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Desalination 204 (2007) 255–264

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DESALINATION

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## Combining air sparging and the use of a static mixer in cross-flow ultrafiltration of oil/water emulsion

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Received 30 January 2006; accepted 20 February 2006

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### Abstract

The simultaneous use of a static mixer and air sparging technique has been investigated during cross-flow ultrafiltration of oil/water emulsion. It is found that the use of a static mixer leads to a considerable increase in the permeate flux. However, the presence of a static mixer reduced hold-up in the feed channel leading to a high pressure drop along the length of the membrane and therefore the increase in energy consumption. Combining air sparging and the use of a static mixer demonstrated that a high permeate flux can be obtained at a relatively low energy consumption. The results suggest that the simultaneous use of these techniques could be interesting in membrane processes which already use gas injection, such as aerobic membrane bioreactors.

*Keywords:* Cross-flow ultrafiltration; Oil/water emulsion; Static mixer; Air sparging

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### 1. Introduction

Ultrafiltration represents a highly attractive and promising technique for treating oily wastewaters, especially stable oil/water emulsions which

require more sophisticated treatment to meet more and more rigorous wastewater effluent standards. The use of an ultrafiltration membrane to separate stable oil/water emulsion has been reported in many studies [1–7]. High water quality and low energy costs provide to ultrafiltration a decisive

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*Presented at EuroMed 2006 conference on Desalination Strategies in South Mediterranean Countries: Cooperation between Mediterranean Countries of Europe and the Southern Rim of the Mediterranean. Sponsored by the European Desalination Society and the University of Montpellier II, Montpellier, France, 21–25 May 2006.*

advantage over conventional treatment of stable oil/water emulsions. However, this process has several limitations, foremost among them the low permeate fluxes due to fouling. Oil/water emulsions induce three kinds of fouling mechanisms: oil drop deposit, concentration polarisation and adsorption of dissolved organic compounds [8].

The reduction of concentration polarisation and membrane fouling has been the focus of many studies. Several methods have been developed in various applications of ultrafiltration: the use of higher cross-flow velocities, pulsed flow [9], production of centrifugal instabilities such as Dean vortices [10], use of corrugated and/or modified membranes [11], operation under uniform transmembrane pressure [12], use of static turbulence promoters such as static mixers [13], screw-treaded inserts [14] or helical baffles [15] and more recently proposed high-shear rotary ultrafiltration [16] and the use of vibrating membranes (VSEP system) [17]. Some of these methods have been investigated for improving the flux in ultrafiltration of oil/water emulsions. Viadero, Jr. et al. [16] have shown that high-shear rotary ultrafiltration allows concentration of oil beyond the typical operating limitations of conventional ultrafiltration modules. Faibish and Cohen [18] have reported the increase by over 20% in oil rejection for a commercial cutting oil emulsion with polymer-modified zirconia-based ultrafiltration membrane compared to the native membrane. Krstić et al. [19] have shown that the use of the Kenics static mixer as a turbulence promoter during cross-flow ultrafiltration of a stable oil/water emulsion can provide flux values up to 5 times higher compared to the fluxes obtained during operation without using the static mixer. However, the increase in pressure drop across the membrane module due to presence of the static mixer, and therefore the increase in energy costs, represents the main limitation of the systems with a turbulence promoter placed in a tubular membrane.

Among the various ways to reduce fouling in ultrafiltration, another interesting process is gas

sparging. Gas bubbles are introduced directly into the feed leading to the creation of a gas/liquid two-phase flow. Gas sparging, and particularly air sparging, has been shown to be efficient in disruption of the concentration polarisation layer, and compared with conventional cross-flow operation, considerable permeate flux increases have been reported with hollow fibres [20,21] and tubular membranes [22,23]. Cui and Wright [22,23] have obtained flux increases of between 60 and 320% when ultrafiltering dextran, dried dextran and BSA solutions. They have shown that the orientation of the membrane (horizontal or vertical) and the kind of flow (upwards or downwards) affect the effectiveness of gas sparging technique. However, few studies have investigated the effect of gas injection on ultrafiltration of oil/water emulsion. Um et al. [24] have shown that nitrogen injection causes positive effect on promoting turbulence leading to flux enhancement, but it also had a negative effect of decreasing the effective membrane area due to partial occupation of membrane pores by bubbles. The efficiency of gas sparging was found to be dependent on bubble fractions in the mixture: at sufficient bubble fraction the higher flux was observed, but at lower bubble fractions the flux decreased compared to the one without gas sparging.

