stream was filtered employing a 40 μm cartridge filter (Merck, Germany). After the filtration, the flat sheet membranes were placed in petri dishes and they were covered with the concentrated streams obtained after treating the effluents from the cotton dyeing, diluted cotton dyeing, wool dying, end-of-pipe mixture 1, and end-of-pipe mixture 2. Contact times of 1 min, 5 min, 15 min, 30 min, 1 h, 5 h and 24 h were evaluated. After this time, the membranes were rinsed with distilled water and dried at room temperature to be compared.

2.2.3. Cleaning in place

To clean the membrane, an osmotic backwash was applied, employing a 0.7 M NaCl solution as feed, whereas reverse osmosis water was used as draw solution. The membrane was considered to be clean when 90 % of the initial water flux was recovered.

2.2.4. Analysis of streams

To characterized the textile wastewaters, chemical oxygen demand (COD) was evaluated by the Dichromate methodology [28], employing a Hach Lange DR3900 spectrophotometer (Hach Lange, USA); total organic carbon (TOC) was assessed by means of a multi N/C 3100 equipment (Analytik Jena, Germany). pH was measured employing a Jenway 3510 pHmeter. The data related to total dissolved solids (TDS) and conductivity were measured employing a ULTRAMETER II™ 4P (Myron L®, USA). Finally, an Osmomat 030 (Gonotec, Germany) was used to measure the osmolality of the samples. Sample characterization was conducted in duplicates.

This characterization allowed to calculate the rejection of the membrane (R), according to the following equation:

$$R\ (\%) = \frac{C_{draw}}{C_{feed}} \cdot 100 \tag{5}$$

Table 2
Molarity of the draw solution employed to treat each type of textile wastewater.

	WASTEWATER				
	Wool dyeing	Cotton dyeing	Cotton dyeing (diluted)	End-of-pipe 1	End-of-pipe 2
CONCENTRATION OF NaCl IN THE DRAW SOLUTION (M)	1.2	6.4	1.2	0.7	0.9

where C_{draw} refers to the concentration in the draw solution and C_{feed} refers to the concentration in the feed solution.

3. Results and discussion

3.1. Characterization of the textile effluents

All textile wastewaters were characterized in terms of COD, TOC, TDS, conductivity, pH, and osmolality. As shown in Table 3, the COD and TOC of all wastewaters surpassed 320 mg L^{-1} and 170 mg L^{-1}, respectively. Regarding the TDS, the cotton dyeing wastewater displayed a much higher value. In fact, the TDS content of the cotton dyeing wastewater was more than five times larger than the TDS of the wool dyeing wastewater, which was the following sample with the greatest TDS content. This was also applicable to the conductivity. Furthermore, the osmolality of the cotton dyeing wastewater was much higher.

The differences between the cotton-derived wastewater and the wool-derived wastewater can be attributed to differences in the dyeing process of both types of textiles. The high conductivity and TDS content of the cotton dyeing wastewater are due to the high proportion of salts and strong bases that are employed to fix the dye to the cellulose fibers [29,30]. A high pH is required to promote the covalent bond between the reactive dye and the hydroxyl group of cellulose. These chemicals highly increase the quantity of free ions in the wastewater, which contributes to increase the conductivity and TDS. In the case of wool, this textile is composed of protein fibers, mainly based on keratin. The amine groups of keratin can establish covalent bonds with reactive dyes at neutral or slightly basic pH [31]. Therefore, strong bases and high quantities of salts are not employed to dye wool. Instead, buffers are preferred. These buffers (based on acetate, ammonia, etc.) allow pH stabilization while reducing the ionic charge of the effluent, therefore lowering its conductivity and TDS in comparison with the cotton effluent.

For these reasons, the cotton dyeing wastewater was diluted five times, in order to consider the parameters of a more common textile effluent. Then, both the original and the diluted cotton dyeing wastewater were processed.

In order to process this kind of cotton-derived textile wastewater, several techniques have been explored before. Activated sludge and anaerobic digestion have been demonstrated to be feasible and economical, but they have limitations regarding the decolorization of the water [32,33]. Coagulation/flocculation and electrocoagulation have been employed to reduce the turbidity and COD of cotton wastewater [34]. Also, adsorption processes have been proposed [35,36]. However, these techniques imply a high cost, derived from the high doses of reagents required, the regeneration of the adsorbent or the generation of a concomitant solid waste. In this study, FO is proposed. Therefore, low energy and economic requirements will be pursued.

Regarding the pH, the wastewaters coming from the dyeing of wool and from the end-of-pipe (in its first variant) were acid, whereas the wastewater from the dyeing of cotton was basic. This is in line with the pH normally observed for the dyeing effluent of cotton and wool [37,38]. As shown by Table 3, the wool dyeing wastewater has a blue color, whereas the cotton dyeing wastewater was brown. Considering the color of the end-of-pipe mixtures, the results suggested that the end-of-pipe wastewater 1 was enriched in wool processing wastewater, whereas the end-of-pipe 2 contained a higher proportion of cotton-related wastewater. In all cases, reactive dyes were present in the wastewater. Reactive dyes are widely used for dyeing cotton and wool because they form strong covalent bonds with the fibers, resulting in excellent colorfastness and durability. The reactive groups in these dyes react with the hydroxyl groups of cellulose in cotton and the amino groups in wool, creating stable bonds that resist washing and fading [39,40].

3.2. Membrane performance

The permeate flux obtained during the treatment of the different wastewaters considered in this study is presented in Fig. 3.

