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Treatment of Terasil Red dye by bio-coagulation–microfiltration process: experimental study and modelling

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ABSTRACT

The present study investigates the effectiveness of acorn leaves (*Quercus canariensis*) as a bio-coagulant and microfiltration technique for removing Terasil Red dye from a synthetic solution. In this study, a zirconium oxide ceramic membrane was employed. Experimental investigations were also conducted to assess the influence of coagulant dose and solution pH on the coagulation performance. Afterward, for microfiltration (MF) and the hybrid process of bio-coagulation–microfiltration, the effect of transmembrane pressure (TMP) was evaluated. The results indicate that the pH of the solution and the dose of coagulant used have a significant impact on coagulation performance. However, the MF process is primarily influenced by TMP. Furthermore, the pH and operating pressure were found to have an important influence on the hybrid process. The experimental results showed that the highest removal efficiency for dye and turbidity removal (96.2% and 98.77%, respectively) was obtained by hybrid process at pH 3 and TMP of 1.7 bar. For the same previous conditions of pH and TMP, the permeate flux increased and reached the highest values. A comparative study between the experimental results and those obtained by simulation with SuperPro Designer software demonstrated close agreement between the findings.

Keywords: Dye; Bio-coagulant; Natural coagulant; Acorn leaves; Coagulation; Membrane filtration; Microfiltration; Hybrid process; Turbidity; SuperPro Designer

1. Introduction

In recent years, the escalating worldwide requirement for textile products has led to a concurrent rise in water pollution. The dyeing processes employed by textile mills necessitate substantial quantities of dyes and water, resulting in the generation of approximately 125–150 L of wastewater per kg of produced goods [1]. This wastewater presents distinctive characteristics, including intense coloration, elevated levels of chemical oxygen demand (COD), dissolved solids, and notable pH variability, all of which contribute to its pollutant nature [2]. Textile effluent is often discharged into watercourses without adequate treatment [3]. Moreover, the synthetic dyes in this effluent are a primary concern, as they exhibit high water solubility, even at low concentrations, compromising the aquatic ecology and posing major environmental threats [4]. Hence, eliminating these pollutants from wastewater is primordial before

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their release in nature. Several methods have been applied to treat dye-contaminated effluents, classified into 3 categories, namely biological, chemical, and physical treatment [3]. Coagulation–flocculation is a well-known treatment process for destabilizing colloids and other dispersed compounds in water, such as dyes. Nonetheless, the utilization of chemical coagulants entails certain drawbacks. These limitations include elevated operational expenses, reduced efficiency in low-temperature water, generation of high sludge volumes, and pronounced impact of wastewater pH variations [5].

However, a recent shift in practice has emerged, promoting the use of natural substances as coagulants. This trend is driven by their environmentally friendly nature in terms of both production and application, mitigating the costs associated with conventional methods while concurrently enhancing the water quality for safe use [6]. Moreover, a multitude of investigations have substantiated the efficacy of numerous plant-derived coagulants in water and wastewater treatment applications, such as *Moringa oleifera* [7], tannin [8], cactus [9], common bean [10], aloe vera [11], plantago major [12], red lentil [13].

Membrane separation methods have found a broad application in potable water and wastewater treatment due to their effectiveness in eliminating particles, turbidity, and bacteria, and reduce chemical and biological oxygen demand (COD and BOD) [14]. However, the main technical limitation in the use of membranes is the reduction of flux during filtration which is primarily caused by the accumulation of suspended solids, microorganisms, and organic substances on the membrane surface or within its pores, a phenomenon commonly referred to as membrane fouling [15]. Membrane cleaning or applying pre-treatment processes are some methods used to control the fouling [16]. In a study, coagulation–flocculation technique was employed as a preliminary treatment step for treating landfill leachate with nanofiltration and reverse osmosis membranes. They reported that ferric chloride or aluminum sulfate increased permeate flux of some membranes by more than 80% [17]. Huang et al. [18] conducted a study to evaluate the impact of magnetic ion exchange (MIEX) pre-treatment on the membrane filtration of natural surface water. The findings indicated that MIEX pre-treatment could potentially be effective in removing dissolved organic carbon (COD) from source waters. However, it was observed that additional pre-treatment methods would be necessary to enhance fouling reduction. Ozay and Dizge [16], investigated the influence of different pre-treatment methods, namely coagulation, electrocoagulation, and electrooxidation, on membrane flux during the treatment of wastewater from a pistachio processing plant using membrane processes. The experimental results demonstrated that the electrocoagulation method, specifically utilizing Al-Al electrode pairs, yielded the highest initial and steady-state flux values. The steady-state flux value was found to be 118 times higher than that of raw wastewater. In another study, the efficacy of adsorption and coagulation processes which is known to contribute to membrane fouling, were investigated. The results revealed a remarkable 95% removal efficiency of dissolved organic matter in dye wastewater Thus, it has been divulged that the pre-treatment step significantly improved membrane flux compared to untreated raw wastewater [19]. In addition, many studies have reported the efficiency of membrane flux performance with pre-treatment methods in fields such as the juice industry [20,21], dairy industry [22,23], brewery industry [24], municipal wastewater treatment [25,26]. This technology is proving to be a viable option for treating textile wastewater due to its notable environmental advantages, such as pollution reduction, water reuse, and the recovery of valuable components from the waste stream [27].

Therefore, the primary aim of this study was to assess the effectiveness of microfiltration (MF) as a membrane-based treatment process and the hybrid bio-coagulation–microfiltration (BC-MF) process. Acorn leaves were employed as a natural coagulant in these processes to remove Terasil Red textile dye from a synthetic solution.