Gas sparging technique and a gas/liquid two-phase flow has proved to be an attractive method to enhance ultrafiltration. The technique is simple and effective involving a high wall shear stress at a relatively low pressure drop across a membrane module. Even though gas sparging does seem to be effective as, for example, using the static turbulence promoters, a gas sparged ultrafiltration is an interesting system from an energetic point of view. Possible limitation of gas sparged ultrafiltration is the problem of gas distribution and its handling in the membrane system. In order to reduce membrane fouling and improve gas distribution into the membrane module, Derradji et al. [25] have suggested a new configuration with injecting air in such a way to obtain a gas/liquid

two-phase flow that passes through a Sulzer static mixer, placed upstream of the membrane. Compared with conventional cross-flow ultrafiltration, flux improvement of up to 180% was obtained during ultrafiltration of sodium alginate.

On the basis of the above-mentioned results, a novel configuration combining air sparging and the use of a static mixer placed in a membrane tube has been proposed. If such a configuration could reduce the main limitations of both techniques, it would be a promising solution for an effective and low-energy consumption ultrafiltration process. Simultaneous use of the static mixer and air sparging was investigated during ultrafiltration of a stable oil/water emulsion.

## 2. Materials and methods

A conventional ultrafiltration set-up was redesigned to operate air sparging in cross-flow mode with a laboratory tubular single-channel membrane module (Fig. 1). The membrane used was a  $ZrO_2$  membrane (Exekia, Pall, USA) with a nominal pore size of 20 nm, length of 250 mm and diameter of 6.8 mm. The Kenics™ static mixer (FMX8124-AC, Omega, USA), consisting of 38 mixing elements with a diameter of 6.35 mm, was placed in the membrane. Detailed characteristics

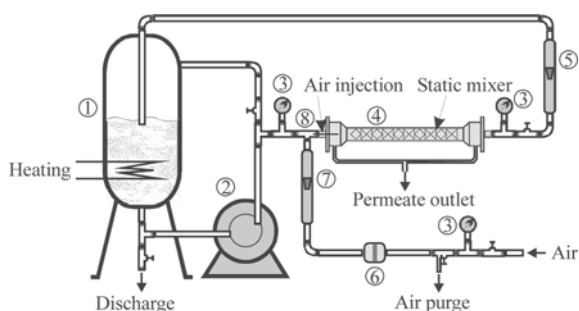


Fig. 1. Schematic diagram of the experimental set-up: 1: Feed tank; 2: Circulation pump; 3: Pressure gauge; 4: Membrane module; 5: Liquid flow meter; 6: Air filter; 7: Air flow meter; 8: Non-return valve.

and image of the static mixer can be found elsewhere [26].

A stable oil/water emulsion was prepared from water-soluble cutting oil (Unisol, Mol, Hungary) in batches of 8 L. The oil concentration in the emulsion was 5% (w/w). All experiments were carried out at 50°C. The feed was pumped from a tank to the membrane module and then recirculated. The liquid volume flow rate ( $Q_L$ ), air volume flow rate ( $Q_G$ ) and transmembrane pressure (TMP) were controlled by means of regulation valves. The permeate flux ( $J_p$ ) was calculated from the time needed to collect 10 mL of permeate. The permeate side was opened to the atmosphere, so TMP could be taken as the average of the gauge readings. The experiments were carried out at the liquid flow rates of 100 and 150 L h<sup>-1</sup> while the air flow rate was varied from 20 to 100 L h<sup>-1</sup>. Volumetric concentration factor (VCF) during concentration of the emulsion was determined as the ratio of feed volume at the beginning of operation ( $V_{feed,i}$ ) to retentate volume in time  $t$  ( $V_{ret,t}$ ):

$$VCF = \frac{V_{feed,i}}{V_{ret,t}} \quad (1)$$

The membrane was cleaned according to the recommendation of the manufacturer prior to each experiment and the pure water flux of the cleaned membrane was measured. The cleaning procedure was repeated until the original water flux was restored.