As can be seen in Fig. 3, a similar permeate flux was obtained for all the textile wastewaters at the beginning of the process, except for the cotton dyeing wastewater. To treat the cotton dyeing wastewater, a draw solution with a much higher concentration of NaCl (in comparison with the rest of the draw solutions) was employed. This high concentration (Table 2) was selected in order to maintain the driving force of the process until a high water recovery was achieved. As the cotton dyeing wastewater already contained a high concentration of TDS at the beginning of the FO process (Table 3), a much higher concentration was expected at the end of the FO, which determined a high concentration of NaCl in the draw solution. This prompted a higher permeate flux for this type of wastewater. The permeate flux of the wool dyeing wastewater, diluted cotton dyeing wastewater, end-of-pipe wastewater 1, and end-of-pipe

Table 3
Characterization of the textile wastewaters in terms of chemical oxygen demand (COD), total organic carbon (TOC), total dissolved solids (TDS), conductivity, pH, apparent color, and osmolality.

	WASTEWATER				
	Wool dyeing	Cotton dyeing	Cotton dyeing (diluted)	End-of-pipe 1	End-of-pipe 2
COD ($Mg{\cdot}L^{-1}$)	1186	326	67.5	468	329
TOC ($Mg{\cdot}L^{-1}$)	528.2	181.7	38.4	225	170.4
TDS ($Mg{\cdot}L^{-1}$)	2825	15690	2940	1600	2276
CONDUCTIVITY ($\mu S{\cdot}cm^{-1}$)	3512	17930	3924	3271	2915
pH	5.5	10	9.6	6	7.1
APPARENT COLOR	Blue	Brown	Light Brown	Blue	Brown
OSMOLALITY ($Osmol{\cdot}kg^{-1}$)	0.06	0.309	0.063	0.068	0.051

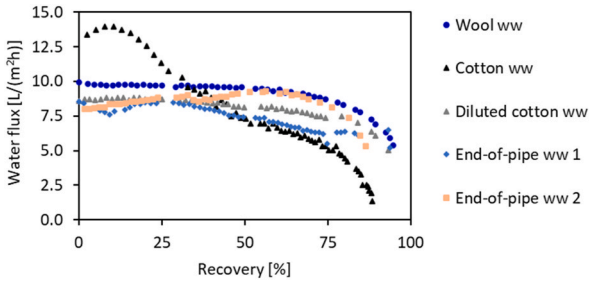


Fig. 3. Evolution of permeate flux with the percentage of water recovered during the forward osmosis process. The treatment of each wastewater (ww) lasted 2.0 ± 0.1 h.

wastewater 2 (which was between 8 and 10 L h^{-1} m^{-2}) was stable during the major part of the experiment. From a 70 % water recovery, a slight decrease in the permeate flux can be observed, which was extended until the end of the process. This decline can be attributed to a progressive loss in the osmotic pressure of the draw solution, due to its dilution at an advanced stage of the FO experiment. Furthermore, the concentration of the feed solution reduced the differences between the osmotic pressure of the feed and draw solutions, then affecting the driving force [18,41]. According to these results, the HFFO2 membrane was considered adequate to treat textile wastewaters and recover water from them, as a good performance was observed in all cases. The only exception was the processing of the dyeing cotton wastewater in its raw state, without any dilution. In this case, a sharp decrease of the initial permeate flux was exhibited when a 30 % of water was recovered, prompted by the much higher concentration of TDS and COD of this stream. This is also visible in Supplementary Figure 1, which contains the evolution of the water recovery during the FO process. A linear increment of water recovery with time was observed for all wastewater except for the raw cotton effluent. As commented in section 3.2.1, the total dissolved solids and COD content of this effluent was more than five times higher than the content of the rest of the wastewaters. This higher concentration of organic matter can cause the fouling of the membrane, thus reducing the permeate flux. Furthermore, the concentration of the feed solution reduced the differences between the osmotic pressure of the feed and draw solutions, then affecting the driving force [18,41]. In fact, as will be shown in section 3.2.4, the dyeing cotton wastewater generated the highest reduction (in comparison with the rest of wastewaters) in permeate flux after the processing of this effluent, which also indicated the highest fouling of the membrane. Also, the osmolality of this solution indicates that its osmotic pressure was much higher than the rest of the processed wastewaters, which also contributed to a lower permeate flux. According to the results obtained with the raw cotton wastewater, the dilution of this stream prior to its processing was considered a suitable strategy. Even though fresh water is required for this dilution, the treatment of the diluted cotton-derived wastewater results in a more productive process, as the membrane fouling is reduced and the permeate flux is higher. Then, a high water recovery can be achieved, regaining the water used for diluting. Also, the quantity of salt required for the draw solution is lower when the dilution is applied.

The concentration of the wastewater in the feed solution of the FO process is reflected in Fig. 4.

