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Short communication

Effect of bubble size and frequency on the permeate flux of gas sparged ultrafiltration with tubular membranes

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Abstract

Gas sparged ultrafiltration experiments are performed using a tubular membrane module with solutions of dextran and human serum albumin (HSA) as the test media. Air is injected, in a controlled manner with the ability to adjust bubble size and frequency independently, into the membrane module to create a gas–liquid two-phase crossflow operation. The effects of bubble size and frequency on the permeate flux of the sparged ultrafiltration are studied experimentally. It is found that the permeate flux increases with the bubbling frequency in the examined range. The effect of bubble size on flux can be divided into two regions, an increasing region for smaller bubbles and a plateau region for larger slugs. The results are discussed on the basis of bubble wake hydrodynamics. © 1997 Elsevier Science S.A.

Keywords: Bubble frequency; Bubble size; Dextran; Gas sparging; Tubular membrane; Ultrafiltration

1. Introduction

Ultrafiltration is a pressure-driven membrane separation process which has a wide range of applications in the dairy, water, chemical and pharmaceutical industries. In practice, two major problems associated with its application arise from concentration polarisation and membrane fouling. Whereas membrane fouling is related to the adsorption of macromolecules on the membranes and depends mainly on physico-chemical interactions between the molecules and the membranes, concentration polarisation is a physical phenomenon related to mass transfer. In ultrafiltration, solutes retained or rejected by the membrane accumulate near the membrane, which causes an increase in solute concentration near the membrane surface. This results in an increase in osmotic pressure, which reduces the effective driving force, and an increase in overall resistance to the permeate. Consequently a decline in permeate flux is inevitable [1].

Concentration polarisation can be reduced by improving the hydrodynamics of the crossflow near the membrane surface. Various techniques, such as pulsatile and reverse flow [2,3], vortex mixing [4–6], and the application of helical and baffle inserts [3] and of corrugated membrane surfaces [7], have been used to enhance ultrafiltration by promoting local mixing near the membrane. Gas sparging, i.e. injecting air bubbles into the feed to generate gas–liquid two-phase crossflow operation, has proved to be an effective, simple and low-cost technique in enhancing ultrafiltration [8,9]. Several mechanisms for this enhancement have been identified, including bubble induced secondary flow, physical displacement of the mass transfer boundary layer, reduction in membrane fouling, and pressure pulsing caused by slugs [10]. Physical displacement and reduction in fouling resistance are thought to be the main reasons for the observed enhancement in gas sparged ultrafiltration with hollow fibre membrane systems [11]. However, it is postulated that for tubular membranes the bubble induced secondary flow plays a major role in the enhancement of ultrafiltration. The secondary flow around bubbles promotes local mixing, reduces the thickness of the mass transfer boundary layer and increases the mass transfer rate of solute molecules from the membrane surface back to the bulk solution [8]. It is of considerable importance to understand the details of bubble dynamics and their effect on ultrafiltration performance so that process optimisation can be carried out to achieve the maximum enhancement with the minimum gas sparging rate.

It is commonly accepted that the turbulence in the bubble wake is responsible for the enhancement of heat and mass transfer in gas–liquid two-phase flow systems [12,13]. Experimental studies on enhanced heat transfer by injecting

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air bubbles into a liquid–solid fluidized bed indicate that the maximum increase in heat transfer rate is exhibited in the bubble wake. The strength and size of the bubble wake depend on the size of the bubble. Tiny bubbles take a spherical shape, and the flow around these spherical bubbles does not separate and hence there are no vortices generated. As the bubble size increases, the shape changes to ellipsoidal and a helical vortex is generated. Large bubbles have the shape of a spherical cap and symmetric vortex rings are created in the wake region [12,13]. The secondary flow is much stronger for the larger bubbles.

In gas sparged ultrafiltration, the effect of the wall (the membrane surface) on the secondary flow pattern can be significant. The permeation wall may also affect the strength of the wake. The hydrodynamics around bubbles may differ from those of free bubbles. The effect of bubble size and frequency on sparged ultrafiltration is yet to be determined.

In this study, experiments were carried out to investigate the effect of bubble size and frequency on the performance of gas sparged ultrafiltration with tubular membranes. During the experiments, air is injected in a controlled manner, with the ability to adjust bubble size and frequency independently so that the effect of the two factors can be identified separately.

