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Influence of dispersed particulates on mass transport in cross-corrugated structures

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Abstract

A study of the effect of particulate solids on mass transport behaviour in cross-corrugated membrane systems is reported. The electrochemical limiting current technique was employed to determine the mass transfer coefficients. The influence of cross-corrugated structures, to simulate membranes, and the effect of the orientation of flow to the angle of corrugation (0°, 45° and 90°) on the mass transfer coefficient is reported. The effect of particulate solids on mass transport was investigated using SiO_2, at concentrations of up to 0.2 wt %. The presence of SiO_2, at a concentration lower than 0.2%, gave higher values of mass transfer coefficient compared to those in the absence of the solid. The mass transport correlations showed that even at low values of Reynolds number there is evidence of turbulence.

List of symbols

C_b	bulk reactant concentration (kmol m^{-3})
D	diffusivity (m^2 s^{-1})
d_h	equivalent diameter (m)
F	Faraday's constant (96 501 C mol^{-1})
j_L	limiting current density (A m^{-2})
k	mass transfer coefficient (m s^{-1})
L	a characteristic length for the geometry of the flow model (m)
Re	Reynolds number ($\frac{uL\rho}{\mu}$)
Sc	Schmidt number ($\frac{v}{D}$)
Sh	Sherwood number ($\frac{kL}{D}$)
u	velocity (m s^{-1})
x	distance from the leading edge (m)

Greek letters

μ	viscosity (kg m^{-1} s^{-1})
v	kinematic viscosity (m^2 s^{-1})
ρ	density (kg m^{-3})

1. Introduction

The last few decades have seen the introduction of a range of separations based on the concept of a membrane to supplement or replace distillation, adsorption, extraction, and crystallisation processes. The performance of a membrane can be defined in terms of two simple factors, flux or permeate rate (mass transfer) and selectivity [1]. The flux achieved through many membrane systems can be limited by the resistance of the membrane itself, by concentration polarisation near the surface of the membrane and by fouling. To limit or prevent fouling the hydrodynamic conditions may be modified so as to increase the shear stress in the boundary layer. This also serves to decrease the effect of concentration polarisation at the membrane surface. Many different approaches can be used to improve the flux in membrane processes. Most of the recent developments in mass transfer enhancement have focused on the design of new membrane modules, including volume displacement rods in tubular membranes, turbulence promoters attached to, or at some distance from, the membrane, fluidised beds in tubular membranes, and mesh screens in flat and spiral-wound membranes [2]. However, flux enhancement is usually achieved at the expense of significant increased frictional pressure drop. More recently, there has been interest in using corrugated membranes as turbulence promoters in order to reduce both concentration polarisation and membrane fouling [3]. It is proposed that a corrugated membrane would promote turbulence preferentially near the wall region, causing less pressure drop in the flow channel along the membrane, than with other techniques [3]. So it would be expected that, using a double layer of corrugated membranes, with flow between the layers, the turbulence of flow should be further increased in comparison to that with a single corrugated membrane layer.

This work was performed to obtain experimental data for a mass transport study of cross-corrugated membranes with different angles of flow to the direction of the corrugation and the influence of particulate solid (SiO_2), using the well known limiting current technique [4, 5]. Correlations relating the Reynolds, Schmidt and Sherwood numbers for the cross-corrugated module are established.

2. Experimental

The reaction used in this study to determine mass transport coefficients is the cathodic reduction of ferricyanide ions to ferrocyanide ions at an inert electrode according to the following reaction:

$$Fe(CN)_6^{-3} + e^- \longrightarrow Fe(CN)_6^{-4} \quad (1)$$

The reverse reaction proceeds at the anode, so that the bulk concentration of the electroactive species remains unchanged. The method constitutes a convenient and accurate procedure for mass transfer rate determination, and can be applied in a wide variety of geometric and hydrodynamic situations [6–8].

Maintaining conditions conducive to electrolyte stability is vital with this system, so the exclusion of light and dissolved oxygen is necessary.

The mass transfer coefficient, k, is determined from the limiting current density, j_L, using

$$k = \frac{j_L}{FC_b} \quad (2)$$

where C_b is the concentration of ferricyanide in bulk solution.

Current densities are calculated on the basis of the actual area of the corrugated electrode.