2. Materials and methods

2.1. Preparation of acorn leaves extract with NaOH (AL-NaOH)

The acorn leaves (AL) were locally collected near the city of Mila, Algeria. After a cleaning process to remove external impurities and washing with water, the leaves were dried at 50°C, crushed, and sieved through a 0.35 mm sieve. The fraction with particle size less than 0.35 mm was selected for use in experiments. For the procedure 25 g of acorn leaves powder was mixed with 1 L of 0.05 M sodium hydroxide ($NaOH$) solution at room temperature (20°C ± 1°C). The suspension was stirred for 20 min followed by centrifugation to extract the active components, responsible for coagulation [28]. A schematic representation for the preparation of acorn leaves extract with NaOH is shown in Fig. 1. It is worth noting that numerous studies have indicated that the extracted active component responsible for the coagulation activity is of a proteinaceous nature [15,29].

2.2. Preparation of synthetic solution of Terasil Red

Terasil Red, a commercial disperse dye powder, was obtained from COTITEX Batna, Algeria and used without additional purification. The synthetic solution, for all experiments, was prepared by adding 10 mg of textile dye Terasil Red in 1 L of water. The solution was stirred gently for 5 min using magnetic stirrer for uniform dye dispersion. Properties of Terasil Red were given in Table 1.

2.3. Coagulation studies

Jar test apparatus was used to investigate the performance of acorn leaves as a coagulant and to optimize its dose and the pH of the solution [30]. Six 1,000 mL beakers were filled with 500 mL of synthetic dye solution and placed on Jar test apparatus. Each beaker received a different dose of coagulant and underwent agitation for 3 min at 160 rpm. The mixing speed was afterward reduced to 30 rpm for 25 min. Finally, the suspensions were allowed to decant for 30 min [31,32]. Hydrochloric acid (HCl, 0.1 N) or sodium hydroxide ($NaOH$, 0.1 N) were used to alter the pH of the solution.

2.4. Microfiltration experiments

The tubular ceramic membrane in this study was prepared using the clay (kaolin) (from the east of Algeria) and zirconia by the sol–gel method [33]. The membrane treatment of dye solution was conducted in a laboratory scale. The schematic presentation of the system is shown in Fig. 2. The studies were carried out in a total recirculation mode to optimize the operating parameters. The main components of the MF experimental setup comprehend a tangential inorganic membrane made of zirconia (ZrO_2) with an effective area of 2.64 × 10^{-3} m^2, and a 10 L water inlet tank. In each experience, 3 L of the synthetic solution were used. Transmembrane pressure (TMP) was controlled using a

Table 1
Properties of Terasil Red dye

Terasil Red	Properties
Color index number	Disperse Red 349
Molecular formula	$C_{23}H_{21}Cl_2N_5O_5$
Molecular weight	518.351 g/mol
UV absorption	λ_{max} 523.5 nm
CAS registry number	79542-46-4

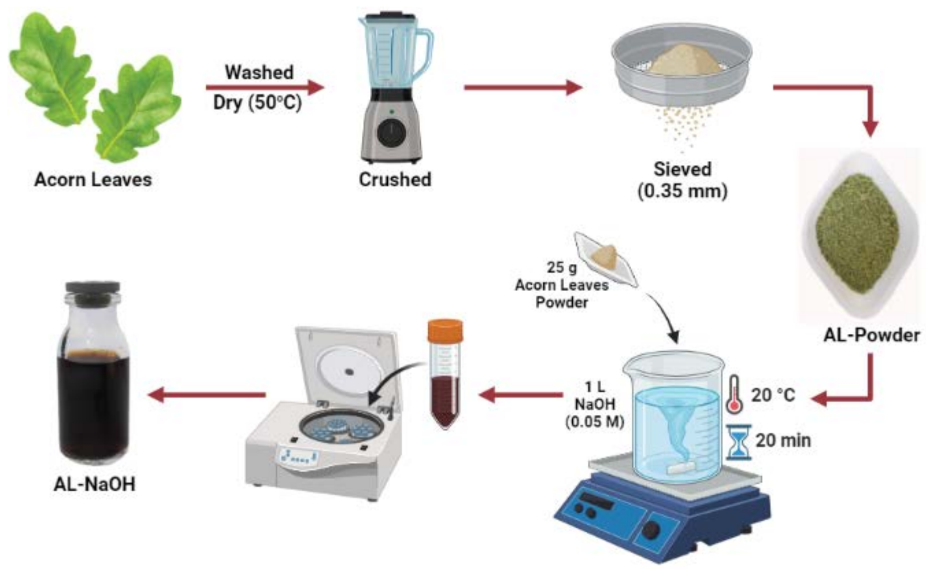

Fig. 1. Synthesis of Al-powder and AL-NaOH.

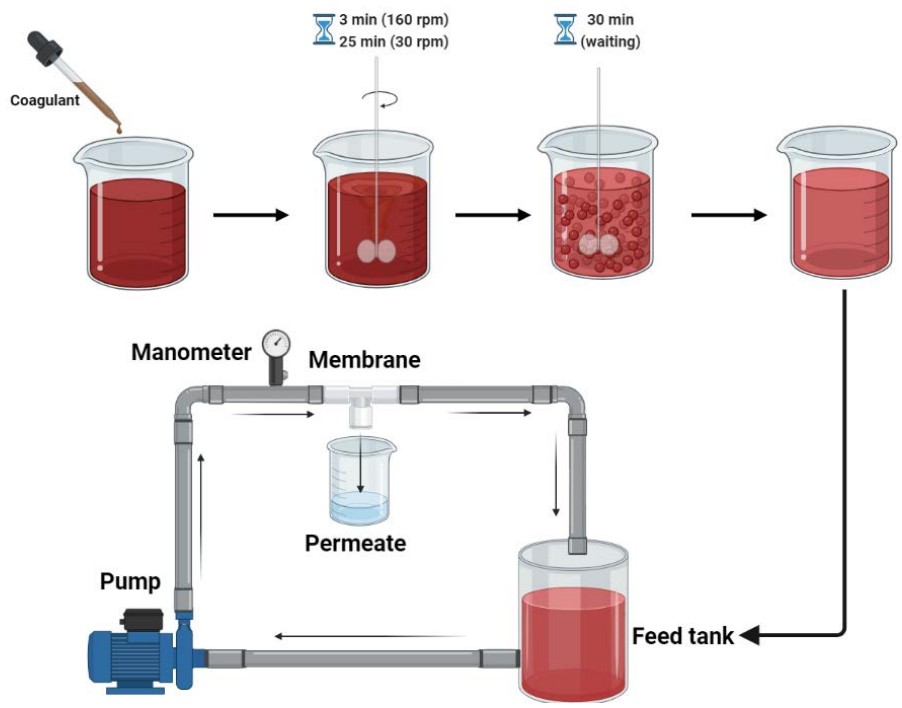

Fig. 2. Use of the solution containing Terasil Red in the bio-coagulation and subsequent membrane system.

