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Study of membrane cleaning with and without ultrasounds application after fouling with three model dairy solutions

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ABSTRACT

The aim of this study was to investigate the behavior of two ultrafiltration (UF) membranes after their fouling with different fouling solutions and cleaning with a surfactant, including the application of ultrasounds (US). Thus, two UF membranes (UH030 and UP005) were fouled with three different whey model solutions that consisted of bovine serum albumin (BSA) with a concentration of 1% (w/w), BSA (1%, w/w) plus $CaCl_2$ with a concentration of 0.17% (w/w) and whey solution (Renylat 45) with a concentration of 2.22% (w/w). Chemical cleaning was carried out with P3 Ultrasil 115 solution at temperatures between 25 °C and 45 °C and concentrations in the range between 0.5% (v/v) and 0.9% (v/v). US were applied in some of the tests at a frequency of 20 kHz and nominal power of 300 W. The results demonstrated that US cleaning was effective to enhance the permeability recovery, although this enhancement was only up to 9%. Concerning fouling, results from resistances calculations showed that for UH030 membrane prevails the reversible fouling whereas for UP005 membrane predominate irreversible fouling.

1. Introduction

Membrane technologies are frequently used for many industrial applications (Delaunay et al., 2008). Especially, in the dairy industry, UF membranes are commonly used for processes such as milk dehydration and whey concentration (Kazemimoghadam and Mohammadi, 2007).

UF processes present many advantages over conventional processes such as the reduction of the cost of separation operations (due to low-energy requirement) and the increasing of the product yield. However, the main concern of UF processes is membrane fouling, since retained particles can accumulate on the membrane surface and inside the pores (Muthukumaran et al., 2004). As a result, an important reduction in the separation efficiency by decreasing the permeate flux is produced. In membranes used in dairy industry, proteins and minerals (mainly calcium and phosphorous) are the two major contributors to membrane fouling (Rice et al., 2009). These compounds foul the membrane by adsorption onto the membrane surface and due to internal pore blockage.

For the restoration of the membrane initial permeability, it is necessary to clean the membranes. Thus, optimization of the cleaning processes for UF membranes (Yee et al., 2009) is of paramount importance. Most works about membrane cleaning are focused on conventional cleaning methods using chemicals such as acids, bases, chelating agents or surfactants. However, these methods often require large amounts of chemicals and make the membrane cleaning process more expensive. Besides, the excessive use of these chemicals can damage the membrane material and decrease the lifetime

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Nomenclature

UF	ultrafiltration
US	ultrasounds
PS	polysulfone
BSA	bovine serum albumin
TMP	transmembrane pressure
K	membrane permeability, $L\,m^{-2}\,h^{-1}\,bar^{-1}$
ΔP	transmembrane pressure applied, bar
C_p	permeate protein concentration, $mg\,mL^{-1}$
C_f	feed protein concentration, $mg\,mL^{-1}$
μ	irreversible resistance, m^{-1}
J_w	permeate flux with distilled water, $L\,m^{-2}\,h^{-1}$
J_t	permeate flux at the end of the fouling step, $L\,m^{-2}\,h^{-1}$
J_{wr1}	permeate flux after the first rinsing step; $L\,m^{-2}\,h^{-1}$
J_{wc}	permeate flux after the second rinsing step, $L\,m^{-2}\,h^{-2}$
CE	cleaning efficiency without US, %
$CE_{test\ with\ US}$	cleaning efficiency applying US, %
CE_{US}	cleaning efficiency improvement with US, %
R_m	membrane initial hydraulic resistance, m^{-1}
R_t	membrane resistance after fouling step, m^{-1}
R_c	membrane resistance after the second rinsing step, m^{-1}
R_{irrev}	irreversible resistance, m^{-1}
R_{rev}	reversible resistance, m^{-1}

of the membrane. Thus, it is important to study and to develop other cleaning methods that reduce the use of these products.