A cell fabricated from acrylic was used to emulate the cross-flow membrane unit with 'membrane simulants'. Two corrugated sheets, 67 mm × 35 mm were used and placed at 90° to each other. The corrugations were 1 mm deep and 2 mm wide as shown in Figure 1.

An EG&G Princeton Applied Research VersaStat was used to conduct the electrochemical reaction under linear sweep voltammetry. The data was logged using EG&G Model 270 Research Electrochemistry Software.

In this study, the electrolyte composition was typically:

0.3 M Na_2CO_3	supporting electrolyte
0.01 M $K_3Fe(CN)_6$	ferricyanide
0.1 M $K_4Fe(CN)_6$	ferrocyanide in excess

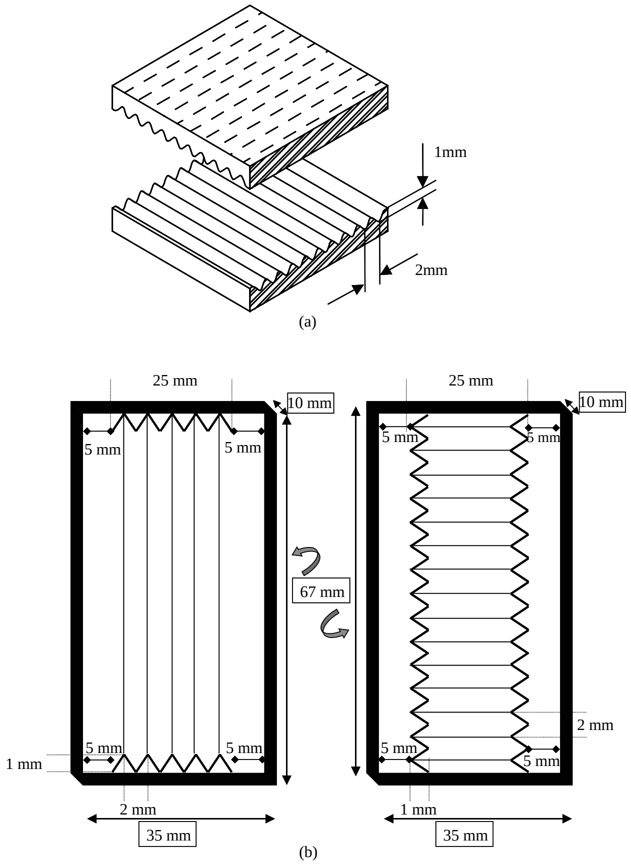

Fig. 1. (a) Corrugated membrane plates (not to scale); (b) dimensions of the membrane plates (not to scale).

A thin (30 μm) nickel cathode, 10 × 10 mm, was pressed into one of the membrane plates to take the shape of the corrugations. Figure 2 is a SEM picture of the electrode used in channels with 0°, 45° and 90° angles. A smooth nickel electrode of approximately four times the surface area of the cathode was used as the anode. Since the cathode was smaller, and the ferricyanide ion concentration was much lower than that of ferrocyanide, the process occurring at the cathode controlled the cell current. The back of the cathode was coated with insulating paint, and an electrical connection established at the side edge to avoid interference to the flow pattern. Nitrogen was bubbled through the electrolyte to expel dissolved oxygen prior to tests.

Experiments were conducted at a constant flowrate, using a slow linear potential sweep applied to the cell, with the corresponding currents measured. As the voltage increased, the current increased to a maximum, and the resultant limiting current plateau range, which extended over a large range of cell voltage, was identified. Once the limiting current was determined (at the mid-potential of the plateau) the process was repeated over a range of flow rates.

The flow between the corrugations was at one of three angles to the corrugated electrode:

1. at 0° to the corrugation at the electrode, i.e. along the trough of the corrugation,
2. at 45° to the corrugation at the electrode,

