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Study and optimization of the ultrasound-enhanced cleaning of an ultrafiltration ceramic membrane through a combined experimental–statistical approach

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ABSTRACT

Membrane fouling is one of the main drawbacks of ultrafiltration technology during the treatment of dye-containing effluents. Therefore, the optimization of the membrane cleaning procedure is essential to improve the overall efficiency. In this work, a study of the factors affecting the ultrasound-assisted cleaning of an ultrafiltration ceramic membrane fouled by dye particles was carried out. The effect of transmembrane pressure (0.5, 1.5, 2.5 bar), cross-flow velocity (1, 2, 3 m s^{-1}), ultrasound power level (40%, 70%, 100%) and ultrasound frequency mode (37, 80 kHz and mixed wave) on the cleaning efficiency was evaluated. The lowest frequency showed better results, although the best cleaning performance was obtained using the mixed wave mode.

A Box–Behnken Design was used to find the optimal conditions for the cleaning procedure through a response surface study. The optimal operating conditions leading to the maximum cleaning efficiency predicted (32.19%) were found to be 1.1 bar, 3 m s^{-1} and 100% of power level.

Finally, the optimized response was compared to the efficiency of a chemical cleaning with NaOH solution, with and without the use of ultrasound. By using NaOH, cleaning efficiency nearly triples, and it improves up to 25% by adding ultrasound.

1. Introduction

Membrane filtration is currently applied within a wide variety of fields: dairy and food technology [1,2], pharmaceutical industry [3], and waste effluents treatment from pulp and paper industry [4] or from textile applications [5] among others. Moreover, membrane filtration is attempting to be introduced into a wide diversity of new other applications [6]. The relatively rapid expansion and increasing popularity of this technology may be attributed to its considerable separation efficiency, lower production costs in comparison with other treatment systems and versatility, among others. Additionally, membrane technologies show unique separation properties among which their simple operation, no phase change and no need for any chemical addition in order to achieve the expected separation, may be emphasized. These properties made them a suitable replacement for other separation and treatment technologies while meeting their separation objectives [7,8]. In particular, ultrafiltration (UF) is a promising separation tool in a wide diversity of industrial processes covering fractionation, concentration and elimination of macromolecular species as well as for the elimination of macrosolutes from diverse industrial effluents [9,10]. One of the applications where UF has been reported as a feasible and economically more favorable technology involves dye effluent treatment [11].

Nevertheless, a significant drawback avoiding an even major expansion of membrane technologies use is related to membrane fouling. This membrane fouling entails a decline in system performance which may be mainly observed either in the permeate flux or membrane selectivity. Fouling extent is dependent on a great number of parameters related to membrane properties, feed solution and hydrodynamics of the process [12,13]. In ultrafiltration, the reduction in permeate flux is a consequence of the increased flow resistance which may be mainly attributed to the build-up of a concentration polarization layer and to membrane fouling [12]. The concentration polarization layer is produced as a consequence of the accumulation of rejected solutes within a thin boundary layer near the membrane surface [14]. This concentration polarization phenomenon has also been observed in the particular case of the filtration of dye-containing effluents [15,16]. Regarding the fouling mechanisms of the ultrafiltration membranes three factors are considered as main responsible i.e. membrane pore blocking, formation of a cake layer on the membrane surface and the adsorption of the fouling materials on the membrane surface or in the pore walls [6,17]. Initially, a rapid and sharp flux decline occurs which may be attributed to the initial internal

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pore blocking by the retained molecules. Further flux decline may be associated to the formation and subsequent growth of the gel and cake layer by the deposition and accumulation of rejected particles [13,18].

As a consequence of the related decrease in process efficiency, significant research efforts have been made involving different approaches in order to limit or reduce membrane fouling and especially concerning membrane cleaning, which may be found within literature. Currently, the most common cleaning technologies include hydraulic, mechanical, chemical, and electrical cleaning methods. The selection of the most suitable cleaning method would depend on different factors such as type of foulants or membrane characteristics [18,19]. In spite of the benefits of each method, diverse disadvantages and limitations may be also found among them. In this way, either the continuous research on mitigation of these disadvantages as well as more innovative cleaning methods become necessary. Recently, novel techniques such as electrical field and ultrasound (US) have proven its suitability for membrane filtration and membrane cleaning [20–23].