2. Experimental

The experimental apparatus is shown schematically in Fig. 1. Tubular PVDF membranes (PCI FP100) installed vertically with a molecular weight cut-off (MWCO) of 100 kD were used in all the experiments. The membranes had an internal diameter of 12.7 mm. Two different length were used, one with an effective length of 1.18 m and an effective surface area of 0.047 m^2, and the other 0.28 m long and with 0.011 m^2 surface area. A section of a transparent tube of length 150 mm and the same diameter as the membrane was fitted in line and upstream of the long membrane tube in order to allow direct observation of the flow pattern at the inlet of the membrane. The test solution was driven from the feed tank by a gear pump and circulated through the membrane module. The permeate was weighed using an electronic balance and recycled into the feed tank at regular intervals through a peristaltic pump in order to maintain a steady feed concentration. The feed tank also acted as a gas–liquid separator during gas sparged ultrafiltration experiments.

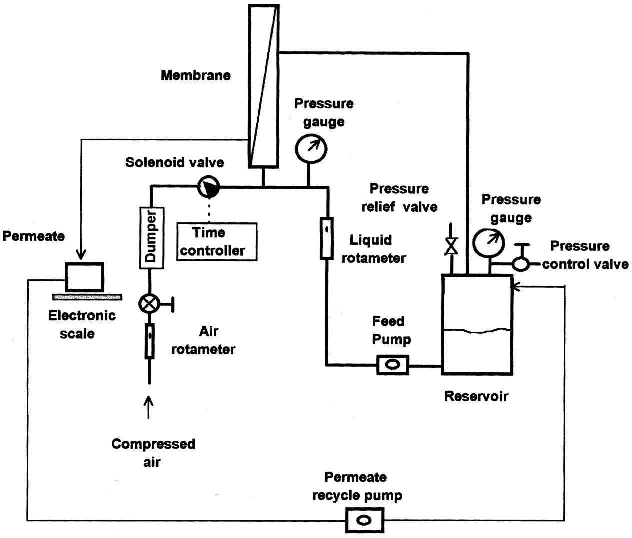

Fig. 1. Schematic view of the experimental rig.

Compressed air was directed to the inlet of the membrane module through a solenoid valve with a time controller to adjust the bubbling frequency and bubble size, incorporating air flow adjustment. The air flow rate was monitored by a rotameter and corrected according to the operating pressure. A container connected between the air rotameter and solenoid valve acted as a ''dump tank'' so as to damp the air flow pulsation, so enabling one to read the upstream air flow rotameter easily. Bubbling frequency was altered by varying the frequency of the electric pulse signal which activated the solenoid valve. Bubble size could then be varied by changing the air flow rate. For a given pressure, the relation ship between bubble volume V_b, frequency f and air flow rate F_g can be described by the following equation

$$F_g = fV_b$$

For a given frequency, varying the air flow rate can change the bubble volume according to the above relationship. It has been observed that, at a low flow rate, small slugs staying mainly in the central region were introduced, whereas at a high flowrate, large slugs were formed to occupy the whole cross-section of the tube. By varying bubble size and frequency, their effect on ultrafiltration can be identified separately. Typical values of f, F_g and V_b used in the experiment are listed in Table 1.

For gas sparged ultrafiltration experiments, the applied transmembrane pressure (TMP) was controlled by adjusting the outflow of the compressed air in the exhaust stream from the feed tank or the gas–liquid separator. In the case of single liquid phase ultrafiltration, a needle valve just downstream of the membrane was used for controlling the TMP.

Solutions of industrial grade dextran with an average molecular weight (MW) of 167 kD (Sigma, production No. D4876) and human serum albumin (HSA) of MW 67 kD (supplied by the Scottish National Blood Transfusion Ser-

Table 1
Bubble size, frequency and air flow rate

f (s^{-1})	F_g (ml min^{-1})			
	V_b = 2.2 ml	V_b = 3.3 ml	V_b = 5.0 ml	V_b = 8.3 ml
0.05	6.7	10.0	15.0	25.0
0.1	13.4	20.0	30.0	50.0
0.2	26.7	40.0	60.0	100.0
0.5	66.7	100.0	150.0	250.0
1.0				500.0

vice) were used as the test media. Dextran solutions were prepared with distilled water and HSA was prepared in 20 mM NaH_2PO_4 buffer of pH 4.7.

The membrane was cleaned prior to and after each experiment by running with 0.25% NaOH solution through the membrane module for at least 30 min, and was rinsed thoroughly with water. According to the manufacture's instructions, the membrane was stored in 0.1% sodium metabisulfite solution after each experiment to prevent bacterial growth. Pure water permeability was always checked before each experiment to assess the effectiveness of cleaning.

In the sparged experiment, the flow rates of air and feed solution, TMP and feed concentration were adjusted to the pre-set values. The weights of permeate at a certain time interval were logged into a computer. Single phase flow experiments without air sparging were also carried out in parallel for comparison. All experiments were repeated to check the reproducibility of the data, and the difference in the repeated experimental results was found to be within 10%.