The images presented in Fig. 4 qualitatively illustrate that the dyes existing in the wastewaters from the dying of cotton, wool, and end-of-pipe (in its two variants) were concentrated as the water was recovered at the other side of the membrane. The textile effluents

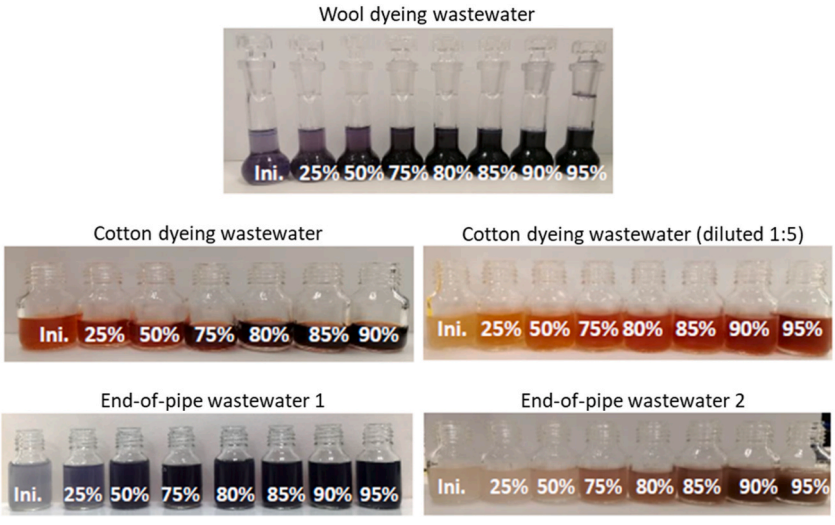


Fig. 4. Concentration of the studied textile wastewaters during the forward osmosis process. The percentage indicates the fraction of recovered water from the initial feed.

may contain a considerable percentage of functional, unfixed dyes [42]. Therefore, the concentration of these wastewaters can be considered a suitable strategy to obtain valuable streams to be used in subsequent dyeing stages, after adjusting the composition of the concentration of chemicals, if necessary. The reuse of these concentrated dyes in a new dyebath can contribute to the circular economy in the textile industry.

As presented in Fig. 3, in all cases, a 95 % water recovery was achieved, except during the treatment of the cotton dyeing wastewater, when a 90 % recovery was achieved. In this case, a higher pH, conductivity, osmolality, and concentration of total dissolved solids was determined in the feed stream, as commented in section 3.2.1. These characteristics contributed to the progressive reduction of the permeate flux, which was already very low at 85 % water recovery.

Fig. 5 shows the variation of the conductivity during the FO process. This parameter was monitored at the feed outlet (Fig. 5A) and at the draw solution outlet (Fig. 5B). The results from Fig. 5A were expected, because the conductivity of the feed solution increased progressively as the water passed through the membrane and the solution became concentrated. At the beginning of the process, all wastewaters (except the raw dyeing cotton wastewater) displayed a conductivity below 4 mS cm^{-1}, and this value surpassed 12 mS cm^{-1} at the end of the FO process. The undiluted dyeing cotton wastewater followed a similar tendency regarding the conductivity of the feed side, but the values of conductivity were much higher.

According to Fig. 5B, the draw solution suffered a fast drop in the conductivity until 25 % of the feed water was recovered. This indicated the fast dilution of the draw solution at the beginning of the process. All of the studied wastewaters displayed the same tendency. Although the draw solution employed with the raw cotton dyeing wastewater presented higher values of conductivity (due to its high concentration of NaCl (Table 2), all draw solutions featured a reduction of the initial conductivity of 1.6 times, approximately. Afterwards, the conductivity of the draw solution was more constant, around 40 mS cm^{-1}. Interestingly, for the undiluted cotton dyeing wastewater, the conductivity of the draw solution increased again after the first drop that occurred until a recovery of 25 % was reached. As reflected in Fig. 3, the permeate flux also suffered a sharp drop when the undiluted cotton dyeing wastewater was concentrated at 25 %, suggesting the accumulation of solutes at the surface of the active layer and the external fouling of the membrane [17,43,44]. Additionally, the visual inspection of the membrane module (Supplementary Figure 2) indicated that some cotton fibers passed through the cartridge filter and entered the feed solution, then contributing to the fouling of the HFFO2 membrane. In this scenario, the results from Figs. 5B and 3 indicate the passage of solutes, which were concentrated at the membrane surface, from the feed side to the draw solution. As a consequence, the conductivity of the draw solution increased, even though this stream was getting continuously diluted. This is supported by the results of Table 4, which presents the rejection values of the COD and TOC obtained during the treatment of the textile wastewaters.

In order to reuse the recovered water, a high rejection of the organic matter was pursued in all cases. The rejection of COD and TOC was always above 90 %. Only in the case of the raw cotton dyeing wastewater, the rejection of COD was below 90 % (86 ± 3 %). This can be explained by the higher concentration of total solids present in this wastewater, in comparison with the rest of them. Therefore, a higher concentration at the membrane surface is expected, then favoring the passage of solutes by diffusion across the membrane [45,46]. Furthermore, the pH of the undiluted cotton dyeing wastewater should be considered. According to Table 1, this effluent has a pH of 10, and it corresponds to the limit of the permitted pH range to preserve the stability of the membrane. Therefore, if the membrane was slightly damaged, it is reasonable that the rejection of solutes was compromised to some degree. This supports the results from Tables 2 and is in line with the decrease in the permeate flux and the increase in the conductivity of the draw solution for the undiluted cotton dyeing wastewater. Nevertheless, the HFFO2 membrane exhibited a high rejection of organic matter even in this case. Therefore, the results reflected in Table 4 were considered satisfactory.