manometer. Additionally, a constant flow pump circulated the solution from the feed tank to the membrane, with the filtrate collected in a beaker for analysis, while the concentrate solution was recirculated back to the feed tank.

The permeate flux was calculated by using Eq. (1):

$$J_P = \frac{V_P}{A \cdot t} \tag{1}$$

where J_P represents the permeate flux (L/m²·h), V_P is the permeate volume (L), A is the effective membrane area (m²), and t is the sampling time (h). The permeate flux is also governed by the so-called general filtration equation (Darcy's law) given in Eq. (2) [34]:

$$J_P = \frac{\Delta P}{\mu R_m} \tag{2}$$

where ΔP (Pa) is the applied pressure, μ (Pa·s) is the solvent viscosity, and R_m (m^{-1}) is the hydraulic resistance of membrane. For polluted water filtration, Darcy's law can be given as follows (resistance in-series model):

$$J = \frac{\Delta P}{\mu\left(R_m + R_c + R_P\right)} = \frac{\Delta P}{\mu R_r} \tag{3}$$

$$R_T = R_m + R_c + R_P \tag{4}$$

where R_T, R_c, and R_P are the total clogging and polarization resistances expressed in (m^{-1}).

2.5. Analysis

The concentration of the synthetic dye solution was measured at λ_{max} = 523.5 nm using a UV spectrophotometer Jasco V-630 (Jasco, Japan). Turbidity was obtained using a Hanna HI88703 Turbidimeter (Hanna Instruments, USA) (expressed in nephelometric turbidity units (NTU)). pH solution was measured using a Jenway 3505 pH meter (Jenway, UK). To analyze the main functions of the dye, bio-coagulant, and produced sludge, a Jasco 4600 FT/IR Spectrophotometer (Jasco, Japan) was employed. The removal efficiency of turbidity and colour after coagulation and MF treatment was determined using the Eq. (5):

$$\text{Removal Efficiency}\left(\%\right) = \frac{X_0 - X_t}{X_0} \tag{5}$$

where X_0 and X_t represent the initial measured concentration or turbidity and those in the treated solution at time t, respectively.

2.6. Simulation study

Maximizing the potential of bioprocesses currently depends significantly on modelling, simulation, and optimization techniques. SuperPro Designer stands out as widely used simulator for this purpose. Various industries, including pharmaceutical, biotechnology, chemical, mineral processing, microelectronics, water purification, wastewater treatment, air pollution control, and other sectors heavily rely on this tool [35]. The main advantages of this simulator include an extensive database of specific chemical compounds, unit operations, economic and ecological process evaluation, as well as a performance index. SuperPro Designer simplifies the modeling, evaluation, and optimization of wastewater treatment processes, capable of handling both biological and physical wastewater treatment. It also provides models for water purification in each working unit.

The following steps should be followed:

After launching the SuperPro Designer software, it should be defined whether the process is batch or continuous. Constituents must then be added, with the option to modify their properties if not available in the database. Then choose unit procedures > type of procedure, selecting: filtration > microfiltration in this case. Once the flow sheet is ready with all the input, output, and intermediate streams, it is necessary to specify the operations that will take place in each section of equipment in the process. To add or remove operations for a particular unit, right-click on the unit and choose: Add/Remove operations.

3. Results and discussion

3.1. Characterization of the prepared membranes

As shown in Fig. 3, the scanning electron microscopy characterization of the membrane support via scanning

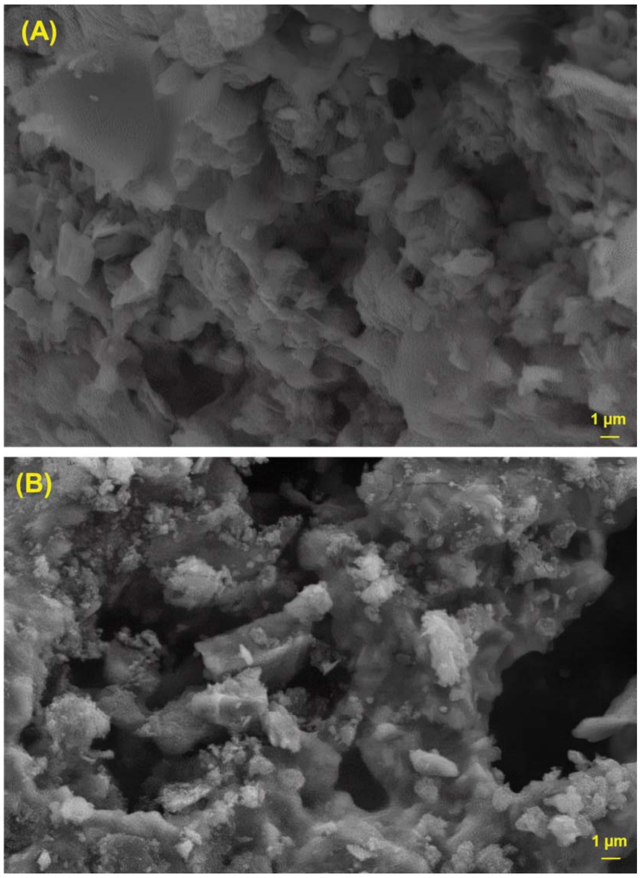

Fig. 3. Scanning electron microscopy images of the support and ceramic membrane: (A) inner and (B) outer surfaces.

electron microscopy, reveals that the inner surface of the membrane has significantly smaller pores compared to the outer surface (support). This feature allows the membrane support to effectively retain particles with diameters exceeding those of the inner surface (membrane). Consequently, the flow of the feed solution is ensured from the inside to the outside of the membrane support.

