Available online at www.sciencedirect.com
ScienceDirect
Separation and Purification Technology 56 (2007) 133–142
Separation and Purification Technology
www.elsevier.com/locate/seppur

Ultrasonic enhancement of membrane-based deoxygenation and simultaneous influence on polymeric hollow fiber membrane

Liying Liu, Zhongwei Ding *, Lijing Chang, Runyu Ma, Zurong Yang

College of Chemical Engineering, Beijing University of Chemical Technology, Beijing 100029, China

Received 30 December 2006; received in revised form 29 January 2007; accepted 29 January 2007

Abstract

This study made an attempt at applying ultrasonic technique to membrane-based deoxygenation process. The mass transfer performance of immersed hollow fiber membrane modules in removing dissolved oxygen from water with or without ultrasonic irradiation was evaluated. Moreover, to understand the influence of ultrasonic irradiation on the membrane, the surface structure of the irradiated membrane was observed by field emission scanning electron microscopy (FESEM). It was found that the mass transfer was greatly improved by ultrasound stimulation in both PP and PVDF membrane module. The mass transfer coefficient increased significantly with the increase of ultrasonic intensity, and up to 2.0 of the enhancement factor was obtained. In addition, the mass transfer improvement depended on the operating vacuum degree as well as the altitude of the immersed membrane module in the reservoir, due to the local difference in ultrasonic intensity. The microscopic images shed light on the changes of membrane structure under ultrasonic treatment. Either PP or PVDF membrane was affected by ultrasonic irradiation, the surface of PP membrane was more easily transformed and damaged than PVDF. In conclusion, ultrasonic irradiation can be applied to membrane-based deoxygenation for the enhancement of mass transfer, but cautions should be taken to choose proper membrane material, ultrasonic intensity and irradiated duration to avoid membrane damage.

Keywords: Ultrasound; Hollow fiber membrane; Mass transfer enhancement; Deoxygenation

1. Introduction

Ultrasonic wave is referred to the acoustic wave with the frequency between 20 kHz and 10 MHz. Several concomitant physical effects, such as the mechanics, thermotics and cavatition effect, present themselves during the propagation of ultrasonic wave in various media, and these effects have been recognized to be beneficial to many physical and chemical processes. Chemical engineering may be among the most that the ultrasonic enhancement technique can be applied to. Ultrasonic enhancement on mass transfer has been realized in such unit operations as leaching [1], extraction [2,3], adsorption and desorption [4,5]. In addition, in the fields of heat transfer [6–8], chemical reaction [9–11], fine particles preparation [12,13] and waster water treatment [14,15], the concerning processes also benefit from ultrasonic irradiation.

Another potential and important application of ultrasonic effect in modern industry is to improve the permeation of membrane filtration, especially for microfiltration (MF) and ultrafiltration (UF). The two processes often suffer from concentration polarization and subsequent fouling, which cause serious decrease of transmembrane flux and membrane lifetime. Conducting MF and UF under ultrasonic irradiation has been shown to be effective in solving these problems. Herein, ultrasound wave is made use of in two ways: on-line ultrasound irradiation during filtration, or being coupled to chemical/water cleaning of fouled membranes. In the cross-flow UF of dextran, Kobayashi and co-workers [16,17] found that ultrasonic irradiation was effective on the enhancement of permeate flux, possibly as the results of the stimulation of mass transfer across the concentrated dextran layer close to the membrane surface and the decline of the osmotic pressure caused by the decrease of solute concentration at the membrane surface. The enhancement could be improved by using higher ultrasonic intensity and lower ultrasonic frequency, and also depended on feed property (MWs of dextran) and the propagation direction of ultrasound. Juang and Lin [18] examined the flux recovery in dead-end UF with ultrasound, and found that ultrasound was a promising method for the recovery of transmembrane flux, particularly for the solution with less fouling potential (70–80% flux recovery was

* Corresponding author. Tel.: +86 10 64454601; fax: +86 10 64436781.
E-mail address: dingzw@mail.buct.edu.cn (Z. Ding).