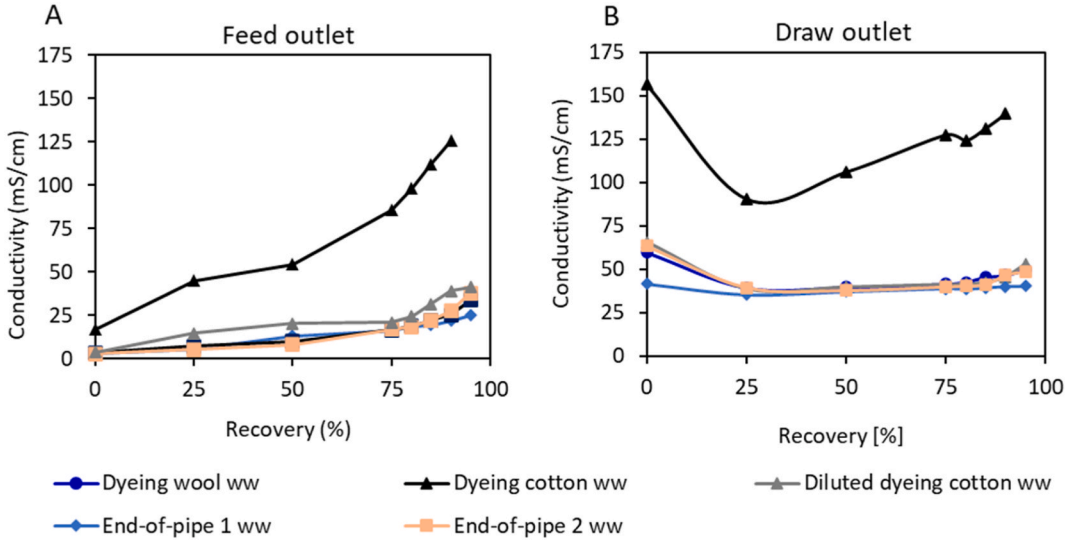


Fig. 5. Variation of the conductivity with the percentage of water recovered during the forward osmosis process. Ww refers to wastewater.

Table 4
Rejection values obtained for each textile wastewater at the end of the forward osmosis experiments. The error refers to the standard deviation. The percentage of recovered water from the feed solution has been also displayed.

	Parameter	Rejection (%)	Water recovery (%)
Wool dyeing wastewater	COD	99 ± 3	95
	TOC	100 ± 3	
Cotton dyeing wastewater	COD	86 ± 3	90
	TOC	88.3 ± 0.4	
Diluted cotton dyeing wastewater	COD	91 ± 1	95
	TOC	97 ± 3	
End-of-pipe wastewater 1	COD	95 ± 3	95
	TOC	100 ± 3	
End-of-pipe wastewater 2	COD	98 ± 4	95
	TOC	99.8 ± 0.4	

To evaluate whether the adsorption of dyes could contribute to these high rejection values, several dyeing tests (at different times) were conducted. Then, several flat sheet FO membranes prepared with the same active layer as the HFFO2 membrane were dyed employing the concentrated textile wastewater obtained after the FO process. Fig. 6 reflects the appearance of the membranes after being dyed, then rinsed with distilled water and finally dried.

As reflected in Fig. 6, most of the membranes remained clean after being dyed with the textile wastewaters. The dyes only remained on the membrane surface after a prolonged time. For most of the wastewater, this time was 24 h. In the case of the wool wastewater, the flat sheet piece retained a light blue color after 5 h. In any case, any of the FO experiments of this study took more than 3 h. Therefore, Fig. 6 indicates that the high rejection values presented in Table 4 cannot be attributed to the adsorption of dyes on the membrane surface. On the contrary, the results suggest that the HFFO2 membrane had an excellent retention capacity that allowed the recovery of water and the simultaneous concentration of textile dyes.

3.3. Cleaning of the membranes

FO operates without the application of any pressure. Therefore, membrane fouling was expected to be reduced. Nevertheless, considering the composition of the employed textile wastewaters (Table 2), a cleaning procedure was evaluated. First, water flux and reverse salt flux of the membrane after the treatment of each textile wastewater were tested and compared with the values corresponding to the pristine membrane. Fig. 7 reflects the comparison of water flux, reverse salt flux and specific reverse flux (J_s/J_w) observed with the HFFO2 membrane before and after the treatment of the wastewaters. The results indicated that the fouling of the membrane was not severe, as water flux after the processing of the wastewater was very similar to the initial water flux of the membrane. The highest reduction of water flux and the highest increase of the specific reverse flux was observed after treating the undiluted cotton dyeing wastewater. As has already been discussed (see sections 3.2.1 and 3.2.2), the cotton dyeing wastewater contained a high concentration of TDS (15690 mg L^{-1}), which prompted a higher fouling of the membrane and, thus, the reduction of water flux after the experiment. The increase in J_s/J_w suggests a possible damage of the membrane structure as a result of processing the cotton dyeing wastewater, which could be related to the high value of pH of this stream. The end-of-pipe wastewater 2 led to similar results. These results were expected, as the appearance and brown color of the wastewater suggested a high proportion of

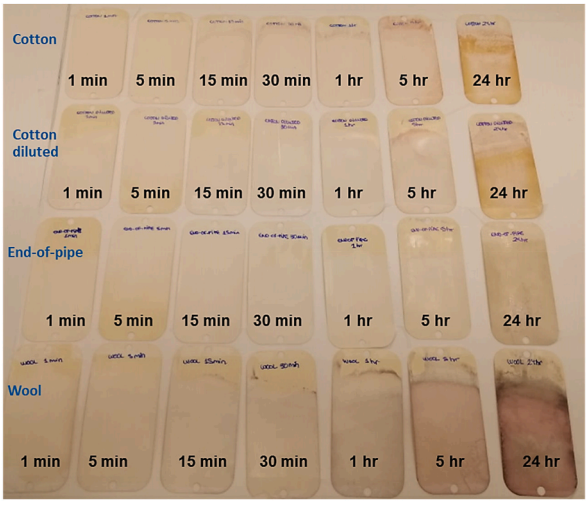


Fig. 6. Flat sheet membranes after being dyed with each concentrated wastewater, rinsed and dried.