In the last years the use of US in the membrane cleaning process has been taken into account (Muthukumaran et al., 2004). In the membrane processes field, US have been applied both in the cleaning membrane procedures (like in this study) to remove the fouling and in membrane filtration processes to prevent it (Kyllönen et al., 2006; Secondes et al., 2014; Muthukumaran et al., 2005b). Membrane cleaning with US uses high-frequency sound waves to create microbubbles that collapse, with a release of energy, which help to remove foulant from the membrane surface, since the interaction between foulant and membrane is weakened. In addition, the high pressures and temperatures generated with the collapsing of microbubbles are a source of -OH radicals, which may lead to the oxidation of organic pollutants and molecules at the gas-liquid interface (Naddeo et al., 2015).

Several authors have studied the use of US in membrane applications. As reported by Muthukumaran et al. (2005b) and Muthukumaran et al. (2007), US applications at low frequencies were effective both for the filtration and the cleaning of UF membranes treating whey. Popović et al. (2010) published that US membrane cleaning was effective for ceramic membranes used for whey ultrafiltration. Muthukumaran et al. (2004) studied the effect of US in combination with surfactant solution for cleaning polysulfone flat sheet ultrafiltration membranes used for whey ultrafiltration. These authors reported that US cleaning were effective and independent of the sonication time. However, in these studies membrane modules were submerged in the US bath instead of applying US in the cleaning solution like in this study.

The main purpose of this work was to study the protein separation effectiveness and membrane fouling of two ultrafiltration (UF) membranes fouled by model proteins solutions (BSA 1%, w/w and 0.06%, w/w in calcium dosed as $CaCl_2$) and commercial whey. In addition, it was evaluated the membrane cleaning, under several cleaning operation conditions, including the application of ultrasounds (US). For this, US were applied in the chemical cleaning solution instead of submerging the membrane module in the US bath, what has been reported in the literature by most authors until now (Kyllönen et al., 2006; Latt and Kobayashi, 2006; Li et al., 2002; Muthukumaran et al., 2005a). Thus, in this study, membrane module was placed outside the US bath, US were generated in the chemical cleaning solution that was recirculated by the membrane system.

As a cleaning agent a specific surfactant, P3 Ultrasil 115, has been tested. Surfactant solutions are common chemical cleaning agents used to restore the membrane initial flux. Particularly, P3 Ultrasil 115 is recommended to clean membranes used in the dairy industry and to remove organic foulants like proteins (Naim et al., 2012). In this work, the membrane flux recovery for various P3 Ultrasil 115 solutions at three temperatures and concentrations was evaluated. US were applied to enhance the flux recovery and to evaluate their effect under different cleaning conditions.

2. Materials and methods

2.1. *Ultrafiltration pilot plant*

A laboratory UF plant (Orelis Environnement SAS, Salindres, France) was used in the experiments. This laboratory plant includes a Rayflow flat sheet module also from Orelis, France. The membrane module has capacity for two flat sheet membranes of 100 cm^2 each one, operating by cross-flow filtration mode and working in series. The US equipment consisted of a US bath and US generator. The tank for the cleaning solution, where US were generated was a TSD-D 18 ultrasonic bath (TSD Machinery, Ultrasonidos JJ.VICEDO, Valencia, Spain) with total volume of 18 L. This tank was connected to US generator TSD RF 300 (TSD Machinery). Ultrasonic equipment consists of an ultrasound generator and special transducers. The produced ultrasonic waves propagate through the liquid medium (like the chemical cleaning solution), causing a succession of compression and expansion cycles. This sequence of events is called cavitation phenomenon and is the responsible to promoting cleaning. The collapse of the bubbles causes the breakup between the foulant and the membrane and removes the foulant from the membrane (Li et al., 2002; Muthukumaran et al., 2005a). A scheme of the UF plant is shown in Fig. 1.

2.2. *Membranes*

The organic UF membranes tested in the experiments were from Microdyn Nadir (Wiesbaden, Germany). Two types of membranes were used. One of them was a polyethersulfone (PES) membrane with a nominal molecular weight cut-off of 5 kDa (membrane UP005) and the other one was a hydrophilic polyethersulfone (PESH) membrane with a nominal molecular weight cut-off of 30 kDa (membrane UH030). Both membranes accept values of pH in the range of 0–14 and work until temperatures of 95 °C.