3. Results and discussion

3.1. Effect of bubble size

The variation of permeate flux with air bubble size (in terms of bubble volume V_b at the operating pressure) at different bubbling frequencies is shown in Fig. 2. The experiments were performed with dextran solutions, and the other experimental conditions are indicated on the graph. In these experiments the permeate flux variation with time is similar to those reported previously [8], and the values reported here are those for the steady state flux after the initial sharp decline. The negligible change with time oberved in the experiments indicated that membrane fouling is not severe in the studied system.

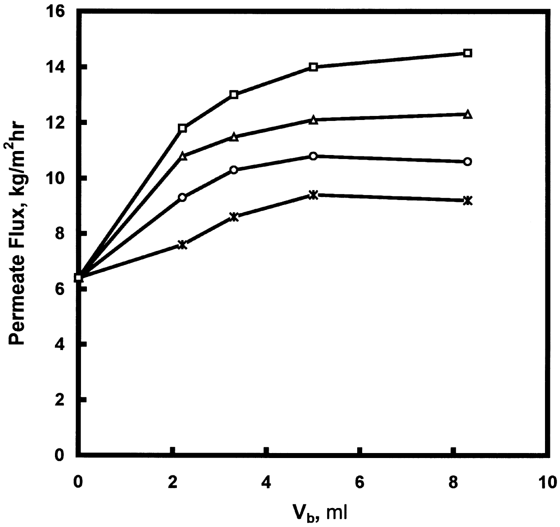

Fig. 2. Effect of bubble size on permeate flux at various frequencies.TMP = 1.0 bar, feedstock dextran $C_f = 10.0$ g l^{-1}, liquid flow rate 1.0 l min^{-1}, membrane length 1.18 mf, s^{-1}. ∗, 0.05; ○, 0.1; △, 0.2; □, 0.5.

The changing pattern of flux vs. bubble size can be divided into two regions, an increasing flux region associated with smaller bubbles and a plateau region with larger slugs. In the increasing flux region, the cross-sectional area of the bubbles is smaller than that of the membrane tube, and these individual bubbles tend to stay in the central region away from the membrane surface. For a given bubbling frequency, as the air flow rate increases, the bubbles grow in both length and cross-sectional area and tend to occupy almost the full cross-sectional area of the membrane tube, forming slugs. In the plateau region, the gas phase exists in the form of slugs, with a thin film separating the slug and the membrane. In this region, increasing bubble volume can only increase the length of the slug.

Research on bubble hydrodynamics and enhanced heat transfer has shown that both bubble size and bubble frequency have a strong influence on bubble motion characteristics [14–17], such as rising velocity, wake size, wake strength and drag force. In a system where wall effects are small, bubble rising velocity increases with bubble size and frequency for bubbles with equivalent diameter greater than 5 mm [14]. The bubble wake size and the strength of circulation in a bubble wake are also increased due to the increase in rising velocity of the bubble [15]. Kojima et al. [16] also concluded that for an isolated bubble the wake volume is proportional to the bubble size in the range of equivalent bubble diameter from 4 to 18 mm.

In the increasing region, as shown in Fig. 2, where bubble diameter is considerably smaller than that of the tube, the effect of the membrane wall is small. The behaviour of the bubbles is similar to that of free bubbles. Increasing bubble size can result in a stronger wake, which enhances the local mixing and mass transfer, and an apparent increase in permeate flux with bubble size is observed. This is in good agreement with the results from a study on bubble hydrodynamics and enhanced heat transfer [17]. Furthermore, the mass transfer boundary layer may be reduced because of the raised shear rate in the flow between bubbles and membrane wall as the bubble cross-sectional area increases, particularly when the bubbles tend to occupy the whole cross-sectional area of the membrane tube and form larger slugs.

However, in the plateau region, slugs occupy almost the whole cross-sectional area of the membrane tube. Further increases in bubble volume can only make the slugs longer. As noticed by Campos and Guedes de Carvalho [18], increasing slug length above a critical length has no effect at all on the wake size. Neither does the shear stress inside the liquid film separating the slug and the membrane surface change much with the increased slug length. As a result, increasing bubble volume in this region has little effect on permeate flux.

3.2. Effect of bubbling frequency

As shown in Figs. 2 and 3, permeate flux increases with bubbling frequency in the range examined for both dextran

