Distillation Absorption 2010
A.B. de Haan, H. Kooijman and A. Górak (Editors)

EFFECT OF HYDRODYNAMIC CONDITIONS ON MASS AND HEAT TRANSFER IN A NOVEL MEMBRANE BASED CONTINUOUS MICRO-DISTILLATION DEVICE: EXPERIMENTAL APPROACH

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Abstract
In this work the influence of hydrodynamic conditions on the mass and heat transfer in a novel micro-separator combining membrane distillation concept with micro-fluidic structures has been investigated. In both feed and permeate channels laminar flow conditions have been established.
The temperature polarization effect was localized in both the feed and permeate channels and was found to be strongly dependent on the feed composition and the hydrodynamic conditions in both channels. On the other hand, the inert gas flow rate was identified as a crucial operating parameter influencing both the mass and heat transfer in the micro-separator.
In addition, the selection of an appropriate membrane liquid-vapor/gas contactor was found to be an important design parameter for the reduction of temperature polarization effects. As compared to existing membrane distillation processes, the new membrane distillation based micro-separator offers a substantial potential to enhance the separation performance particularly for mixtures with high methanol concentration.

Keywords: Micro-separation, sweep gas membrane distillation, micro-fluidic structure, liquid-vapor/gas membrane contactor, aqueous mixture with low to high methanol concentration

1. Introduction

A continuous micro-distillation device was designed and tested for the separation of a mixture of methanol and water[1]. The novel micro-separator consists of two horizontal polycarbonate plates (length: 60 mm, width: 30 mm, thickness: 1 mm) which are joined together holding a flat micro-porous polymeric hydrophobic / oleophobic membrane (active area: 171 mm^2) in between (Fig. 1). In each plate a meandering rectangular channel is milled (channel height H: 1 mm, channel width W: 1 mm, and channel length l: 342 mm). Each separation plate is imbedded into a slit machined into the respective cover polycarbonate plate (external dimension: 110 mm × 110mm × 30mm, internal dimensions: 80 mm × 80 mm × 20 mm) used for mechanical stability, thermal insulation and for visual inspection. Both cover plates are screwed together. The heated feed (sub-cooled liquid) is circulated through one of the micro-channels and the cold carrier inert gas (dry nitrogen) is circulated through the other one. The two streams can be arranged in co-current or counter-current mode, tangentially to the membrane surface. The membrane serves as a liquid-gas contactor using interfacial tension to stabilize the liquid-gas interface between the process fluids repelling the liquid phase and creating a liquid-vapor interface at the entrance of the pores. The volatile components (methanol, water) evaporate, and diffuse through the membrane pores. The inert gas sweeps the permeated distillate through the membrane carrying it outside the device.

In this paper the influence of the hydrodynamic conditions in both feed and permeate channels on the partial distillate fluxes of methanol and water is investigated for two types of membrane contactors.