3.2. Optimization of bio-coagulation

3.2.1. Effect of coagulant dose

Determination of the optimal coagulant dose is an important parameter that directly influences the treatment's effectiveness. An inadequate dose can result in poor treatment [31]. Fig. 4 shows the effect of coagulant dose as powder and AL-NaOH on the turbidity removal. The investigation was conducted at a pH 8 and initial turbidity of 45.7 NTU. The results show the positive effect of acorn leaves on turbidity removal, with the active components extracted from AL-NaOH proving to be more efficient than acorn leaves in powder form. This difference in performance is probably related to the high protein content in AL-NaOH solution, which enhances coagulation activity [13,32]. Fig. 4 also indicates a gradual decrease in turbidity. Maximum suspend solid removal corresponds to an optimal coagulant dose of 10 mL. Beyond this dosage, a turning point is observed, leading to increased turbidity. Similar findings have been reported in numerous studies [15,33]. These phenomena are related to the coagulation mechanisms, specifically, adsorption and charge neutralization. An increasing coagulant dosage leads to particle destabilization and turbidity increase [36]. Further reduction is noticed beyond the coagulant dose of 45 mL. This may be explained by the sweep coagulation mechanism, which occurs at high coagulant dosages. It is worth noting that a low optimal coagulant dose is preferable, as excessive dosing results in an elevated concentration of organic matter in the solution, which can lead to microbial growth [15].

3.2.2. Effect of pH of synthetic dye solution

The pH of the solution is another important parameter that can affect the performance of coagulation since it can control the surface charge of the coagulant [37]. Fig. 5 shows the turbidity removal upon pH ranging from 3 to 13. Notably, the most significant removal efficiency occurs at pH levels of 3 and 13. Similar results have been reported in the studies conducted by Šćiban et al. [32], Hussain et al. [37] and Diaz et al. [38]. It was perhaps attributed to the presence of amino acids within proteins which is responsible for this turbidity removal. In an acidic medium, positively charged amino acids such as lysine and arginine control the elimination process by charge neutralization. In an alkaline pH solutions, negatively charged amino acids such as aspartate become active, leading to removal through adsorption and bridging [39,40]. On the other hand, Terasil Red is strongly affected by the alkaline medium resulting in precipitation within this pH range [41].

3.2.3. Characterization

The Fourier-transform infrared spectrum of Terasil Red, acorn leaves, and settled sludge are shown in Fig. 6. From this figure, among the functional groups of Terasil Red observed from the peaks include primary amines group with NH stretch at band 3,413 cm^{-1}, alkyl groups (2,926 cm^{-1}), may be ammonium ions (2,438 cm^{-1}), aromatic compounds (1,673 cm^{-1}), the C=N function at 1,616 cm^{-1}, and aliphatic amine or primary alcohols at 1,050 cm^{-1} [42].

In the case of the acorn leaves, the main functional groups observed can be described as follows: a band at 854 cm^{-1} is supposed to be characteristic of the hydro-peroxides bond or aromatic group, pyridines (1,614 cm^{-1}), the carboxylic acid (1,315 cm^{-1}), aliphatic amine at 1,032 cm^{-1} (strong band), at 1,483 cm^{-1}, a possible methylene or an aromatic C=C, primary amines at 1,614 cm^{-1}, the aldehydes at 1,728 cm^{-1}, and the probable presence of a methylene group at 2,850–2,918 cm^{-1}; the presence of alcohols, mainly the phenols at

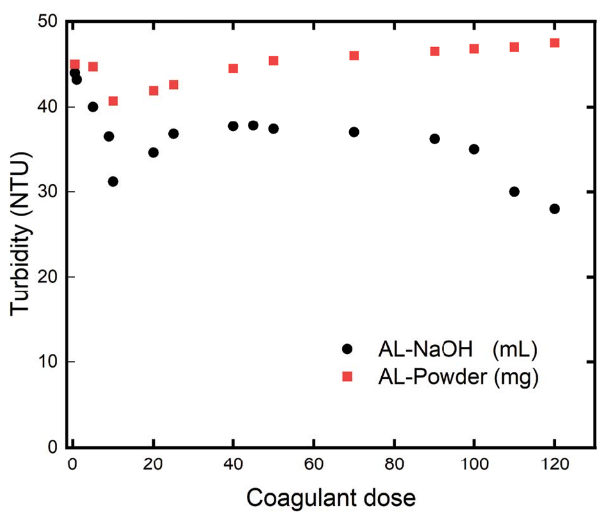

Fig. 4. Effect of coagulant dose on turbidity removal (initial conditions: dye concentration = 10 mg/L; turbidity = 45.7 NTU).

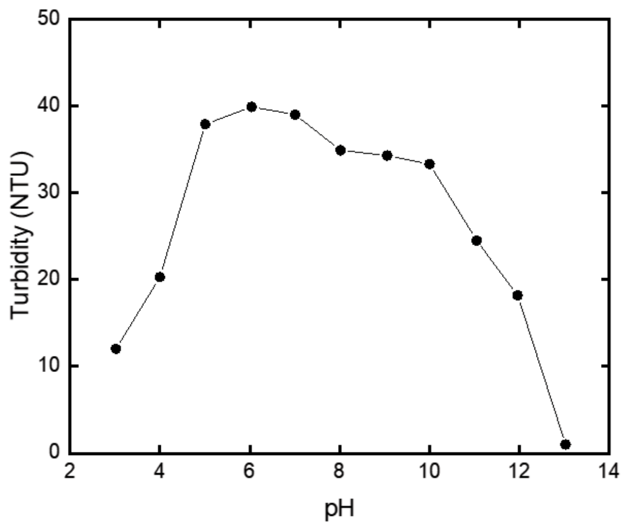

Fig. 5. Effect of pH on turbidity removal (initial conditions: dye concentration = 10 mg/L; turbidity = 45.7 NTU).