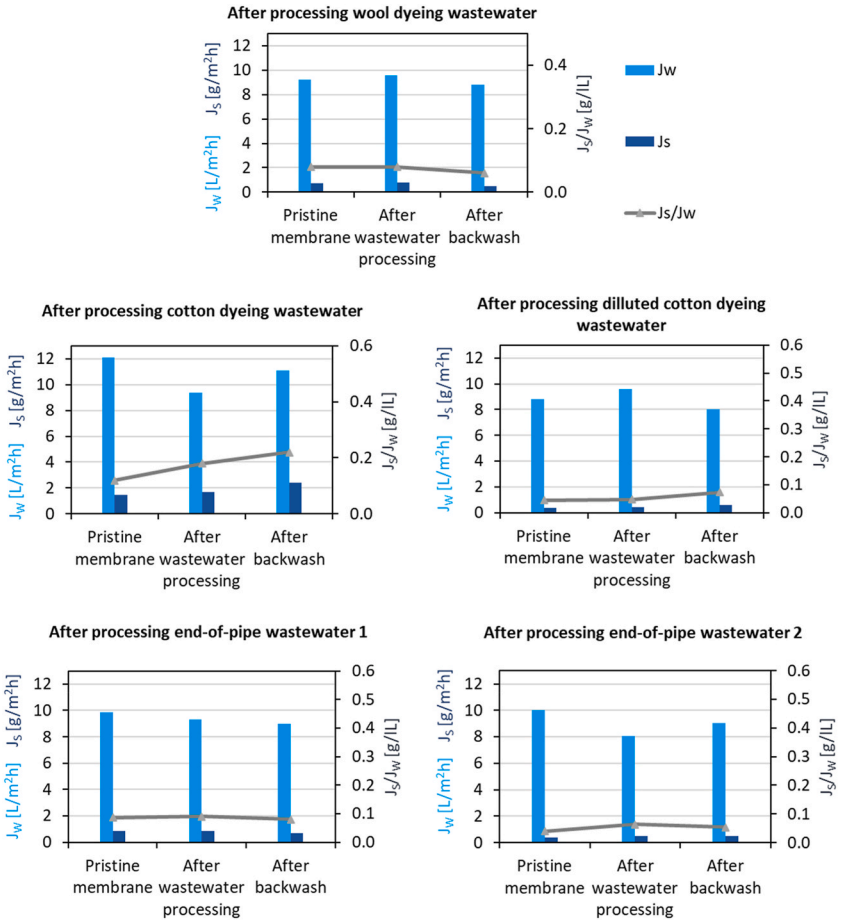


Fig. 7. Water flux (J_w), reverse salt flux (J_s) and specific reverse flux (J_s/J_w) determined for the HFFO2 membrane before its use, after the processing of each wastewater and after backwash.

cotton-derived wastewater in the mixture. In any case, the membrane was not highly fouled, as the reduction of water flux was never higher than 22 %.

As previously commented, the cleaning of the membrane was assessed by performing a backwash after the processing of each textile wastewater. The aim of this process was to induce a reverse flux of water that drove the solids deposited on the membrane surface back to the bulk solution [47]. During the processing of textile wastewaters, the adsorption of solutes on the membrane surface was plausible. Nevertheless, the results from Fig. 7 indicate that these solutes were removed during the cleaning stage. In fact, reactive dyes are designed to establish a strong interaction with wool fibers and cellulose fibers from cotton, as these materials contain hydroxyl groups, amine groups and thiol groups [48–50]. As the active layer of the FO membrane is made of polyamide, a strong interaction between the dyes and the membrane was not expected, because the affinity between reactive dyes and amide groups is reduced.

In all cases, the recovery of water flux was greater than 90 %, as can be observed in Fig. 7. This demonstrated that the application of the backwash was an effective process to remove membrane fouling.

4. Conclusions

In this work, the urgent need for treating textile-derived wastewater, such as the wastewater from cotton and wool dyeing processes, has been assessed through FO. To that end, five different types of textile wastewaters were processed, one of them featuring highly challenging characteristics, in terms of total dissolved solids and osmolality. In all cases, the HFFO2 membrane (Aquaporin) performed satisfactorily, as the recovery of clean water surpassed 90 %, being the rejection of total dissolved solids and COD above 88.3 ± 0.3 % and 86 ± 3 %, respectively. Furthermore, textile dyes were concentrated on the feed side of the membrane, which enables the recovery of water and dyes in a subsequent stage of cloth dyeing, reducing the cost and the impact of the process. Finally, the water flux of the HFFO2 membrane was recovered after an osmotic backwash process, enabling the reutilization of the membrane and contributing to the sustainability of the FO process.

CRediT authorship contribution statement

Carmen M. Sánchez-Arévalo: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation. **Laura García-Suarez:** Methodology, Investigation. **Maria Salud Camilleri-Rumbau:** Writing – review & editing, Visualization, Validation, Supervision, Investigation. **Jorg Vogel:** Writing – review & editing, Validation, Supervision. **Silvia Álvarez-Blanco:** Writing – review & editing, Visualization, Supervision, Investigation, Funding acquisition. **Beatriz Cuartas-Uribe:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Investigation, Funding acquisition. **M. Cinta Vincent-Vela:** Writing – review & editing, Visualization, Validation, Supervision, Resources, Project administration, Investigation, Funding acquisition.