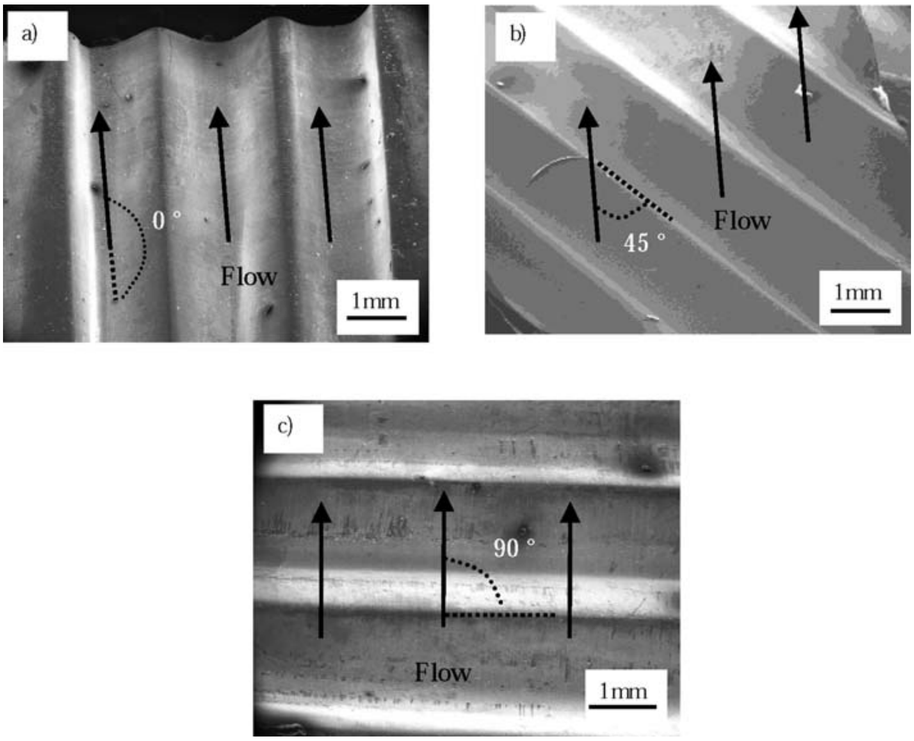

Fig. 2. Details of the angle channels of the corrugated electrodes with the flow direction: (a) 0°; (b) 45°; (c) 90°.

3. at 90° to the corrugation at the electrode, over the ribs of the corrugation.

The corrugated membrane plates were crossed as follow:

1. 0° crossed with 90°,
2. 90° crossed with 0°,
3. 45° crossed with 90°,
4. 90° crossed with 45°,

where the first angle indicates the angle of the corrugation of the 'membrane' into which the electrode was pressed.

To investigate the influence of suspended particulates, inert SiO_2 solids (size range 0.074–0.165 mm) were used in the wt % range of 0 to 0.2%. The influence of viscosity and thus Schmidt number was investigated by adding different percentages of glycerol to the electrolyte solution. Viscosity was calculated using published relationships [9]. The diffusion coefficients for the ferricyanide ion were calculated using the well-known Wilke–Chang equation [9, 10] at 298 K:

$$Sc = 1483 : D = 6.76 \times 10^{-10}\ \mathrm{m\,s^{-2}}$$

$$Sc = 4998 : D = 3.57 \times 10^{-10}\ \mathrm{m\,s^{-2}}$$

3. Results and discussion

3.1. *Effect of the* SiO_2

Figure 3 shows the variation in mass transfer coefficients with Reynolds number for three cross-corrugated membranes and different percentages of SiO_2, from 0 to 0.2% of solid. Values of mass transfer coefficient were high, up to 2×10^{-4} m s^{-1}, with this cross-corrugated structure. The presence of SiO_2 in the solution produced a slight increase in the mass transfer. As the percentage of SiO_2 increased the mass transfer coefficient increased up to 0.1% SiO_2. There was no difference in the mass transport coefficient for 0.1 and 0.2% SiO_2; suggesting a limit to the effect of suspended solids in enhancing mass transfer in cross-flow filtration.

When data for the influence of angle of corrugations are compared it can be seen that the greatest mass transport was accomplished at a cross-corrugated membrane of 90° crossed with 45° (Figure 4). Figure 4 shows representative data for mass transfer coefficient vs Reynolds number, at 0.1% of SiO_2, for the different cross-corrugated membranes studied. In the range of Reynolds number studied, there was a steady increase in mass transport coefficient from approximately 4×10^{-5} to 2×10^{-4} m s^{-1}. This represents a substantial enhancement in mass transport with this cross-corrugated membrane system.