3,281 cm^{-1} [42]. Furthermore, Fig. 6 reveals a significant similarity between the IR spectra of the settled sludge and the bio-coagulant (AL), suggesting that the settled sludge also contains the bio-coagulant with a slight offset of the peaks; it is possible that some functional groups of the dye may interact with those of the bio-coagulant.

3.3. Treatment of synthetic dye solution by microfiltration and bio-coagulation–microfiltration

3.3.1. Variation of dye concentration and turbidity by microfiltration

The effectiveness of membrane filtration treatment was evaluated by dye and turbidity elimination. Experiments were conducted at normal pH of 7.92 and TMP of 1.7 bar. The results presented in Fig. 7 demonstrate that the dye concentration and turbidity in concentrate are higher than in the permeate. After 50 min of treatment, the maximum dye reduction achieved was 0.21 mg/L, corresponding to an elimination efficiency 'EE' of 94.7%. Turbidity removal reached 1.7 NTU (EE: 96.3%). These results can be attributed to two key characteristics of disperse dye: their negative surface charge and hydrophobic nature, which enhance the affinity and interaction of dye particles with the positively charged membrane surface [43].

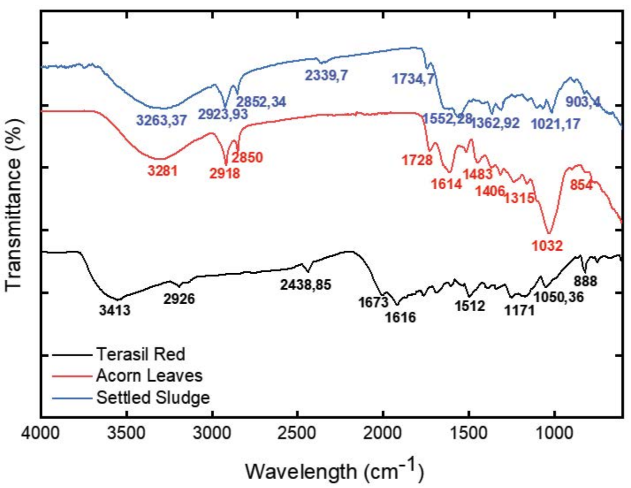

Fig. 6. Fourier-transform infrared spectrum of Terasil Red, acorn leaves, and settled sludge.

3.3.2. Permeate flux variation

The permeate flux is the volume of effluent passing through a defined membrane area per unit of time, expressed in units of litter per square meter per hour (LMH). A reduction in permeate flux indicates the accumulation of foulants on the membrane surface. Fig. 8A presents a variation of dye solution flux over time. Filtration was carried out under a TMP of 1.7 bar and normal pH of 7.92. As shown in the figure, the flux steadily decreases with filtration time. The most significant drop occurs in the first initial 15 min due to pore blockage by small dye particles (internal fouling) [30,44]. This phase is followed by a slower decline in the flux between 15 and 25 min, caused by larger dye particles forming a layer cake on the inner membrane surface [30,45]. Finally, in the last phase (25–50 min), a pseudo-steady state of flux diminution is observed.

Permeates flux variations of pure water and dye solution, as a function of applied pressure, are illustrated in Fig. 8B. Filtration time was fixed at 5 min, and TMP ranged between 0.6 and 1.7 bar.

It was observed that both fluxes increase with rising the TMP, following Darcy's law [46]. However, pure water flux was higher than dye solution due to the accumulation of dye substances on the membrane surface, acting as the counterforce to TMP [47].

The permeates flux are also studied for hybrid process, bio-coagulation–microfiltration (BC-MF), with a filtration time of 50 min, pH ranging from 3 to 10, and TMP varied from 0.6 to 1.7 bar. The results presented in Fig. 9 reveal similar permeate streams curves to those obtained in Fig. 8A, with a notable difference in flux values, influenced by pH and TMP. However, when solution pH

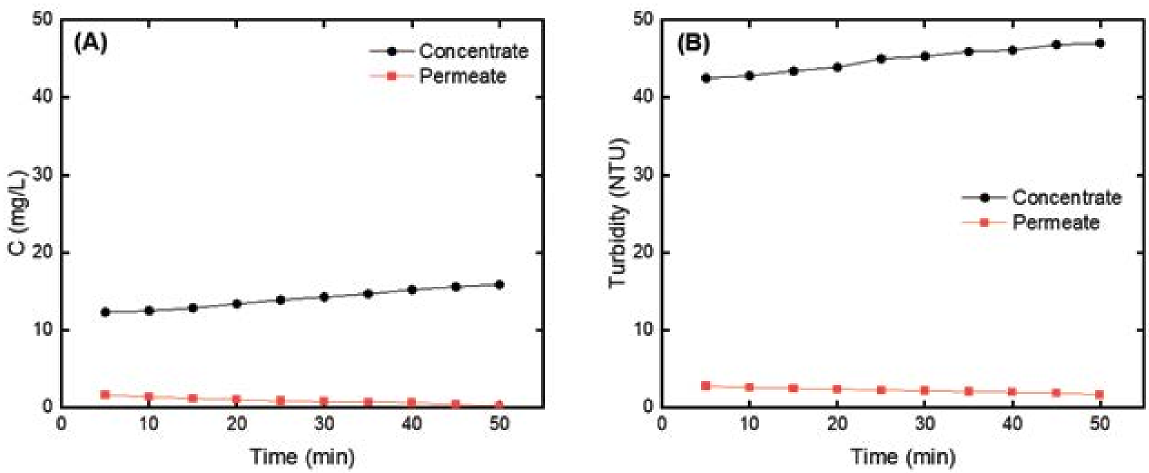

Fig. 7. Variation of dye concentration and turbidity with time: (A) dye concentration and (B) turbidity (initial conditions: dye concentration = 10 mg/L; turbidity = 45.7 NTU).