Data availability

Experimental data can be found in the Zenodo community of the TEXMEM project (https://zenodo.org/communities/texmem/).

Funding

This work was supported by the research project PID2022-138299OB-I00, funded by MCIN/AEI /10.13039/501100011033 and by FEDER, UE.

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.

Acknowledgements

Authors would like to acknowledge L.P. Hansens Eftf. Textilfarveri in Bredsten, Denmark, (Dilling Underwear) for providing the real textile wastewaters. Also, authors are thankful to Erica Key Lundqvist for supporting the chemical analysis and experimental work.

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.heliyon.2024.e40742.

References

[1] E. Sahinkaya, A. Yurtsever, Ö. Çınar, Treatment of textile industry wastewater using dynamic membrane bioreactor: impact of intermittent aeration on process performance, Sep. Purif. Technol. 174 (2017) 445–454, https://doi.org/10.1016/j.seppur.2016.10.049.
[2] R. Kant, Textile dyeing industry an environmental hazard, Nat. Sci. 4 (2012) 22–26, https://doi.org/10.4236/ns.2012.41004.
[3] N. Nahar, M. Sazzadul Haque, S.E. Haque, Groundwater conservation, and recycling and reuse of textile wastewater in a denim industry of Bangladesh. https://doi.org/10.1016/j.wri.2024.100249, 2024.
[4] W. Tianzhi, W. Weijie, H. Hongying, S.T. Khu, Effect of coagulation on bio-treatment of textile wastewater: quantitative evaluation and application, J. Clean. Prod. 312 (2021) 127798, https://doi.org/10.1016/J.JCLEPRO.2021.127798.
[5] G. Han, C.Z. Liang, T.S. Chung, M. Weber, C. Staudt, C. Maletzko, Combination of forward osmosis (FO) process with coagulation/flocculation (CF) for potential treatment of textile wastewater, Water Res. 91 (2016) 361–370, https://doi.org/10.1016/J.WATRES.2016.01.031.
[6] A. Mortadi, E.G. Chahid, A. Elmelouky, M. Chahbi, N. El Ghyati, S. Zaim, O. Cherkaoui, R. El Moznine, Complex electrical conductivity as a new technique to monitor the coagulation-flocculation processes in the wastewater treatment of the textile Industry, Water Resour. Ind. 24 (2020) 100130, https://doi.org/10.1016/J.WRI.2020.100130.
[7] M.S. Nawaz, M. Ahsan, Comparison of physico-chemical, advanced oxidation and biological techniques for the textile wastewater treatment, Alex. Eng. J. 53 (2014) 717–722, https://doi.org/10.1016/j.aej.2014.06.007.
[8] M. Nuid, A. Aris, R. Krishnen, S. Chelliapan, K. Muda, Pineapple wastewater as co-substrate in treating real alkaline, non-biodegradable textile wastewater using biogranulation technology, J. Environ. Manag. 344 (2023) 118501, https://doi.org/10.1016/J.JENVMAN.2023.118501.
[9] L. Rendón-Castrillón, M. Ramírez-Carmona, C. Ocampo-López, F. González-López, B. Cuartas-uribe, J.A. Mendoza-Roca, Case Studies in Chemical and Environmental Engineering Treatment of water from the textile industry contaminated with indigo dye : a hybrid approach combining bioremediation and nanofiltration for sustainable reuse Leidy Rend o, Case Stud. Chem. Environ. Eng. 8 (2023) 100498, https://doi.org/10.1016/j.cscee.2023.100498.
[10] R. Kishor, D. Purchase, G.D. Saratale, R.G. Saratale, L.F.R. Ferreira, M. Bilal, R. Chandra, R.N. Bharagava, Ecotoxicological and health concerns of persistent coloring pollutants of textile industry wastewater and treatment approaches for environmental safety, J. Environ. Chem. Eng. 9 (2021) 105012, https://doi.org/10.1016/J.JECE.2020.105012.
[11] J. Lin, W. Ye, M. Xie, D.H. Seo, J. Luo, Y. Wan, B. Van der Bruggen, Environmental impacts and remediation of dye-containing wastewater, Nat. Rev. Earth Environ. 411 (4) (2023) 785–803, https://doi.org/10.1038/s43017-023-00489-8.
[12] Z. Zhang, Y. Wu, L. Luo, G. Li, Y. Li, H. Hu, Application of disk tube reverse osmosis in wastewater treatment: a review, Sci. Total Environ. 792 (2021), https://doi.org/10.1016/J.SCITOTENV.2021.148291.
[13] G. Mahmodi, R.R. Bafti, N.I. Boroujeni, S. Pradhan, S. Dangwal, B. Sengupta, V. Vatanpour, M. Sorci, M. Fathizadeh, P. Bikkina, G. Belfort, M. Yu, S.J. Kim, Improving cellulose acetate mixed matrix membranes by incorporating hydrophilic MIL-101(Cr)-NH2 nanoparticles for treating dye/salt solution, Chem. Eng. J. 477 (2023) 146736, https://doi.org/10.1016/j.cej.2023.146736.