Air sparging systems are analysed via liquid and gas velocities far more often than via liquid and gas flow rates. The gas and liquid velocities are calculated as superficial velocities:

$$U_G = \frac{Q_G}{S} \quad \text{and} \quad U_L = \frac{Q_L}{S} \quad (2)$$

where  $S$  represents the effective cross-section area of the membrane tube.

Three kinds of flow patterns can be distinguished during two-phase flow in vertical tubes:

bubble, slug and annular flow. Each flow pattern corresponds to values of the injection factor defined as [21]:

$$\varepsilon = \frac{U_G}{U_G + U_L} \quad (3)$$

For values of  $\varepsilon < 0.2$ , a bubble flow is assumed. A slug flow corresponds to the values  $0.2 < \varepsilon < 0.9$ , while an annular flow is a flow pattern at the values of  $\varepsilon > 0.9$ .

Besides permeate flux, one of the most important parameter from an economical point of view is the specific energy consumption ( $E$ ) defined as the power dissipated per unit volume of permeate. In the case of liquid flow only, the hydraulic dissipated power is directly related to the pressure drop along the membrane module ( $\Delta P$ ) and the specific energy consumption can be calculated as:

$$E = \frac{Q_L \cdot \Delta P}{J_p \cdot A} \quad (4)$$

where  $J_p$  is the permeate flux and  $A$  is the membrane surface area.

In the case of gas/liquid two-phase flow, the specific energy consumption is equal to the ratio of sum of hydraulic and pneumatic powers to the permeate flow rate and is given by [27]:

$$E = \frac{Q_L \cdot \Delta P + \left( \frac{\gamma}{\gamma - 1} \right) \cdot P_D \cdot Q_{G,D} \cdot \left[ \left( \frac{P_U}{P_D} \right)^{\left( \frac{\gamma - 1}{\gamma} \right)} - 1 \right]}{J_p \cdot A} \quad (5)$$

where  $P_D$  and  $P_U$  are the downstream and upstream pressures,  $Q_{G,D}$  the gas flow rate expressed at the downstream pressure  $P_D$  and  $\gamma$  the specific heat ratio.

From Fig. 1 it can be seen that the apparatus set-up cannot provide separate measuring of the

liquid and air flow rates but only of the gas/liquid two-phase flow rate ( $Q_{G,L}$ ). Therefore, the specific energy consumption during air sparging was calculated as:

$$E = \frac{Q_{G,L} \cdot \Delta P}{J_p \cdot A} \quad (6)$$

It should be emphasized that Eq. (6) gives higher values of the specific energy consumption than Eq. (5) under the applied operation conditions.

### 3. Results and discussion

#### 3.1. The effect of air sparging on pure water flux

Before conducting the experiments with the oil/water emulsion, the air sparging system was tested with pure water in order to check whether the injected air modifies the membrane water permeability. Furthermore, the operation characteristics of the configuration with air sparging and static mixer were checked and compared with those of the air sparging configuration.

When the static mixer was used (SM mode of operation), the effective cross-section area was smaller for the value of a sectional area of the static mixer from that of the empty tube. Different effective cross-sectional areas resulted in about 20% higher velocities at the same flow rates in the case of using the static mixer compared to the velocities during operation without the use of the static mixer (NSM mode of operation). Therefore, for a given water flow rate, the pure water flux was determined for several air flow rates for both modes of operation, with the static mixer (SM) and without the static mixer (NSM). The results are presented in Fig. 2.

The results clearly show that the membrane water is not modified by air sparging which means that there is no decrease of the effective membrane surface area in contact with the liquid flow due to partial occupation of the membrane surface and/