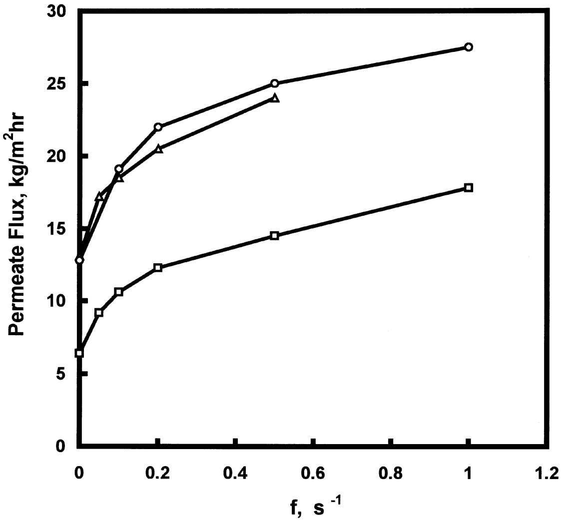

Fig. 3. Effect of bubbling frequency on permeate flux. □, 10.0 g l^{-1} dextran, $V_b = 8.3$ ml, liquid flow rate = 1.0 l min^{-1}, membrane length 1.18 m, TMP 1.0 bar. △, 4.5 g l^{-1} HSA, $V_b = 5.0$ ml, liquid flow rate 1.0 l min^{-1}, membrane length 1.18 m, TMP 1.0 bar. ○, 4.5 g l^{-1} HSA, $V_b = 8.3$ ml, liquid flow rate 0.5 l min^{-1}, membrane length 0.28 m, TMP 1.0 bar.

and HSA solutions. It is seen that the permeate flux of HSA is significantly higher than that of dextran. This may be because the size of the HSA molecule (MW 67 000) is smaller than the pore size of the membrane (MWCO 100 kD), and hence a certain amount of HSA can pass through the membrane. The observed membrane rejection is about 75% for HSA and almost 100% for the dextran.

The dependence of permeate flux on bubbling frequency in the tubular membrane system agrees well with that obtained from a study on heat transfer enhanced by a chain of bubbles in a fluidized bed, where the overall heat transfer rate was found to increase with bubbling frequency [17]. Increasing bubbling frequency means more bubble passages in unit time. When bubble frequency is low, which is the case in this investigation, the enhancement from each bubble is similar and the interaction between bubble wakes is not significant. The higher the bubble frequency, the more wakes inside the system, and the higher the permeate flux.

Another factor, which also influences the bubble induced secondary flow, is the increase of the bubble rising velocity in bubble chains. A negative pressure region is generated in the wake of a rising bubble. When the bubble below moves up near this negative pressure region, it will be sucked in and accelerated rapidly. The rising velocity of bubbles in a chain is thus higher than that of isolated bubbles [15]. The secondary flow and shear rate around the bubbles in a chain are therefore stronger, so enhancing the effect on membrane performance.

These results are somewhat different from those observed in gas sparged ultrafiltration with hollow fibre membranes [11]. In hollow fibre membrane systems, the permeate flux in ultrafiltration of dextrans and human serum albumin was found to be insensitive to the bubbling frequency in the range 0.1–0.25 Hz. In this study of tubular membrane ultrafiltration, substantial increases in permeate flux were observed when the bubble frequency was increased from 0.1 to 0.2 Hz. This indicates that different dominant mechanisms are involved to account for the observed flux enhancement.

In hollow fibre membrane systems, the fibre internal diameters are 0.2 mm and 0.5 mm. Any injected gas flows inside the fibre in the form of a long bubble train. As discussed in our previous study [11], these bubbles can extend to very near the membrane surface, creating a liquid film of <10 μm. The mass transfer boundary layer has been estimated to be thicker than this, hence it is suggested that the top part of the boundary layer is physically removed by the bubbles. Increasing bubble frequency has a positive effect on enhancing the mass transfer, but this will reduce the effective membrane area for filtration. These counter effects result in the observed insensitivity of the permeate flux to bubble frequency [11].

In the tubular membranes used in this study, the bubble frequency is less than 1 Hz. The interaction between adjacent slugs is small and thus they behave like individual slugs or bubbles. It is therefore observed that increasing bubble frequency results in a higher permeate flux.

If the bubble frequency is increased further, when the space between adjacent slugs becomes smaller than the length of the primary wake of each bubble, the primary wakes of neighbouring bubbles may tend to overlap each other and the wake regions of each bubble become indistinguishable, continuously occupying the whole membrane tube. Increasing the bubbling frequency may not improve the mass transfer further but may result in the coalescence of slugs, and transition of the flow pattern to churn flow or annular flow may occur. However, as our interest is focused on low flow rate gas injection to enhance ultrafiltration and to minimise bubble damage to macromolecules in solution, experiments with high bubbling frequency are not explored in this study.

4. Conclusions

Bubble size and frequency have strong influence on the permeate flux of sparged ultrafiltration. Using a 0.0127 m ID membrane tube, it is found that, over the range of bubble volumes from 2.2 to 8.3 ml, the dependence of permeate flux on bubble size has two regions, an increasing region associated with small slugs and a plateau region with large slugs occupying the full cross-sectional area of the membrane tube. The enhancement of gas sparging is increased with bubbling frequency in the examined range of less than 1 Hz.

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