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A new way to apply ultrasound in cross-flow ultrafiltration: Application to colloidal suspensions

N. Hengl [a,*], Y. Jin [a], F. Pignon [a], S. Baup [a], R. Mollard [a], N. Gondrexon [a], A. Magnin [a], L. Michot [b], E. Paineau [c]

[a] *Laboratoire Rhéologie et Procédés, Université Joseph Fourier Grenoble I, Grenoble-INP, CNRS, UMR 5520, BP 53, 38041 Grenoble Cedex 9, France*
[b] *Laboratoire Environnement et Minéralurgie, CNRS-Université de Lorraine UMR 7569, 15 Avenue du Charmois, BP 40, 54501 Vandœuvre Cedex, France*
[c] *Laboratoire de Physique des Solides, CNRS-Université Paris XI UMR 8502, Bat. 510, 91405 Orsay Cedex, France*

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A new coupling of ultrasound device with membrane process has been developed in order to enhance cross-flow ultrafiltration of colloidal suspensions usually involved in several industrial applications included bio and agro industries, water and sludge treatment. In order to reduce mass transfer resistances induced by fouling and concentration polarization, which both are main limitations in membrane separation process continuous ultrasound is applied with the help of a vibrating blade (20 kHz) located in the feed channel all over the membrane surface (8 mm between membrane surface and the blade). Hydrodynamic aspects were also taking into account by the control of the rectangular geometry of the feed channel.

Three colloidal suspensions with different kinds of colloidal interaction (attractive, repulsive) were chosen to evaluate the effect of their physico-chemical properties on the filtration.

For a 90 W power (20.5 W cm^{-2}) and a continuous flow rate, permeation fluxes are increased for each studied colloidal suspension, without damaging the membrane. The results show that the flux increase depends on the initial structural properties of filtered dispersion in terms of colloidal interaction and spatial organizations.

For instance, a Montmorillonite Wyoming–Na clay suspension was filtered at 1.5×10^5 Pa transmembrane pressure. Its permeation flux is increased by a factor 7.1, from 13.6 L m^{-2} h^{-1} without ultrasound to 97 L m^{-2} h^{-1} with ultrasound.

1. Introduction

Membrane filtration is one of the main technologies used to concentrate, purify or remove solute from a solution. Cross-flow ultrafiltration, which belongs to pressure driven membrane processes, is widely applied in many industrial fields such as food industry (milk concentration [1], fruit juice production [2,3], beverage process [4]), or waste water treatment [5,6]. Although it presents numerous advantages (large active area per volume unit, easy to operate, many configurations, modularity), this technology also bears some serious limitations associated to fouling [7] and concentration polarization [8]. Fouling is generally associated to an irreversible phenomenon where physical, chemical and biological effects can modify the membrane permeability [9,10]. In contrast, concentration polarization is considered to be reversible and can be controlled by increasing shear rate [8,10] or changing hydrodynamic conditions.

Several techniques such as backflushing [11–13], pulsing mode [14], vibration [15], air sparging [16], chemical cleaning have been developed to reduce the appearance of these unfavorable phenomena. The effectiveness of these techniques has been proved, but all of them present drawbacks. For instance backflushing or air sparging imposes stopping the process to remove fouling at the membrane surface (discontinuous mode and time consuming). Furthermore, some of these techniques are difficult to implement on a large scale, often need to be repeated for improved efficiency, induce post treatment or can degrade the membrane quality. These two latter drawbacks are typically associated to chemical cleaning.

Within this framework, applying ultrasound appears as a relevant solution for reducing fouling and concentration polarization upon membrane filtration [17]. The purpose of the present work is to propose a new experimental device coupling cross-flow ultrafiltration and low-frequency ultrasound applied continuously for long operating times (few hours) without damaging the membrane. This challenge has been reached by the development of a vibrating blade allowing controlling and applying ultrasound in the feed channel directly to the treated suspension all along the membrane.

* Corresponding author. Tel.: +33 668204428.
E-mail address: hengl@ujf-grenoble.fr (N. Hengl).

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Furthermore, this membrane cell has been specially designed with a rectangular channel section to control the hydrodynamics.