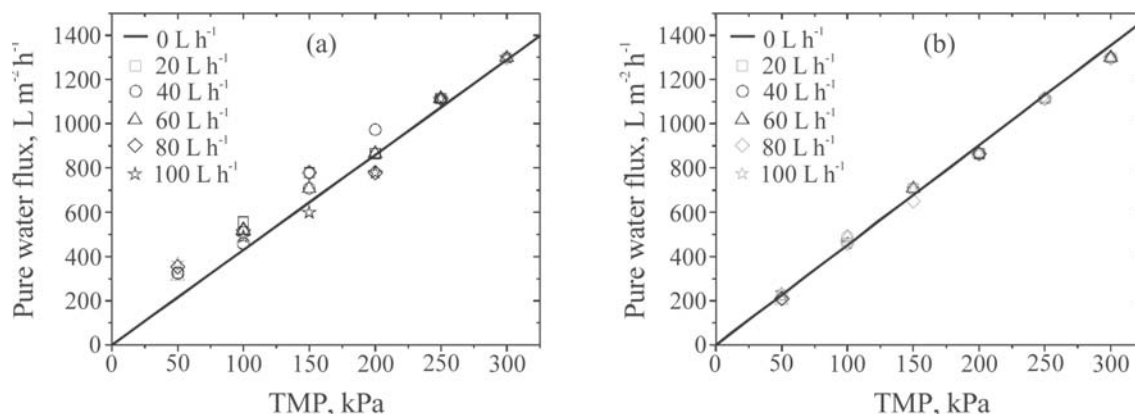


Fig. 2. Pure water flux vs. TMP for different air flow rates. Mode of operation: (a) NSM; (b) SM.  $Q_L = 100 L h^{-1}$ .

or pores by air. Moreover, the insertion of the static mixer did not disturb the operation of the apparatus and regulation of the main operation parameters.

The experiments with pure water demonstrated that the presence of air has no influence on the membrane water permeability, so the proposed configuration with air sparging and the static mixer will be examined with a stable oil/water emulsion as a feed solution in the continuation of the study.

### 3.2. Air sparging with the permeate recirculation

In order to investigate the effect of air sparging on permeate flux, the configuration without inserted static mixer was tested with the permeate recirculation to the feed tank according to following procedure: a liquid flow rate was set at the beginning of the experiment and the permeate flux was measured without the air injection. After the pseudo-steady state flux was achieved, the air was injected, first at a low flow rate and then the air flow rate was increased and the permeate flux was measured. The results are shown in Fig. 3.

The flux increase of about 6% was obtained at  $U_G$  of  $0.15 m s^{-1}$  ( $Q_G = 20 L h^{-1}$ ) which corresponds to the bubble flow in the tube ( $\epsilon = 0.16$ ). Further

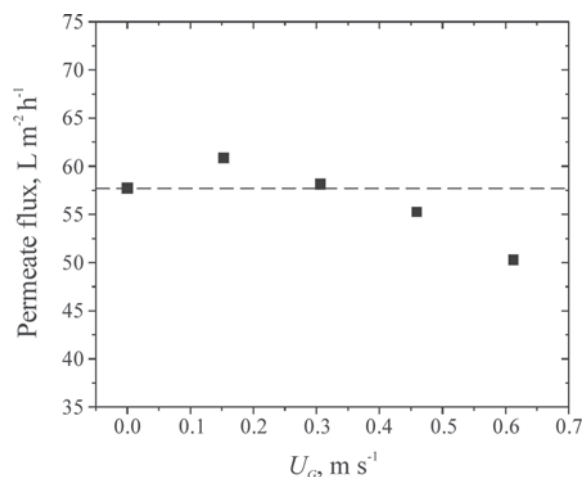


Fig. 3. The effect of air sparging on permeate flux in ultrafiltration of oil/water emulsion. TMP = 200 kPa.  $U_L = 0.77 m s^{-1}$  ( $Q_L = 100 L h^{-1}$ ).

increase of the gas velocity even lead to the decrease of the permeate flux: at  $U_G$  of  $0.61 m s^{-1}$  ( $Q_G = 80 L h^{-1}$ ), which corresponds to the slug flow pattern ( $\epsilon = 0.44$ ), the flux was around 13% lower than the flux obtained without air sparging. These results are in agreement with the results obtained during gas sparging in membrane processes involving oil/water emulsions. Um et al. [24] investigated the effect of nitrogen injection