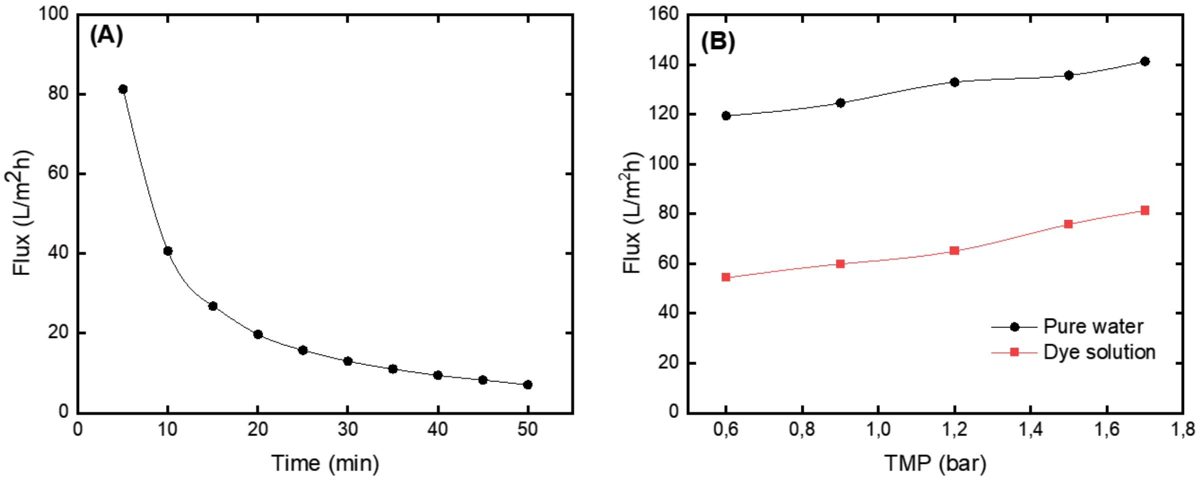

Fig. 8. (A) Variation of dye permeate flux as a function of time (MF) and (B) variation of permeate flux of pure and dye solution as a function of TMP (MF).

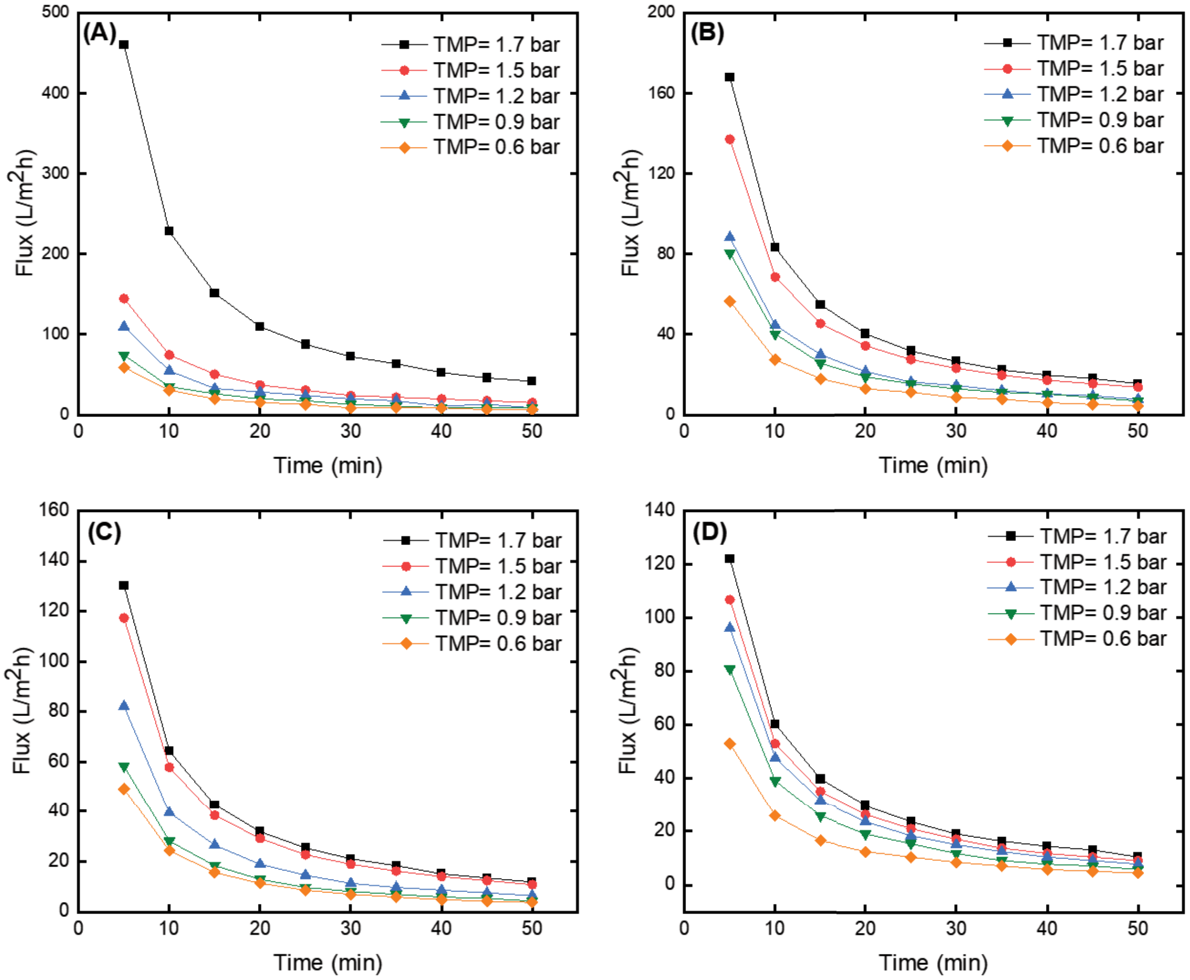

Fig. 9. Variation of permeate flux with time for different values of pH and TMP: (A) pH 3, (B) pH 6, (C) pH 8, and (D) pH 10.