Ultrasound waves are propagated via series of compression and decompression through the whole irradiated solution. Under sufficient power, this can lead to the formation of cavitation bubbles. Other mechanical combined effects such as acoustic streaming, micro-streaming, micro-streamers, and micro-jets can also be associated. In aqueous solutions, collapsing cavitation bubble yields significant mechanical and chemical effects that generate either motion in both the fluid [18] and close to the membrane surface, or free radicals available for chemical reactions [19]. In previous work, several membrane processes involving ultrasound device have been used to reduce fouling or to improve transmembrane fluxes [20–23]. All those studies have shown a positive effect of low frequency ultrasound (between 20 kHz and 50 kHz) for reducing fouling as highlighted by Kyllönen et al. [24] and have also revealed a moderate enhancement of permeation flux through the membrane. Nevertheless, in all these previous studies, the ultrasound device used was a simple ultrasonic bath in which a membrane module is immersed. Accordingly, ultrasound waves have to be transmitted from the transducer to the membrane surface through the membrane module, which results in significant energy loss during irradiation. Furthermore, such a configuration does not allow a precise control of the applied ultrasound. To improve process efficiency, ultrasound should be applied where fouling or concentration polarization occurs, in other words close to the membrane surface. Along those lines, Chen et al. [25] have developed an ultrafiltration process with ceramic membranes with an immersed ultrasound device using colloidal silica model particles. They show that an improvement of 100% of the transmembrane flux can be achieved, and that changing the distance between the membrane surface and the transducer from 1.7 cm to 3.5 cm leads to a reduction of the permeation flux too. Simon et al. [26] studied the effect of low frequency ultrasound device plunged in Amicon dead-end ultrafiltration cell for the filtration of polymer solution. Using polymeric polysulfone membranes, they observed an increase of the permeation flux, without damaging the membranes, for both continuous and pulsed modes. Liu et al. [27] used the same device and found similar results for the dead end microfiltration of grape pomace extract. Mirzaie and Mohammadi [28] proposed to adapt an Amicon membrane cell (based on Simon et al. set up) in order to pass from a dead end configuration to a cross-flow one for milk microfiltration with an average irradiation time of 30 min. Even if different probe tip diameters are used (2, 3, 6 and 12.7 mm), the membrane (40 mm diameter) is not fully irradiated. It is important to notice that the hydrodynamics at the vicinity of the membrane is not totally controlled. Kyllönen et al. [24] used a more achieved device in the case of the filtration of wastewaters: a rectangular cross-flow channel is irradiated by ultrasound, located at 1 cm of the membrane surface. Nevertheless in this device, ultrasound is used for a few seconds in a pulsed mode.

In the present work, the effects of 20 kHz ultrasound, transmembrane pressure (TMP), and properties of filtered solutions on permeation flux were studied during ultrafiltration process. Optical Microscopy (OM) by incident and transmitted light and Scanning Electron Microscopy (SEM) were also performed to understand the effect of ultrasound waves on the structure of the polymeric membrane.

2. Materials and methods

2.1. Experimental setup

A cross-flow ultrafiltration cell coupled with an "*in situ*" ultrasonic system is the base of the experimental setup (Fig. 1). The initial ultrafiltration cell [29,30] has been adapted to receive the ultrasonic system as shown in Fig. 1b. The ultrasonic device is a metallic blade specially shaped (4 mm × 110 mm × 50 mm) connected to an ultrasonic transducer linked to a generator (Sonifier 450, Branson). This blade, which is the upper side of the membrane module, is immersed directly in the feed channel of the polycarbonate membrane cell. The distance from the membrane surface to the bottom of the blade is 8 mm. This configuration will help to improve the propagation of ultrasound through the system. The frequency used for the experimental study is 20 kHz and a continuous ultrasound irradiation was chosen.

The feed solution is stored in a 5 L high pressure vessel from Millipore (Fig. 1a). It flows in the upper compartment with a rectangular channel section of 4 mm × 8 mm (width × height) and a length of 120 mm, allowing a good control of the hydrodynamics. A volumetric pump (Mono pump LF series, Axflow) imposes a constant cross-flow flux measured by a magnetic flowmeter (Optiflux 6300C, Krohne). The feed solution circulates in a close loop. Pressure is applied to the rig via purified compressed air, and is measured at both the input and output of the filtration cell using pressure gauges (FP 110 FGP Sensors & Instrument). Two temperature sensors (YC-747D with k thermocouples) are also placed at the input and the output of the cross-flow cell to control the inlet temperature and estimate the raise in temperature due to ultrasound. The input temperature is kept at 25 °C thanks to a thermostatic bath (Thermo Scientific SC 150 A25, Haake). Finally, the permeate flux J is continuously monitored and its mass is registered every 10 s with a 0.001 g of accuracy (Precisa 400M) which allowed getting a mean value of permeation flux each 10 s as described [30]. Each experiment lasts at least 1 h.

The transmembrane pressure (TMP) varied from 0 to 1.5 bar when the permeate compartment is at atmospheric pressure. Cross-flow rate was maintained constant and equal to the value of 0.45 L min^{-1} giving a 0.23 m s^{-1} mean velocity in the feed channel and a mean shear rate of 175 s^{-1}.

For all the experiments performed with ultrasound, the energy supply was maintained constant at a fixed value of 90 W, corresponding to an output power per unit area of the transducer surface of 20.5 W cm^{-2}. This value represents the total energy used by the generator system and not the intrinsic ultrasound power transferred by the vibration of the metallic blade to the solution.

2.2. Experimental procedure

The permeate flux can be expressed as the Darcy's law according to resistance in series model (Eq. (1)):

$$J = \frac{1}{\mu \cdot R_{tot}} \cdot \Delta P \tag{1}$$

with J: the permeate flux ($m^3\, m^{-2}\, s^{-1}$); μ: the dynamic viscosity of the solvent (Pa s); ΔP: the transmembrane pressure (TMP) (Pa), $\Delta P = \left(\frac{P_{input}+P_{output}}{2}\right) - P_{atm}$; R_{tot}: the total resistance (m^{-1}).

Espinasse et al. [31] have proposed an experimental approach to identify concentration polarization phenomena and membrane fouling during membrane filtration. This relevant procedure that alternates positive and negative pressure changes and evaluates the variation of steady state fluxes for a same pressure [32] was systematically used in the present study. The objective is to assess the reversibility or irreversibility of these phenomena in order to characterize the efficiency of ultrasound and their ability to control the accumulation of particles at the membrane surface.