during ultrafiltration of a 5% wt. cutting oil emulsion. Flux improvements of up to 20% were achieved under operation conditions which correspond to a bubble flow. On the other hand, Ducom et al. [8] observed the decrease of permeate flux at low gas velocities, with flux enhancements when the gas velocity was increased during nanofiltration of a 10% vol. stabilised oil emulsion. The decrease of around 9% was observed at operation conditions which correspond to a slug flow in the module ( $U_G = 0.1 \text{ m s}^{-1}$ ;  $U_L = 0.12 \text{ m s}^{-1}$ ) while the flux was enhanced by a factor 1.2 at  $U_G = 1 \text{ m s}^{-1}$ . The flux decrease at low gas velocities was explained with fractionation of the emulsion by air sparging. Due to fractionation, oil droplets become smaller which can lead to increased membrane fouling and decrease of permeate flux. By injecting air, the viscosity of a gas/liquid mixture decreased and so the turbulence in the module was increased, which could lead to an increase of permeate flux. Ducom et al. [8] suggested that the fractionation effect is predominant at low superficial gas velocities, whereas at higher superficial gas velocities, viscosity and turbulence effect predominate.

A possible explanation for the gradual decrease of the permeate flux at higher gas velocities (Fig. 3) could be the increased irreversible fouling during the duration of the experiment. Moreover, foaming was observed in the feed tank and the foam-breaker was added (Amarel 1500, OMV, Austria). The addition of a foam-breaker could have also contributed to the increased irreversible fouling of the membrane.

From the results presented in Fig. 3, it is obvious that air sparging does not seem to be an effective technique for flux enhancement in ultrafiltration of oil/water emulsions. Flux increases obtained for a bubble flow in the tube are too low compared to flux increases of up to 270% achieved with the use of the static mixer during ultrafiltration of the same oil/water emulsion [19]. However, the presence of the static mixer in a tubular membrane can cause a very high pressure drop across

the membrane which can diminish a positive effect of the permeate flux enhancement due to high energy demands of the process. On the other hand, air sparging technique can be used for maintaining a given permeate flux with a relatively low pressure drop across the membrane, and therefore low energy consumption. Combining air sparging and the use of the static mixer could be an interesting solution for improving performance. If air sparging could reduce a high pressure drop across the module and the static mixer could still provide considerable flux enhancement, the performance of the process would be significantly improved.

Fig. 4 shows the variations of the permeate flux and specific energy consumption with air flow rate during simultaneous use of the static mixer and air sparging. The experiments were started without air sparging. Then the air flow rate was increased in steps but the gas/liquid two-phase flow rate was kept constant on the value set at the beginning of the experiment. This experimental procedure caused the decrease of the liquid flow rate through the membrane due to the air flow rate increase, which resulted in a gradual decline of the permeate flux (Fig. 4a). However, the flux value of about  $180 \text{ L m}^{-2} \text{ h}^{-1}$  achieved at  $Q_G$  of  $80 \text{ L h}^{-1}$  and  $Q_{G-L}$  of  $100 \text{ L h}^{-1}$ , for example, is significantly higher than the flux of  $50 \text{ L m}^{-2} \text{ h}^{-1}$  obtained during conventional cross-flow ultrafiltration under the same operation conditions without the static mixer.

The main limitation of the configuration with the static mixer is the increase of pressure drop across the membrane module. The values of pressure drop across the module for two-phase flow during the increase of air flow rate are shown in Table 1.

The injection of air significantly reduced the pressure drop across the membrane. For example, the increase of the air flow rate up to  $80 \text{ L h}^{-1}$ , while the two-phase flow rate was kept constant on the value of  $100 \text{ L h}^{-1}$  resulted in the pressure drop of only 23 kPa in the configuration with the static mixer (Table 1), approaching the value of