increases (Fig. 9A–D), flux decreases, particularly at low pressure (due to decreased driven force at lower TMP). Consequently, fouling phenomena are important under primary medium to low TMP. In addition, it is worth mentioning that the dye precipitation occurs as the solution pH increases without the addition of bio-coagulant. Therefore,

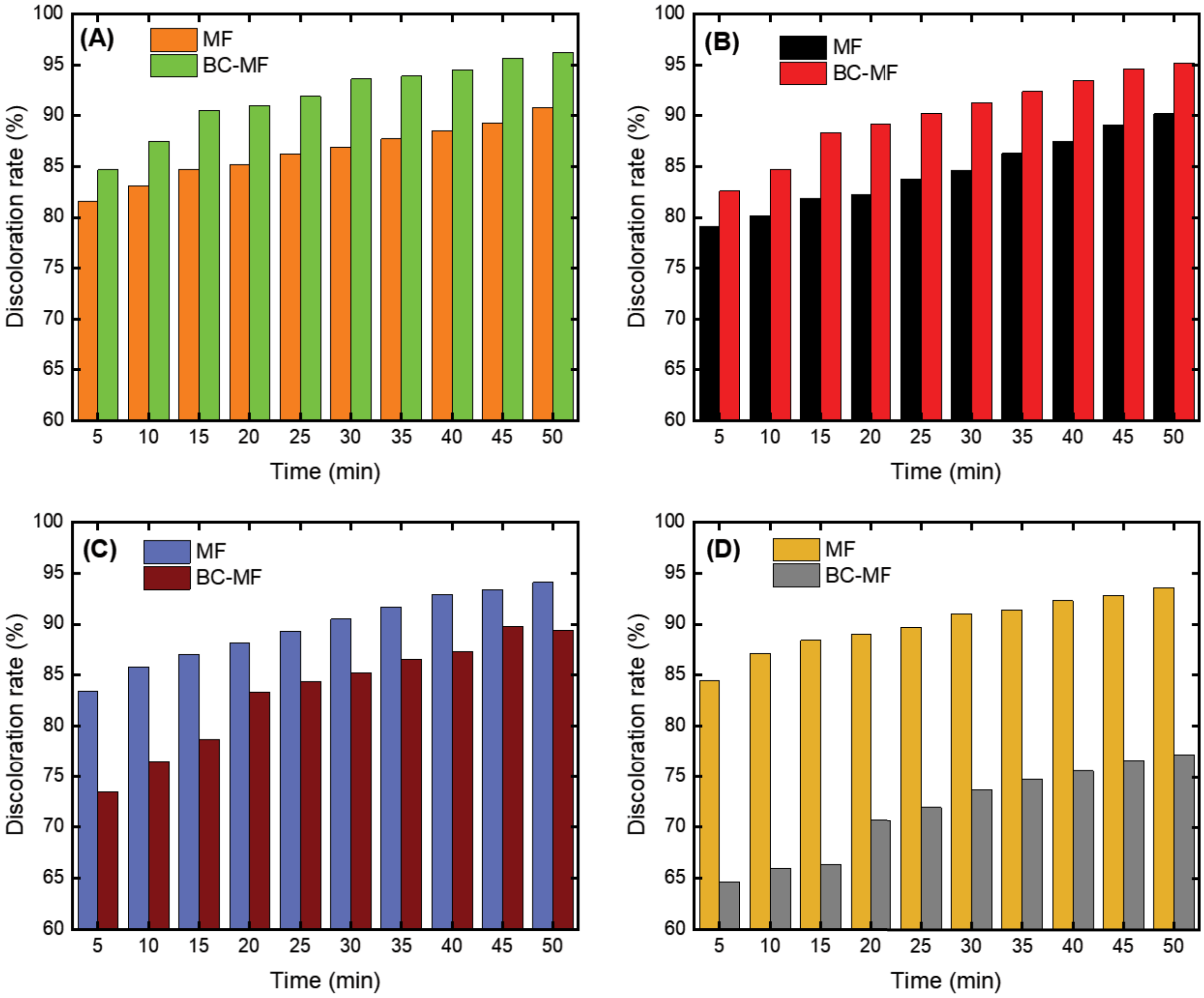

Fig. 10. Comparative study of the variation of discoloration rate (%) between MF and BC-MF with time for different pH values: (A) pH 3, (B) pH = 6, (C) pH = 8, and (D) pH = 10.