[14] Y. Hou, H. Yang, Q. Fu, J. Yu, H. Zhong, W. Lin, J. Wang, J. Fang, F. Zhu, G. Ouyang, Peroxymonosulfate based in situ chemical oxidation: an efficient strategy for mitigation of membrane fouling in real seawater reverse osmosis desalination, Chem. Eng. J. 473 (2023) 145416, https://doi.org/10.1016/J.CEJ.2023.145416.
[15] M. Cifuentes-Cabezas, L. García-Suarez, J.L. Soler-Cabezas, B. Cuartas-Uribe, S. Álvarez-Blanco, J.A. Mendoza-Roca, M.C. Vincent-Vela, Feasibility of forward osmosis to recover textile dyes using single salts and multicomponent draw solutions, Membranes 13 (2023) 1–16, https://doi.org/10.3390/membranes13120911.
[16] J. Song, M. Yan, J. Ye, S. Zheng, L.Y. Ee, Z. Wang, J. Li, M. Huang, Research progress in external field intensification of forward osmosis process for water treatment: a critical review, Water Res. 222 (2022) 118943, https://doi.org/10.1016/j.watres.2022.118943.
[17] K. Lutchmiah, A.R.D. Verliefde, K. Roest, L.C. Rietveld, E.R. Cornelissen, Forward osmosis for application in wastewater treatment: a review, Water Res. 58 (2014) 179–197, https://doi.org/10.1016/j.watres.2014.03.045.
[18] J. Korenak, C. Hélix-Nielsen, H. Bukšek, I. Petrinić, Efficiency and economic feasibility of forward osmosis in textile wastewater treatment, J. Clean. Prod. 210 (2019) 1483–1495, https://doi.org/10.1016/j.jclepro.2018.11.130.
[19] A.H. Behroozi, A. Hemmati, MXene-based membranes for water desalination, in: MXenes Their Compos. Synth. Prop. Potential Appl., Elsevier, 2021, pp. 617–648, https://doi.org/10.1016/B978-0-12-823361-0.00002-2.
[20] M. Yasmeen, M.S. Nawaz, S.J. Khan, N. Ghaffour, M.Z. Khan, Recovering and reuse of textile dyes from dyebath effluent using surfactant driven forward osmosis to achieve zero hazardous chemical discharge, Water Res. 230 (2023) 119524, https://doi.org/10.1016/J.WATRES.2022.119524.
[21] A. Ammar, I. Dofan, V. Jegatheesan, S. Muthukumaran, L. Shu, Comparison between nanofiltration and forward osmosis in the treatment of dye solutions, Desalin. Water Treat. 54 (2015) 853–861, https://doi.org/10.1080/19443994.2014.908419.
[22] C.S. Lin, K.L. Tung, Y.L. Lin, C. Di Dong, C.W. Chen, C.H. Wu, Fabrication and modification of forward osmosis membranes by using graphene oxide for dye rejection and sludge concentration, Process Saf. Environ. Protect. 144 (2020) 225–235, https://doi.org/10.1016/J.PSEP.2020.07.007.
[23] M. Li, X. Wang, C.J. Porter, W. Cheng, X. Zhang, L. Wang, M. Elimelech, Concentration and recovery of dyes from textile wastewater using a self-standing, support-free forward osmosis membrane, Environ. Sci. Technol. 53 (2019) 3078–3086, https://doi.org/10.1021/acs.est.9b00446.
[24] K. Li, M. Li, W. Zhang, Y. Xue, Z. Wang, X. Zhang, A staged forward osmosis process for simultaneous desalination and concentration of textile wastewaters, ACS ES T Water 3 (2023) 1817–1825, https://doi.org/10.1021/acsestwater.2c00314.
[25] H. Wang, T.S. Chung, Y.W. Tong, Study on water transport through a mechanically robust Aquaporin Z biomimetic membrane, J. Membr. Sci. 445 (2013) 47–52, https://doi.org/10.1016/j.memsci.2013.05.057.
[26] V. Sanahuja-Embuena, G. Khensir, M. Yusuf, M.F. Andersen, X.T. Nguyen, K. Trzaskus, M. Pinelo, C. Helix-Nielsen, Role of operating conditions in a pilot scale investigation of hollow fiber forward osmosis membrane modules, Membranes 9 (2019), https://doi.org/10.3390/membranes9060066.
[27] M. Cifuentes-Cabezas, S. Álvarez-Blanco, J.A. Mendoza-Roca, M.C. Vincent-Vela, J.M. Gozálvez-Zafrilla, Theoretical model for the prediction of water flux during the concentration of an olive mill wastewater model solution by means of forward osmosis, Membranes 13 (2023), https://doi.org/10.3390/membranes13080745.
[28] C. Bendicho, I. Lavilla, Water analysis | sewage, in: Encycl. Anal. Sci., Elsevier, 2019, pp. 371–381, https://doi.org/10.1016/B978-0-12-409547-2.11519-7.
[29] W. Ad, Advance Research in Textile Engineering Effect and Role of Salt in Cellulosic Fabric Dyeing, vol. 6, 2021, pp. 2–6.
[30] J. Koh, Dyeing of cellulosic fibres, in: Handb. Text. Ind. Dye., Elsevier, 2011, pp. 129–146, https://doi.org/10.1533/9780857094919.1.129.
[31] L. Chen, B. Wang, J. Chen, X. Ruan, Y. Yang, Characterization of dimethyl sulfoxide-treated wool and enhancement of reactive wool dyeing in non-aqueous medium, Textil. Res. J. 86 (2016) 533–542, https://doi.org/10.1177/0040517515591784.
[32] D. Georgiou, A. Aivasidis, Cotton-textile wastewater management: investigating different treatment methods, Water Environ. Res. 84 (2012) 54–64, https://doi.org/10.2175/106143011X13203357278381.