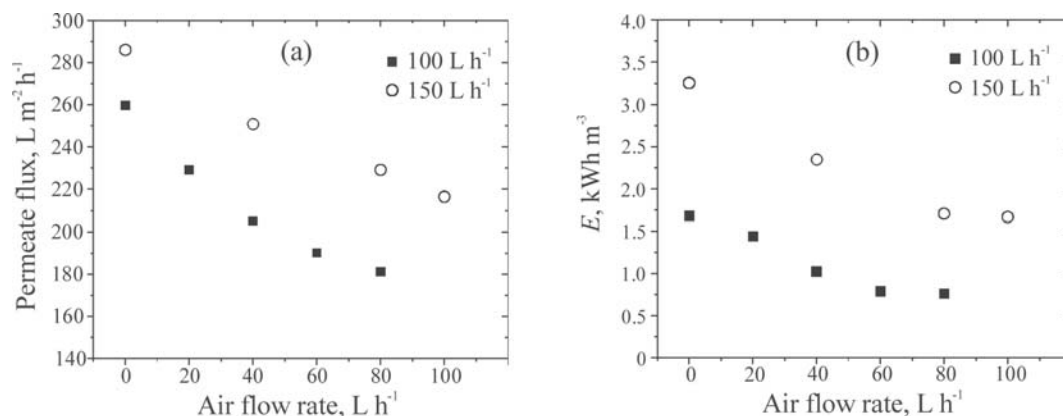


Fig. 4. The variations of permeate flux (a) and specific energy consumption (b) with air flow rate during the operation with the static mixer at  $Q_{G-L}$  of 100 and 150 L h<sup>-1</sup>. TMP = 200 kPa.

Table 1  
Pressure drop across the module for gas/liquid two-phase flow

Air flow rate (L h <sup>-1</sup> )	Pressure drop (kPa)	
	$Q_{G-L} = 100 \text{ L h}^{-1}$	$Q_{G-L} = 150 \text{ L h}^{-1}$
0	75	150
20	55	—
40	35	95
80	23	65
100	—	60

pressure drop across the empty tube (20 kPa) at a liquid flow rate of 100 L h<sup>-1</sup>.

According to Eqs. (4)–(6), a reduction of pressure drop across the membrane at a given flow rate through the module leads to reduction of dissipated power, and considerable energy saving is possible. The variation of specific energy consumption, calculated by Eq. (6), with the air flow rate during air sparging at constant  $Q_{G-L}$  is shown in Fig. 4b. Despite the flux decline (Fig. 4a), such a way of combining air sparging and the use of static mixer resulted in about 2 times lower energy consumption compared to that obtained during the operation without air sparging. Moreover, the results presented in Fig. 4b suggest the existence

of a critical air flow rate, over which further increase of the air flow rate would not improve the process performance.

### 3.3. Volumetric concentration of oil/water emulsion

The experiments with the recirculation of the permeate provided the guidelines of how the ultrafiltration involving simultaneous use of static mixer and air sparging should be carried out in order to improve the process performance. Volumetric concentration of the feed using the ultrafiltration configuration with the static mixer and air sparging was performed under a constant two-phase flow rate. The experimental procedure was the following: a liquid flow rate was set at the value of 100 L h<sup>-1</sup> and the process was started without air sparging. In this way, the positive effect of using the static mixer, such as increased wall shear rate in the vicinity of the membrane, was used at the beginning of the experiment (at low volumetric concentration factors) in order to obtain permeate flux as high as possible. On the other hand, as the feed is concentrated, permeate flux decreases due to fouling and increase of the feed viscosity and using the static mixer could only lead to high

energy consumption. Furthermore, Um et al. [24] showed that air sparging is more effective at higher concentration factors. Therefore, after a certain value of volumetric concentration factor (VCF) was achieved, the air was injected, first at a low flow rate and then the air flow rate was increased by step. Gas/flow two-phase flow rate was kept constant at the value of  $100 \text{ L h}^{-1}$ .

The permeate flux as a function of a VCF during simultaneous use of the static mixer and air sparging is shown in Fig. 5. For comparison, the variation of the permeate flux during operation only with the static mixer is also shown in the figure.

From Fig. 5, a similar pattern of the flux decline with air sparging compared to the one without injecting the air can be noticed. Moderate flux improvement was observed at an air flow rate of  $20 \text{ L h}^{-1}$ , which is in agreement with the results presented in Fig. 3. However, further increase of the air flow rate did not result in increased flux decline, as could be expected from the results obtained during the experiments with permeate recirculation. One possible explanation could be in the way the experiment was performed. Using the static mixer alone, at the beginning of the experiment, resulted in the creation of such oil deposit layer on the membrane surface that air sparging afterwards could not have any significant effect on the membrane fouling through potential fractionation of the oil droplets or even partial occupation of the membrane pores by bubbles. However, the presence of air in the membrane tube led to the reduction of the pressure drop across the module.

The variations of specific energy consumption and pressure drop across the membrane module with VCF obtained during the operations with air sparging and the static mixer simultaneously and with the static mixer alone are shown in Fig. 6.