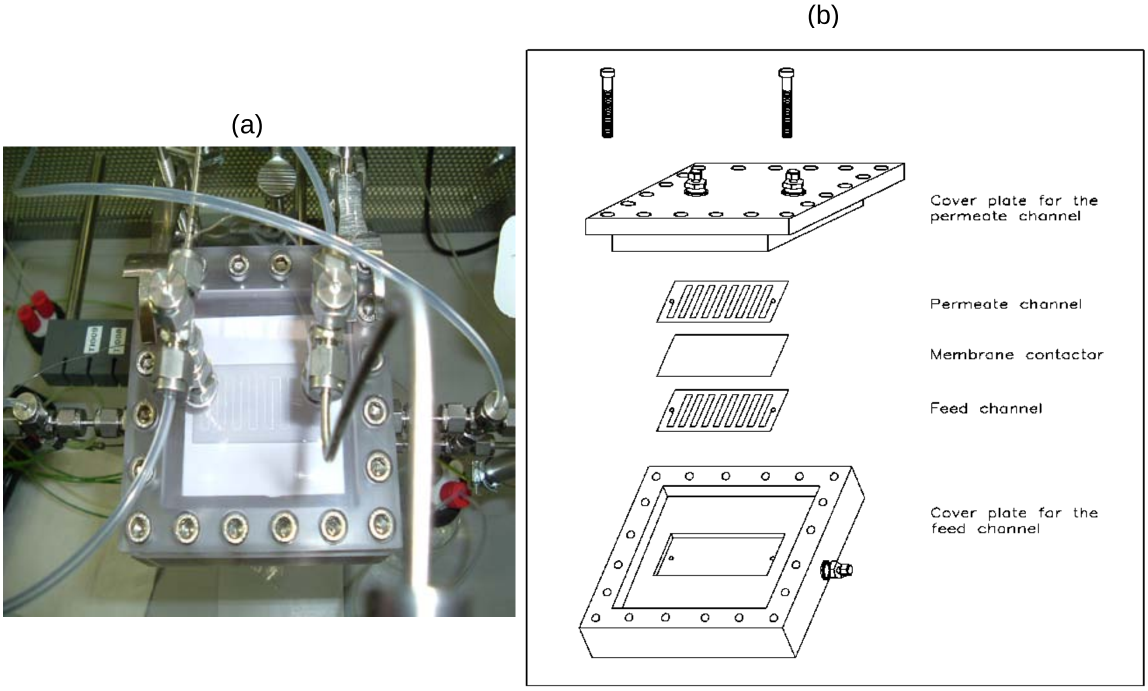

Figure 1. a) Micro-separator, b) Exploded view of the micro-separator

2. Material and methods

Feed mixtures have been prepared with ultra pure water (Millipore, USA) and methanol (99.9 %, Merck, Germany). The carrier gas dry nitrogen (99 %) was supplied by Westfallen (Germany). Analysis of the distillate was performed by means of a gas chromatograph (Agilent 6890 series) equipped with a flame ionization detector (FID), a thermal conductivity detector (TCD) and a HP Innowax column (L=30 m, ID = 0.25 mm). The experimental set-up is described elsewhere[1]. Two commercial types of membranes already tested in our previous study[1] have been used as liquid-vapour/ gas contactors: (a) an oleophobic polyethersulfone polymer cast on a non-woven polyester support membrane (PES40), provided by Pall (USA) and (b) an oleophobic expanded polytetrafluoroethylene membrane (ePTFE45) on a non-woven polyester support supplied by Gore (USA). These membranes are characterized by their thickness, pore size, volume fraction void (porosity), thermal conductivity, and water entry pressure LEP_w. The latter constitutes the main constraint for the process operation as it represents the breakthrough pressure from which the liquid penetrates into the pores of the membrane. To avoid liquid intrusion into the membrane pores, the pressure differences across the membrane are continuously controlled by means of differential pressure transmitters IDM 331 (ICS, Germany) placed at the inlet and outlet of the micro-separator and maintained below the breakthrough pressure of the membrane. Table 1 summarizes the characteristics of the above cited membranes.

Table 1. Characteristics of the membrane contactors

Membrane	Pore size (µm)	Thickness (µm)	Porosity (vol %)	LEP_w (bar)
PES40	0.40	166	0.55	1.37
ePTFE45	0.45	250	0.91	1.30

3. Experimental results and discussion

The effect of the hydrodynamic conditions on the mass and heat transfer in the novel micro-separator in terms of partial distillate fluxes of methanol (N_1) and water (N_2) has been investigated in both feed and permeate channels. For this purpose three sets of experiments have been performed for mixtures with low to high methanol concentration and for each membrane contactor. For each set of experiments the feed composition x_F and the flow rate of one process stream (feed mixture (L_F) / inert gas nitrogen (G)) were held constant while the flow rate of the other one (inert gas nitrogen / feed mixture) was varied within the allowable operating range with respect to the operating constraint on the membrane contactors. For all experiments, inert gas nitrogen was introduced at room temperature (22 ^{0}C) and the feed temperature T_F was kept constant. In each channel, the Reynolds number writes:

$$Re_{F(G)} = \frac{v_{F(G)}.\rho_F.d_h}{\mu_{F(G)}}, \qquad (1)$$

where the hydraulic diameter d_h is given by

$$d_h = \frac{2.H.W}{(H+W)}. \qquad (2)$$

The thermo-physical properties of the feed mixture are calculated for a given feed composition at the average temperature value between the inlet and outlet of the corresponding channel. Whereas those of the carrier gas nitrogen are determined for the temperature and pressure conditions at the inlet of the permeate channel. Both sets of properties have been obtained by using Aspen-Properties tools.