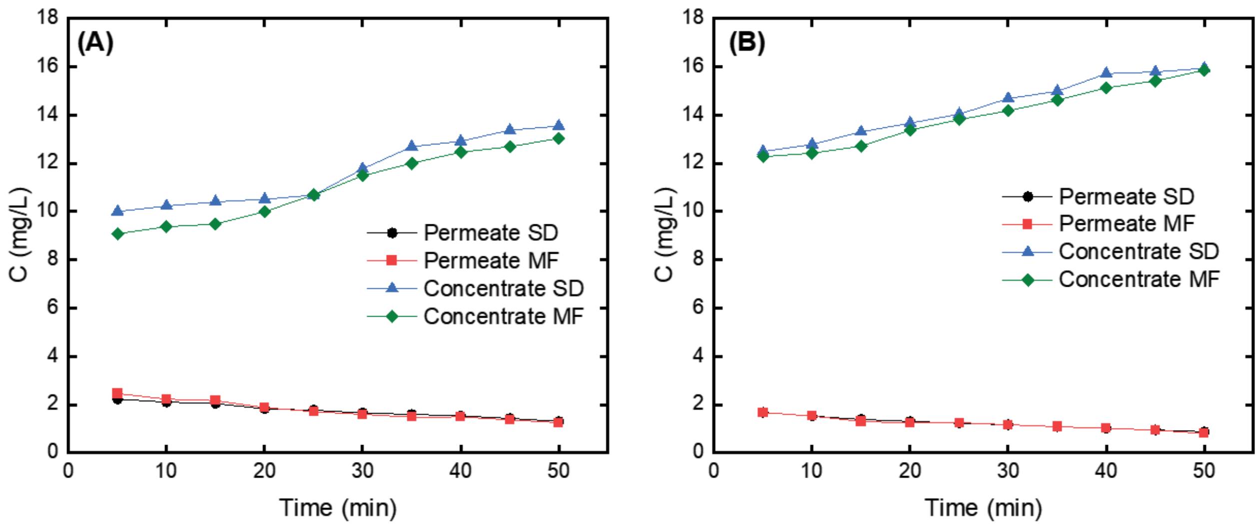

Fig. 11. Comparative study of the variation of dye concentration with time between MF and SuperPro Designer: (A) TMP = 0.9 bar and (B) TMP = 1.7 bar.

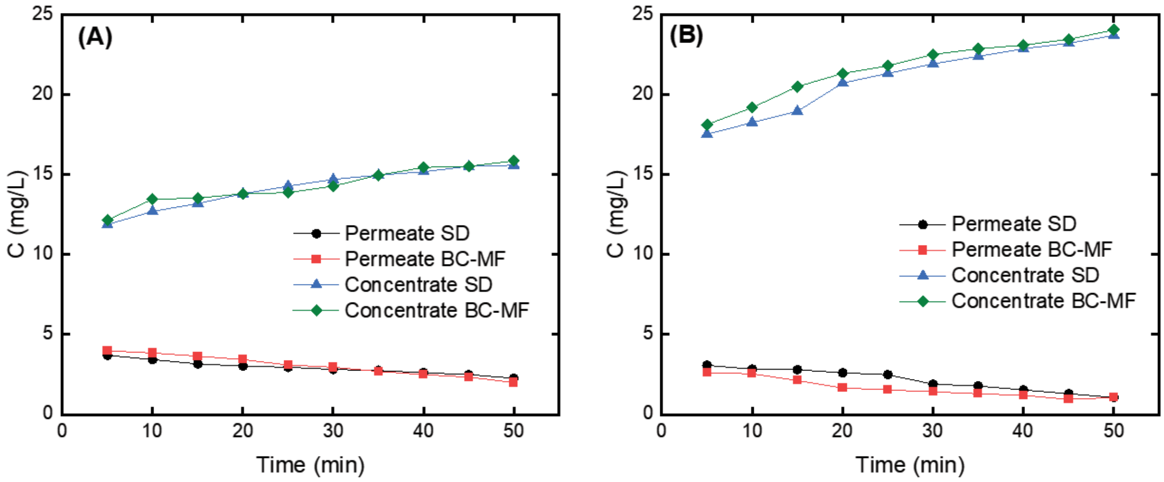

Fig. 12. Comparative study of the variation of dye concentration with time between BC-MF and SuperPro Designer: (a) TMP = 0.9 bar and (b) TMP = 1.7 bar.

the combined process (BC-MF) results at pH of 3 and a TMP of 1.7 bar demonstrate better performance compared to the other experimental conditions.

3.4. Comparative study

3.4.1. Microfiltration and bio-coagulation–microfiltration

Comparative studies between MF and BC-MF are presented in Fig. 10. Experiments were conducted at different pH values with a constant TMP of 1.7 bar. Discoloration rates ranged from 90% to 94% for the MF process, and from 70% to 96% for the BC-MF process. The removal efficiency of Terasil Red by MF in the hybrid process highly depends on the pH of the synthetic solution.

It is also observed that acidic pH in the hybrid process enhances dye removal efficiency. However, microfiltration gives better results than the combined process in an alkaline medium, which can be explained by the dye precipitation within this pH range. Therefore, it can be concluded that, for Terasil Red, simple microfiltration suffices to achieve excellent results for synthetic solution.

3.4.2. Simulation and experimental results

Simulation results obtained by SuperPro Designer software were compared to the experimental results for MF and BC-MF process. The operating conditions were consistent in both cases, with TMP of 0.9 and 1.7 bar (with pH = 3 and C_0 = 10 mg/L). Figs. 11 and 12 show that the experimental results are in good agreement with those obtained by simulation for both processes (MF and BC-MF) for concentrate and retentate. This indicates that the SuperPro Designer can be considered a very reliable tool for simulating membrane separations.

4. Conclusion

To improve the removal of Terasil Red dye from a synthetic solution, a combination of two processes, namely bio-coagulation (BC) and microfiltration (MF), was employed, knowing that coagulation aids in destabilizing dye particles in the solution, making them easier for MF to retain.

The BC process was conducted using acorn leaves as a natural coagulant, both in powdered form and as a solution extracted with NaOH. The results show the process's strong dependence on solution pH and coagulant dose.

The active components extracted with NaOH presented greater efficiency, especially at a coagulant dose of 10 mL and a pH of 3, which was identified as the optimal condition. Furthermore, it was observed that under basic pH conditions, dye particles precipitated without the need for additional bio-coagulant.

It was found that using MF employing a ceramic membrane composed of kaolin from the wilaya of Mila (Algeria) and zirconium oxide, yielded very satisfactory results for Terasil Red, achieving over 90% removal of dye concentration and turbidity. While the BC-MF hybrid process slightly improves the dye removal efficiency under acid pH conditions (achieving 96.2% and 98.77% removal for dye concentration and turbidity, respectively, at pH 3 and TMP of 1.7 bar), it is essential to note that transmembrane pressure significantly influenced the permeate flux, following Darcy's law, with the optimal performance observed at 1.7 bar.

The comparative study between the experimental and calculated results using SuperPro Designer software revealed a perfect similarity in the permeate and concentrate fluxes for both MF and BC-MF cases.

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