1383-5866/$ – see front matter © 2007 Elsevier B.V. All rights reserved.
doi:10.1016/j.seppur.2007.01.023

Nomenclature

A	membrane area (m^2)
C	dissolved oxygen concentration in the water (ppm)
C_0	speed of sound in the water (m/s)
C^0	initial concentration of dissolved oxygen in the water (ppm)
C^*	aqueous phase oxygen concentration in equilibrium with the gas phase (ppm)
f	ultrasonic frequency (kHz)
I	ultrasonic intensity (W/cm^2)
K	overall mass transfer coefficient (m/s)
M	free-field sensitivity of the hydrophone (μV/Pa)
p	vacuum degree or acoustic pressure (Pa)
P	ultrasonic power (W)
t	operating time (min)
U	peak value of outlet voltage of the hydrophone (V)
V	volume of water in the reservoir (m^3)
Z	distance to the bottom of the reservoir (cm)
Greek letter	
ρ	density of water (kg/m^3)

obtained for Cu-PEI solution). Simon et al. [19] compared classical stirred and ultrasonically assisted dead-end UF. They thought the effect of ultrasound was similar to that brought about by a classical mechanical stirrer. This agitation of ultrasound results from the convective currents during the propagation of acoustic waves as well as the physical effects of cavitation, greatly mitigating the concentration polarization. Kobayashi et al. [20] used ultrasonic technique in the water cleaning of fouled UF and MF membrane, and compared the results with that obtained without ultrasound. It was found that cleaning with ultrasound was more effective for flux recovery, especially at the lower frequency. Muthukumaran et al. [21] studied the optimization of ultrasonic cleaning procedures for dairy fouled UF membranes, in which factors affecting the process were examined. The results showed that ultrasonic cleaning improved the cleaning efficiency under all experimental conditions, and the enhancement was more significant in the absence of surfactant, but was less influenced by temperature and transmembrane pressure.

Besides membrane filtration, ultrasound is also applied to other membrane process. Zhu et al. [22,23] introduced ultrasonic technique to an air gap membrane distillation system. The flux enhancement up to 200% was obtained. It was also found that the enhancement can be improved by increasing ultrasonic intensity, decreasing ultrasonic frequency or decreasing the solution temperature.

While ultrasonic enhancement has been applied to membrane process, it is observed that ultrasound irradiation would damage the membrane, a serious problem in membrane lifetime. The extent of the structural damages depends on the mechanical resistance of the polymer to ultrasound and the initial membrane structure. Masselin et al. [24] observed the influence of ultrasound on different polymeric membrane materials at a frequency of 47 kHz, and found that the overall surface of polyethersulphone (PES) membrane was transformed by the irradiation, but the polyvinylidenefluoride (PVDF) and hydrophilic polyacrylonitrile (PAN) membranes showed no observable change. Wang et al. [25] conducted similar work, in which four different polymeric membranes (PES, nylon 6 (N6), the mixed ester of cellulose nitrate with cellulose acetate (CN–CA) and PVDF) were examined under ultrasonic irradiation of low intensity, and found that only the PVDF membrane presented no significant change in the measured parameters within the duration of ultrasound, whereas those of others were noticeably affected. Juang and Lin [18] observed slight damage on a regenerated cellulose membrane in an ultrasonic field generated by a horn transducer with the tip distance of 10 mm and the power of 80 W. However, with the tip distance longer than 20 mm, the membrane could be used under 240 W transducer power without any detected damage in membrane structure.

In previous studies, the ultrasonic enhancement was mainly applied to the filtration process of flat sheet membrane, few works were reported in other membrane process and involved hollow fiber membranes. As a result, the performance of hollow fiber membrane contactor in ultrasonic field is yet to be understood. This study investigates the ultrasonic enhancement on the mass transfer in membrane-based deoxygenation, and the subsequent influence on the structure of hollow fiber membrane. The mass transfer performance of immersed hollow fiber membrane modules in removing dissolved oxygen from water with or without ultrasound irradiation is evaluated. For understanding the influence of ultrasonic irradiation on the membrane structure, field emission scanning electron microscopy (FESEM) was used to observe the surface of the irradiated membrane. Two kinds of polymeric hollow fiber membranes, polypropylene (PP) and PVDF, were employed in both mass transfer and membrane examination experiments.

2. Theory

2.1. Mechanisms of ultrasonic enhancement

Ultrasound wave can propagate through whichever medium of solid, liquid or gas in the form of energy, and interact with these media in mechanics, thermotics and cavitation effects. Cavitation is usually considered playing the most important role in ultrasonic enhancement of membrane process for liquid–liquid and liquid–solid system. Cavitation phenomenon is referred to the formation, growth, compression, and sudden collapse of micro bubbles (cavitation bubbles) in liquids. The collapses of the micro bubbles bring significant mechanical and thermal effects, generating temperature and pressure that is above 5000 K and 500 atm (50 MPa) in the bubbles [26], and associated with which powerful shock wave and microstreaming with the speed of about 110 m/s are created. The implosions occur within very short lifetime, less than 0.1 μs [27,28]. These effects are considered beneficial to the mass transfer of membrane process. Firstly, the macroflow of liquid resulted from the