3.1 Effect of hydrodynamic conditions in the feed channel

At fixed feed temperature, the operating range for the feed flow rate was established by inspecting visually the wettability limit for each membrane contactor for a given feed mixture[1]. For the three tested feed compositions, Fig. (2) illustrates the influence of the Reynolds number in the feed channel Re_F on both methanol and water fluxes. For both membrane contactors similar trends were obtained with higher values for ePTFE45 as compared to PES40. Fig. (2.a) shows that for mixtures with low methanol concentration, the effect of increasing Re_F on the methanol flux is almost negligible, whereas for mixtures with moderate to high methanol concentration, increasing Re_F results in a slight increase of methanol flux as shown in Figs. (2.b) and (2.c). On the other hand for mixtures with low to moderate methanol concentration, the water flux increases slightly with Re_F whereas it remains almost constant for mixtures with high methanol concentration.

It is worth pointing out that for the three levels of feed compositions (mixtures with low, moderate and high methanol concentration) the corresponding ranges of water flux obtained over the whole range of Re_F are almost identical. Such as the effect of methanol content in the feed on the water flux is almost negligible. On the other hand, higher the methanol concentration in the feed higher is the respective flux. This behaviour can be attributed to the improvement of the heat transfer in the feed channel with increasing feed flow rate and resulting therefore in the enhancement of the driving forces with a higher extent for methanol transport. However it is worth mentioning that for each set of experiments corresponding to a given feed composition, the effect of increasing Re_F on the separation factor is almost negligible[1].