[33] S. Kim, C. Park, T.-H. Kim, J. Lee, S.-W. Kim, COD reduction and decolorization of textile effluent using a combined process, J. Biosci. Bioeng. 95 (2003) 102–105, https://doi.org/10.1016/S1389-1723(03)80156-1.
[34] Y.G. Asfaha, F. Zewge, T. Yohannes, S. Kebede, Investigation of cotton textile industry wastewater treatment with electrocoagulation process: performance, mineralization, and kinetic study, Water Sci. Technol. 85 (2022) 1549, https://doi.org/10.2166/wst.2022.061.
[35] P. Pathe, A. Biswas, N.N. Rao, S. Kaul, Physico-chemical treatment of wastewater from clusters of small scale cotton textile units, Environ. Technol. 26 (2005) 313–327, https://doi.org/10.1080/09593332608618562.
[36] G. Kadry, H.A. El-Gawad, Synthesis of azo dyes derived from 4-nitroaniline for textile coloration and their removal from effluents using chemically modified sugarcane bagasse adsorbent, Fibers Polym. 25 (2024) 3853–3873, https://doi.org/10.1007/s12221-024-00704-3.
[37] C. Pinto, A. Fernandes, A. Marques, L. Ciríaco, R.A.L. Miguel, A. Lopes, M.J. Pacheco, Reuse of wool dyeing wastewater after electrochemical treatment at a BDD anode. https://doi.org/10.1016/j.jwpe.2022.102972, 2022.
[38] A. Khatri, M.H. Peerzada, M. Mohsin, M. White, A review on developments in dyeing cotton fabrics with reactive dyes for reducing effluent pollution, J. Clean. Prod. 87 (2015) 50–57, https://doi.org/10.1016/j.jclepro.2014.09.017.
[39] R. Christie, Reactive dyes for textile fibres, in: Colour Chem., The Royal Society of Chemistry, 2014, pp. 194–211, https://doi.org/10.1039/BK9781849733281-00194.
[40] L. Hao, R. Wang, J. Liu, R. Liu, Ultrasound-assisted adsorption of anionic nanoscale pigment on cationised cotton fabrics, Carbohydr. Polym. 90 (2012) 1420–1427, https://doi.org/10.1016/j.carbpol.2012.07.010.
[41] A. Tayel, P. Nasr, H. Sewilam, Forward osmosis desalination using pectin-coated magnetic nanoparticles as a draw solution, Clean Technol. Environ. Policy 21 (2019) 1617–1628, https://doi.org/10.1007/s10098-019-01738-5.
[42] L. Shu, M. Pannirselvam, V. Jegatheesan, Nanofiltration of dye bath towards zero liquid discharge: a technical and economic evaluation, in: M. Pannirselvam, L. Shu, G. Griffin, L. Philip, A. Natarajan, S. Hussain (Eds.), Water Scarcity Ways to Reduce Impact, Springer International Publishing, Cham, 2019, pp. 47–61, https://doi.org/10.1007/978-3-319-75199-3_3.
[43] I. Ibrar, O. Naji, A. Sharif, A. Malekizadeh, A. Alhawari, A.A. Alanezi, A. Altaee, A review of fouling mechanisms, control strategies and real-time fouling monitoring techniques in forward osmosis, Water 11 (2019) 695, https://doi.org/10.3390/w11040695.
[44] F.A. Siddiqui, Q. She, A.G. Fane, R.W. Field, Exploring the differences between forward osmosis and reverse osmosis fouling, J. Membr. Sci. 565 (2018) 241–253, https://doi.org/10.1016/j.memsci.2018.08.034.
[45] A. Imbrogno, A.I. Schäfer, Micropollutants breakthrough curve phenomena in nanofiltration: impact of operational parameters, Sep. Purif. Technol. 267 (2021) 1–13, https://doi.org/10.1016/j.seppur.2021.118406.
[46] C.M. Sánchez-Arévalo, T. Croes, B. Van der Bruggen, M.C. Vincent-Vela, S. Álvarez-Blanco, Feasibility of several commercial membranes to recover valuable phenolic compounds from extracts of wet olive pomace through organic-solvent nanofiltration, Sep. Purif. Technol. 305 (2023), https://doi.org/10.1016/j.seppur.2022.122396.
[47] L. Lv, J. Xu, B. Shan, C. Gao, Concentration performance and cleaning strategy for controlling membrane fouling during forward osmosis concentration of actual oily wastewater, J. Membr. Sci. 523 (2017) 15–23, https://doi.org/10.1016/j.memsci.2016.08.058.
[48] C.H. Lee, Y. Wang, Y.L. Tang, C.W. Kan, Dyeing wool knitted fabric in nano-scale reverse micelle with reactive dyes — a computer colour matching study, Fibers Polym. 22 (2021) 1320–1332, https://doi.org/10.1007/s12221-021-0846-8.
[49] L. Wang, B. Xie, H. Hu, G. Bai, Z. He, Y. Huang, L. Liu, C. Meng, Regulating the surface charge and reactivity of cellulose through quaternized modification to achieve salt-free reactive dyeing process, Int. J. Biol. Macromol. (2024) 135898, https://doi.org/10.1016/j.ijbiomac.2024.135898.
[50] U.H. Siddiqua, S. Ali, M. Iqbal, T. Hussain, Relationship between structure and dyeing properties of reactive dyes for cotton dyeing, J. Mol. Liq. 241 (2017) 839–844, https://doi.org/10.1016/j.molliq.2017.04.057.