An analogous set of experiments was conducted with a value of solution viscosity, which gave a Schmidt number of 4998. The results are plotted in Figure 5, as mass transfer coefficients for different Reynolds numbers with different percentages of SiO_2. In this case, 0.2% SiO_2 produced a slight decrease in the mass transfer with regard to the experiments without solids. With 0.15% SiO_2 a slight increase in the mass transport, which was higher with the cross-corrugated membrane positioned as 90° crossed with 45°, was observed.

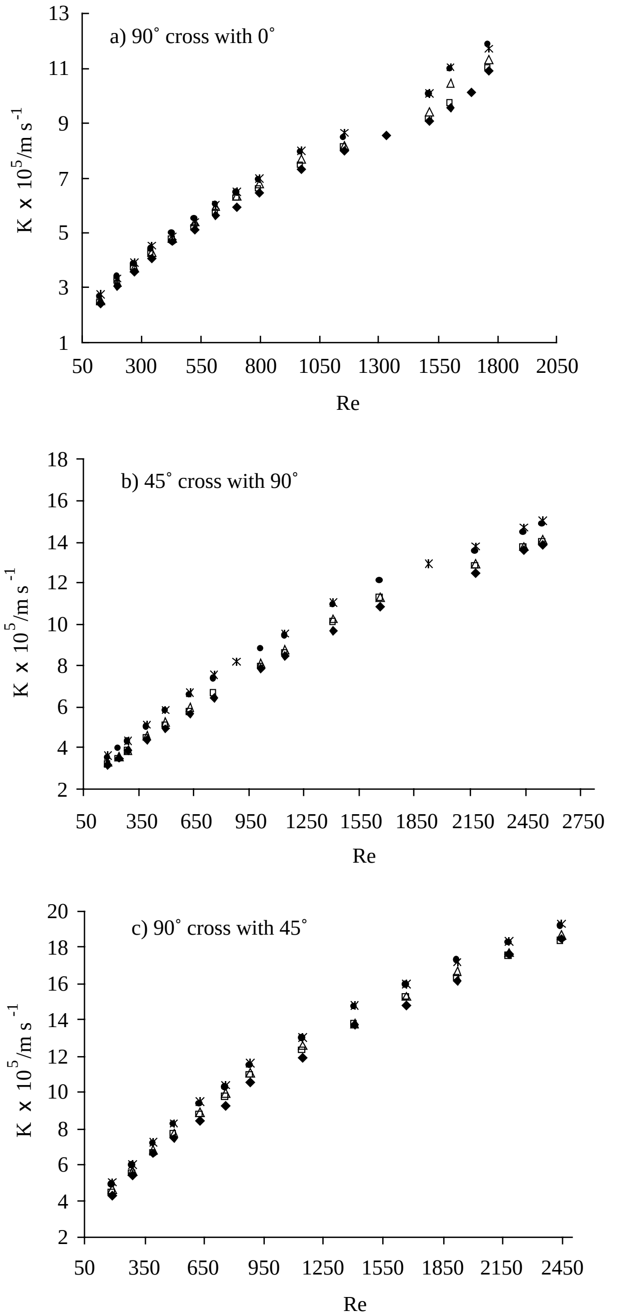

Fig. 3. Mass transfer coefficients at Reynolds numbers and Schmidt number of 1483. (a) 90° cross with 0°; (b) 45° cross with 90°; (c) 90° cross with 45°; (♦) 0% solid; (□) 0.025% solid; (△) 0.05% solid; (●) 0.1% solid; (∗) 0.2% solid.

As the Schmidt number increased, the mass transfer coefficient decreased for all the cross-corrugated membranes studied. For the Schmidt number of 4998 and the following conditions: 0.1% SiO_2 and a cross-corrugated design of 90° crossed with 45°, the highest value of mass transfer coefficient was 9.6×10^{-5} m s^{-1}, whereas, for a Schmidt number of 1483 and the same Reynolds number and with 0.1% SiO_2, the mass transfer coefficient was 1.3×10^{-4} m s^{-1}. Therefore, it appears that percentages of SiO_2 higher than 0.2% may not enhance mass transport.