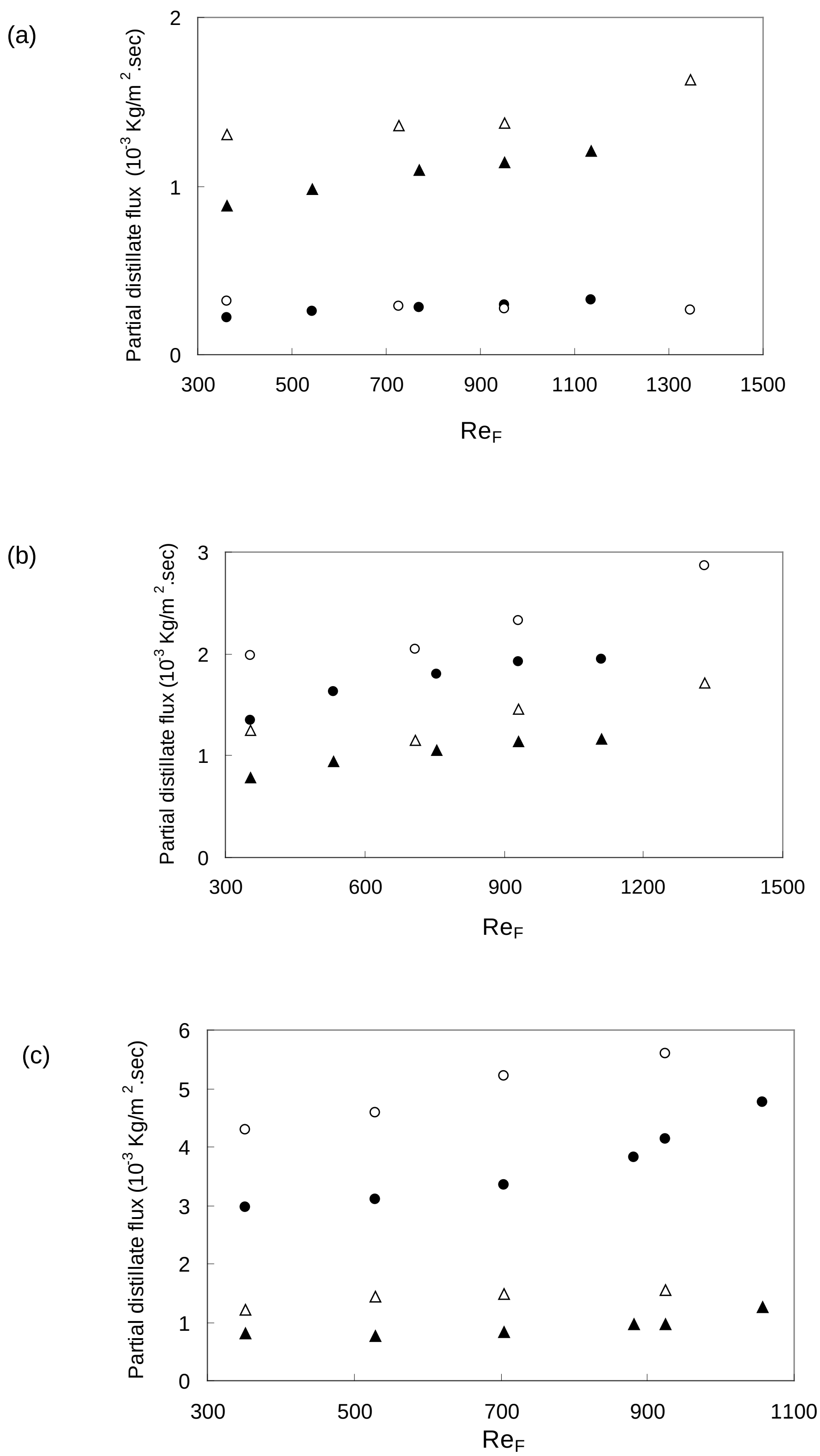

Figure 2. Impact of a variation of the Reynolds number in the feed channel Re_F on the partial distillate fluxes (methanol, water) for: a) x_F = 0.05 Kg/Kg, b) x_F = 0.30 Kg/Kg, c) x_F = 0.60 Kg/Kg, T_F = 60 ºC, Re_G = 74, (●) N_1 (PES40), (○) N_1 (ePTFE45), (▲) N_2 (PES40), (△) N_2 (ePTFE45)

3.2 Effect of hydrodynamic conditions in the gas channel

For the three set of experiments, the range of Re_G in the gas channel has been obtained by considering the allowable operating range of inert gas flow rate such as the pressure in the permeate channel remains smaller than that in the feed channel.