In particular, US-enhanced membrane cleaning has been reported to be a very effective cleaning method because of its particular characteristics [20,24]. Ultrasound has widely been used for cleaning different materials and surfaces due to the primary phenomenon of cavitation and strong convective currents, known as acoustic streaming, combined with the effect of microstreaming, microstreamers, microjets, shock waves and heating [12,21,22,25]. Ultrasound wave propagates via an alternating adiabatic compression and rarefaction (decompression) cycle waves induced in the molecules of the medium. In some cases, at a sufficiently high power, the rarefaction cycle may exceed the attractive forces of the molecules of the medium (liquid), which would involve a negative net pressure applied to the medium and consequently causing cavitation bubbles formation. When these cavitation bubbles collapse due to the compression cycle, they may release sufficient energy to overcome the foulant–membrane interactions [26], removing portions of the fouling layer from the membrane surface, and/or preventing the deposition of particles that lead to membrane fouling [25]. Furthermore, the high temperatures and pressures generated at the collapsing spots are also a source of $^{\cdot}OH$ radicals, which may produce the oxidation of organic pollutants and molecules at the gas–liquid interface. This sonochemical degradation of organic pollutants through chemical oxidation has been already proven as a feasible method for the treatment of dye-containing effluents [27,28].

The efficiency of the US-enhanced cleaning is dependent on a significant number of factors including frequency, power intensity, feed water qualities, membrane characteristics and process hydrodynamics such as transmembrane pressure (TMP) and cross-flow velocity (CFV) among others [12,29].

When such a significant number of factors may influence the process performance, the effect of the different influential operating variables may be dependent on each other and thus, the study of each variable at a time would not be either accurate or appropriate. Consequently, the effects of the multiple variables together with their interactions may be better studied through the response surface methodology (RSM) [30,31].

RSM is a collection of statistical and mathematical techniques useful for developing, improving, and optimizing processes in which a response of interest is influenced by several variables and the objective is to optimize this response [32]. Extensive literature regarding the optimization of different processes by means of RSM is available [33–35].

A significant number of investigations regarding US-assisted membrane cleaning may be found in the literature. However, the use of this cleaning method applied to tubular ceramic membranes previously fouled with dye particles is not as thoroughly studied and a lesser number of studies can be found. In this way, the effectiveness of US on enhancing membrane cleaning and permeability recovery is evaluated in this work. Furthermore, the interest of this study is increased by coupling experimental and statistical approaches. To that end, the main objective of this study is related to determine the influence of different operating parameters such as TMP, CFV and both US power level and frequency on the cleaning process of an ultrafiltration membrane used for the removal of a reactive dye. Additionally, the optimized response in terms of cleaning efficiency is calculated by obtaining a mathematical model using the RSM approach which allows describing the cleaning process. Finally, the optimized response is compared with the performance of a chemical cleaning procedure using a NaOH solution with and without the use of ultrasound.

2. Materials and methods

2.1. Experimental set-up

Fig. 1 is a schematic representation of the experimental set-up and the ultrasonic cleaning unit used during the study. The temperature- and pressure-controlled cross-flow ultrafiltration system, has been described elsewhere [36]. The temperature was controlled with an accuracy of ±1 °C. The variable speed plunger pump (3CP1140, Cat pumps) was used to pump the feed solution through the system, allowing the cross-flow velocity to be regulated. Besides, the TMP was adjusted by means of the needle valve after the membrane module. The membrane was placed inside a tubular stainless steel holder (TAMI Industries, Nyons, France).

In those cases in which NaOH solution was fed during the cleaning procedure, the bypass was used in order to prevent the filter from being damaged by the high pH conditions. The permeate flux was continuously determined gravimetrically using an electronic balance (KB120 2N, Kern®), connected to a computer which acquired data by means of a data acquisition software (Balance Connection 4.0, Kern®). Data were recorded at 1 min interval. During the experiments both the retentate and permeate were recirculated to the feed tank after flux measurements.

The ultrasonic cleaning was performed in a US bath (S70H, Elmasonic) made of cavitation-proof stainless steel. It was equipped with sandwich-type performance transducer systems of 220 W output (adjustable between 40% and 100% of the full nominal power level) for emitting ultrasound at two ultrasonic frequencies, switchable: 37 and 80 kHz. The US unit had also the possibility to operate at a mixed wave mode. When this mode was selected, ultrasonic frequency switched automatically between 37 and 80 kHz at 30 s intervals. The inner dimensions of the US bath were 505 mm $\times$ 137 mm $\times$ 100 mm and it was filled with 4 L of deionized water. The membrane module was placed in the holder and immersed in the liquid bulk, 40 mm away from the bath bottom, where the transducers were located. This distance allowed the holder to be totally immersed in the liquid but without touching the bottom of the tank, since this might lead to damages to the unit. The membrane holder was immersed throughout the entire experimental period but sonication only turned on when necessary. Additionally, the US bath was fitted with the sweep function device for an optimized sound field distribution within the bath.

2.2. Materials

An INSIDE CéRAM® multichannel tubular ceramic membrane manufactured by TAMI Industries (Nyons, France) was used in all the experiments. The membrane consisted of a ZrO_2–TiO_2 layer on a TiO_2 support with a molecular weight cut-off of 150 kDa. It