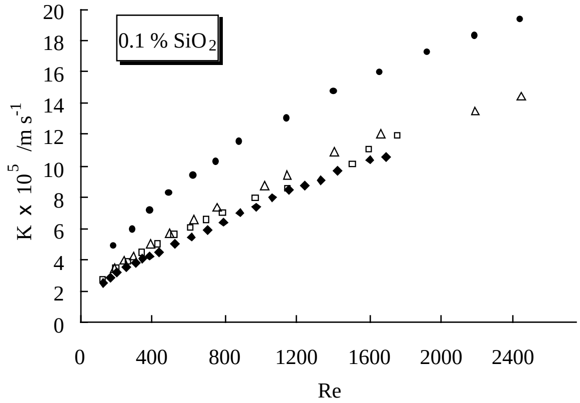

Fig. 4. Mass transfer coefficients at Reynolds numbers, 0.1% of SiO_2, and Schmidt number of 1483; (♦) 0° cross with 90°; (□) 90° cross with 0°; (△) 45° cross with 90°; (●) 90° cross with 45°.

Figure 6 shows the influence of different percentages of SiO_2 on mass transfer coefficient for both Schmidt numbers studied and similar values of Reynolds numbers for the configuration 90° crossed with 45°. The mass transfer coefficients were higher for the lower Schmidt number, because of the lower viscosity of solution. The effect of the solid was similar for both

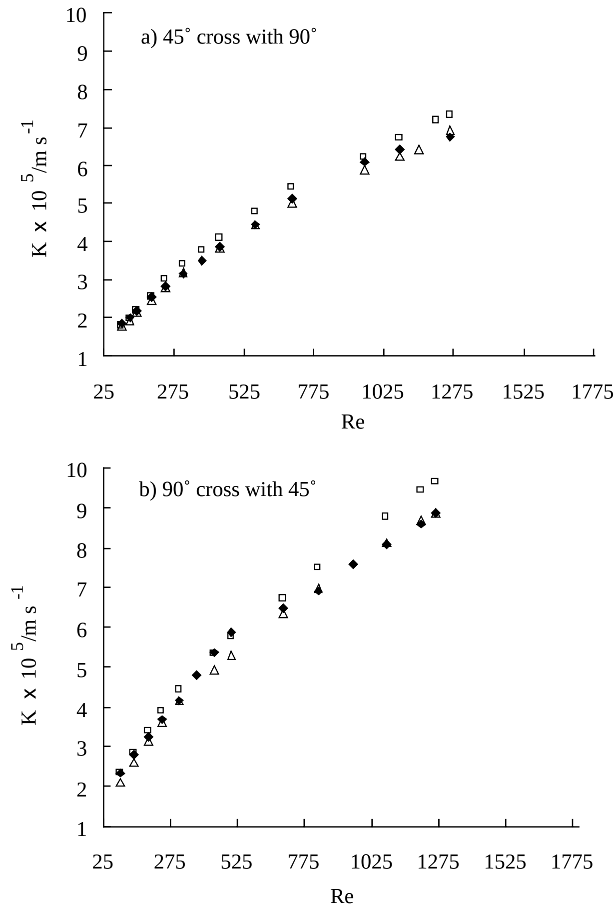

Fig. 5. Mass transfer coefficients at Reynolds numbers and Schmidt number of 4998. (a) 45° cross with 90°; (b) 90° cross with 45°; (♦) 0% solid; (□) 0.15% solid; (△) 0.2% solid.

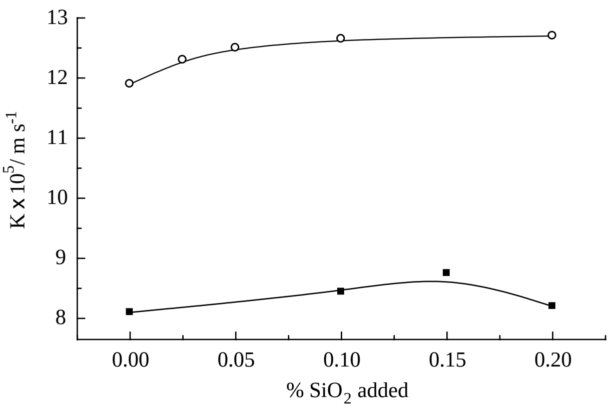

Fig. 6. Mass transfer coefficient at% of SiO_2 added; (–○–) $Sc = 1483$; $Re = 1150$; (–■–) $Sc = 4988$; $Re = 1080$.

Schmidt numbers studied, due to the fact that the flow of solid particulates produces micro-turbulence which enhances mass transfer.