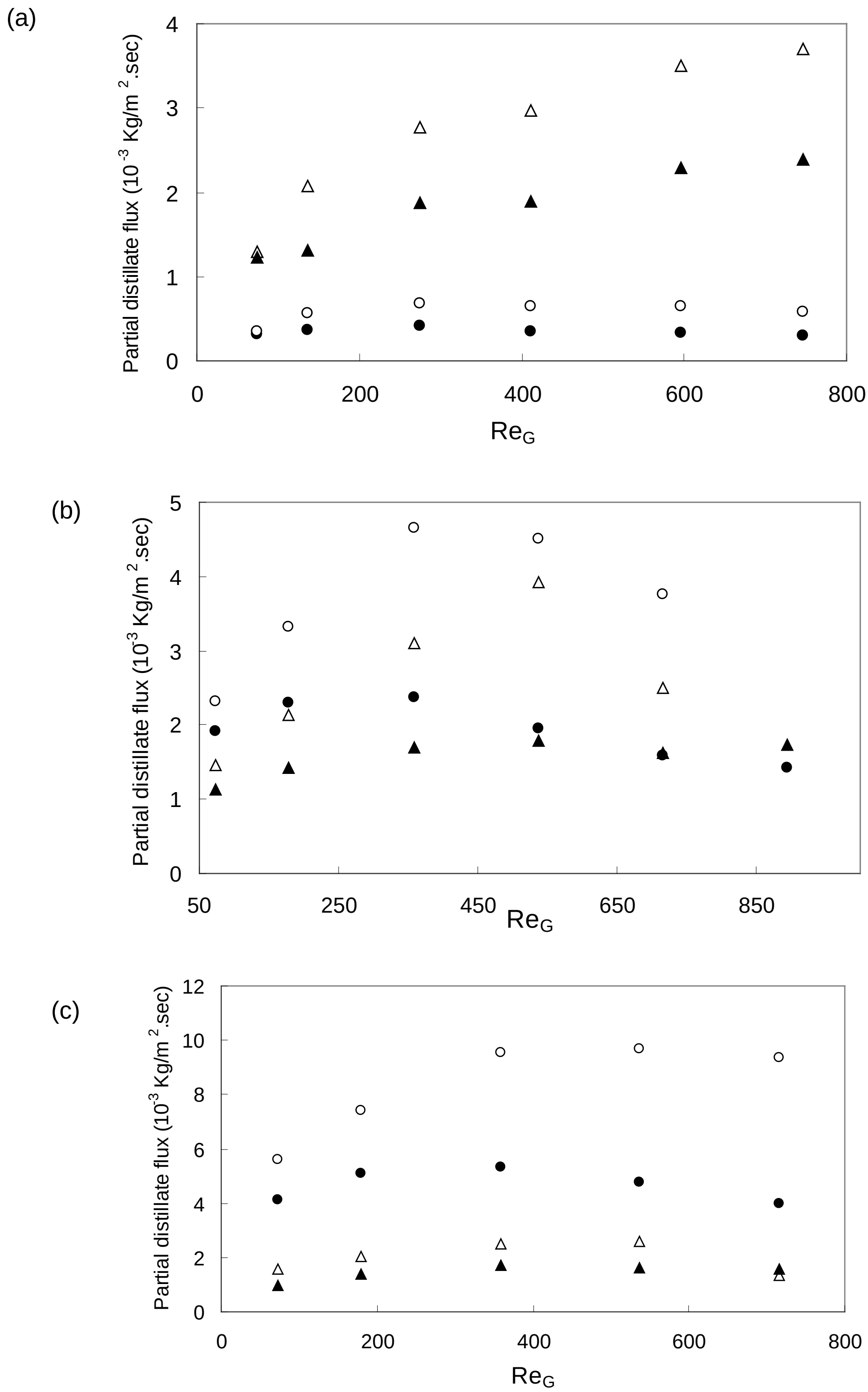

Figure 3. Impact of a variation of the Reynolds number in the gas channel Re_G on the partial distillate fluxes (methanol, water) for: a) x_F = 0.05 Kg/Kg, b) x_F = 0.30 Kg/Kg, c) x_F = 0.60 Kg/Kg, T_F = 60 ºC, Re_F = 953, (●) N_1 (PES40), (○) N_1 (ePTFE45), (▲) N_2 (PES40), (△) N_2 (ePTFE45)

For both liquid- vapor /gas contactors similar trends for the partial distillate fluxes have been obtained with higher values for ePTFE45 as compared to PES40. Fig.(3.a) shows that for mixtures with low methanol concentration, the water flux increases with increasing Re_G until reaching a limiting value corresponding to the operating constraint on the membrane contactors, whereas the methanol flux remains almost constant over the whole range of Re_G. Fig (3.b) and (3.c) show that both methanol and water fluxes increase with increasing Re_G until the latter reaches a threshold value beyond which any increase of the latter results in the decline of both methanol and water fluxes. This behavior may be explained by the trade off between the reduction of the temperature polarization effect and the increase of the resistance to mass transfer in the membrane pores and the permeate channel, both due to the increase of inert gas flow rate.

4. Conclusions

1. In both feed and permeate channels laminar flow conditions have been established within the operating constraints of the micro-separator.
2. The temperature polarization effect is localized in both the feed and permeate channels and is strongly related to the methanol concentration in the feed and to the hydrodynamic conditions in the respective channels. Whereas, the concentration polarization effect is localised mainly in the permeate channel and is strongly dependent on the inert gas flow rate which constitutes therefore a crucial operating parameter for the micro-separator.
3. Polymeric micro-porous hydrophobic membranes used commonly for microfiltration purposes have been successfully used as liquid- vapor / gas contactors for the new micro-distillation device, and constitute an important design parameter for the reduction of the temperature polarization effect. Indeed for all performed experiments the partial distillate fluxes obtained with PES40 were lower than those obtained with ePTFE45. This behaviour can be attributed among other factors to the higher porosity and pore size[2] of the latter so that the mass transfer is more favored.
4. It is worth pointing out that as compared to the existing membrane distillations processes applied only for water evaporation[3] or for the separation of dilute volatile organic compound-water mixtures[4,5,6,7], the novel micro-separator allows to separate aqueous mixtures with low to high methanol concentration. These results indicate the potential of using micro-structures to reduce the concentration polarization effect into membrane distillation based processes.

1. Further investigations will focus on the determination of the influence of the permeate channel height on the separation performance of the micro-separator.

Nomenclature

Symbol

V	Velocity	m/s
ρ	Mass density	Kg/m^3
μ	Viscosity	Pa.s

Subscript

G	Gas
F	Feed

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