Air sparging reduced the pressure drop across the membrane (Fig. 6b), resulting in considerable energy saving (Fig. 6a). After a VCF of 2 was achieved, the injection of air at a flow rate of  $20 \text{ L h}^{-1}$  lead to a large reduction of the specific

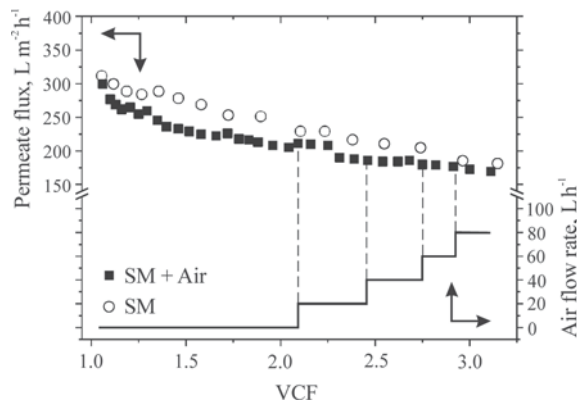


Fig. 5. Permeate flux and air flow rate vs. VCF for different modes of operation. TMP = 160 kPa.

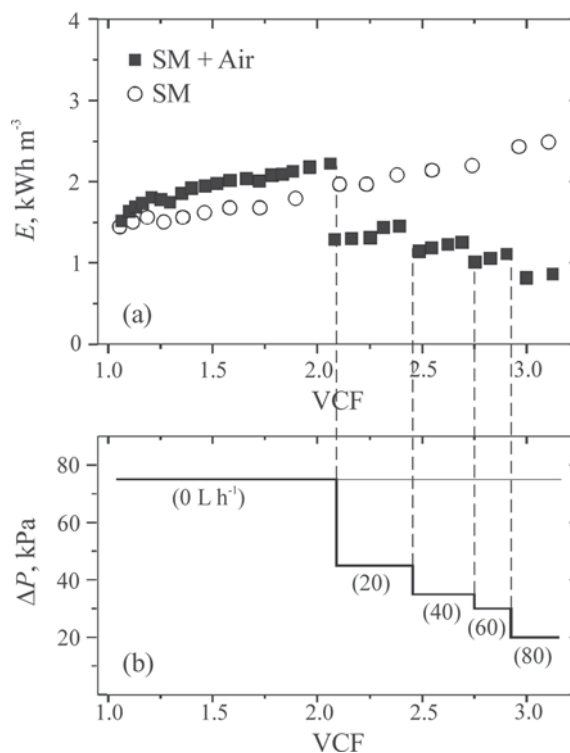


Fig. 6. Specific energy consumption (A) and pressure drop across the membrane (B) as a function of VCF for different modes of operation. TMP = 160 kPa.

energy consumption. Fig. 6 clearly shows that the specific energy consumption could be easily maintained below the value as low as  $1.5 \text{ kWh m}^{-3}$  by increasing the air flow rate step by step.

The experiments involving volumetric concentration of the feed indicated that the appropriate use of air sparging and static mixer could result in improved ultrafiltration of oil/water emulsion.

#### 4. Conclusions

The present study reports on the efficiency of a gas/liquid two-phase flow and static mixer used simultaneously in ultrafiltration of a stable oil/water emulsion. Preliminary experiments with pure water as a feed demonstrated that air sparging does not modify the membrane water permeability over a range of given operating conditions — i.e., air sparging did not cause a decrease of the effective membrane surface area in contact with the liquid flow. The results obtained during air sparging during the ultrafiltration of oil/water emulsion do not seem to be very effective for flux enhancement and it is unlikely that this technique, used alone, will have any immediate industrial application in ultrafiltration of oil emulsions. However, combining air sparging and the use of the static mixer resulted in high permeate fluxes with low energy consumption. The use of the static mixer for flux enhancement with the use of air sparging for energy saving, ensured the improved process performance. It seems that the application of this novel configuration would be especially interesting in processes where both air sparging and static mixer, used separately, can significantly improve flux or in processes already involving gas/liquid two-phase flows, such as aerobic membrane bioreactors.

#### Acknowledgement

This research was supported by the Ministry for Science and Environment Protection, Republic of Serbia (project No. 142045), CEEPUS Founda-

tion (network No. H-0158-04/05) and the Federal Ministry for Education, Science and Culture, Republic of Austria (Ernst Mach Grant).

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