3.2. *Correlation of data*

A correlation relating the Reynolds, Sherwood, and Schmidt numbers for the cross-corrugated module, 90° crossed with 45° was established. The following equation, valid for mass transfer in turbulent channel flow [4, 11], was used to correlate the data:

$$Sh = C\,Re^{0.58}Sc^{0.333} \tag{3}$$

Figure 7 shows the correlation of experimental data for 0 and 0.1% SiO_2, for a Schmidt number of 1483, and for a cross-corrugated membrane of 90° crossed with 45°.

Figure 8 is similar to Figure 7 but in this case the percentages of SiO_2 shown are 0 and 0.15% and the Schmidt number is 4998. The data are correlated by Equation 3.

Table 1 gives the values of the constant *C*, and the Reynolds exponent, a, for both Schmidt numbers, with and without solids. These values are obtained by the linear regression of Figures 7 and 8. The slope of the correlations is higher than the theoretical expressions in the laminar region and is very similar to those shown by the theoretical turbulent expression. This behaviour has also been observed by several authors [12–14] for a wide group of commercial and homemade cells. It can be concluded, that the data fit an equation corresponding to a system exhibiting turbulent characteristics.

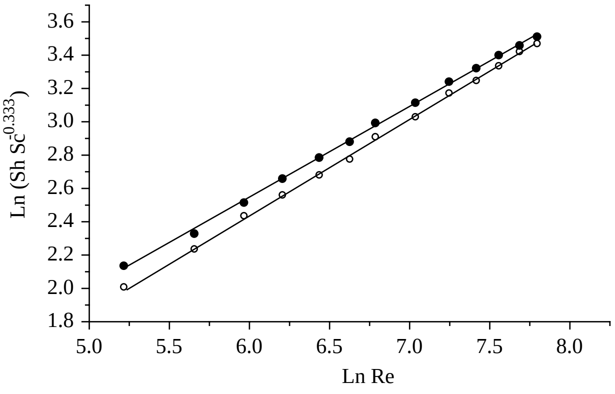

Fig. 7. Experimental data and best fit of equations assuming laminar and turbulent conditions. 90° cross with 45°, Schmidt number 1483, 0% of SiO_2 and 0.1% of SiO_2; (○) exp. data (0% SiO_2); (●) exp. data (0.1% SiO_2).

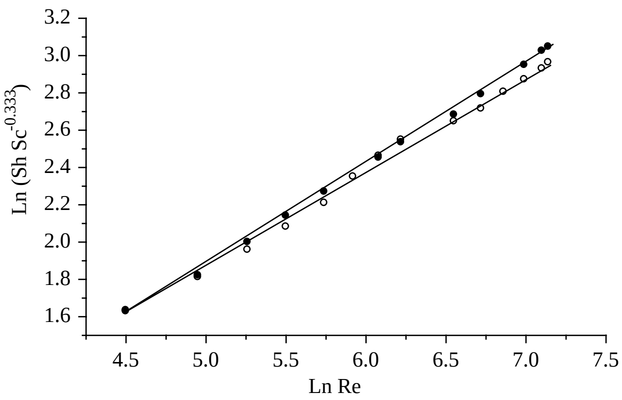

Fig. 8. Experimental data and best fit of equations assuming laminar and turbulent conditions. 90° cross with 45° and Schmidt number of 4998, 0% SiO_2 and 0.15% SiO_2; (○) exp. data (0% SiO_2); (●) exp. data (0.15% SiO_2).

Table 1. Values of constant *C* and exponent Reynolds number *a* with and without SiO_2

	$Sc = 1483$		$Sc = 4998$	
	0% SiO_2	0.1% SiO_2	0% SiO_2	0.15% SiO_2
C	0.368	0.476	0.487	0.432
a	0.57	0.55	0.52	0.54
R^2	0.998	0.998	0.995	0.997

4. Conclusions

Mass transport coefficients at the surface of cross-corrugated plates, with different angles of liquid flow were determined. The turbulence created by the system of cross-corrugated plates of 90° crossed with 45° was the highest of all the systems studied. The presence of SiO_2 particles in the electrolyte produced a slight increase in the mass transport. The mass transport correlation showed that the laminar flow regime, normally associated with low Reynolds numbers, does not represent the conditions in the cross-corrugated module. It is likely that the irregular flow path caused by the membrane corrugation promoted turbulent flow and enhanced mass transfer.

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