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# Selection of membrane contactors used in a membrane distillation based micro-separator

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In this paper the separation performance of a membrane distillation based micro-separator has been investigated. Particularly the influence of the membrane properties (surface chemistry, material and characteristics) on the distillate quality has been studied. The experimental results showed that the surface chemistry of the membrane with respect to its free surface energy constitutes the main limiting factor for the separation. Indeed, independently of their material and characteristics, the tested oleophobic membranes present lower free surface energy than hydrophobic membranes. Consequently they are more appropriate as non selective liquid-gas contactors for the separation of aqueous mixtures with low to high methanol concentration. Furthermore, it is shown that the impact of membrane characteristics (porosity, thickness, pore size) on the distillate quality (separation factor) is almost negligible since the resistance to mass transfer is concentrated in both feed and gas channels for the investigated micro-separator design. That suggests that the separation factor may be further enhanced by decreasing the feed/gas channel height in the micro-separator.

## 1. Introduction

A membrane distillation based micro-separator (Fig. 1) has been recently fabricated and tested by the authors (Adiche and Sundmacher, 2010) for the separation of aqueous mixtures with low to high methanol concentrations. The 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: 1 mm, channel width: 0.5 mm, and channel length: 342 mm). Each separation plate is imbedded into a slit machined into the respective cover polycarbonate plate (external dimension: 110 mm × 110 mm × 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 preheated liquid mixture of methanol and water 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- or counter-current mode, tangentially to the

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membrane surface. The membrane serves as a non selective 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 and / or convect through the membrane pores. The inert gas sweeps the permeate mixture (distillate) carrying it outside the device.

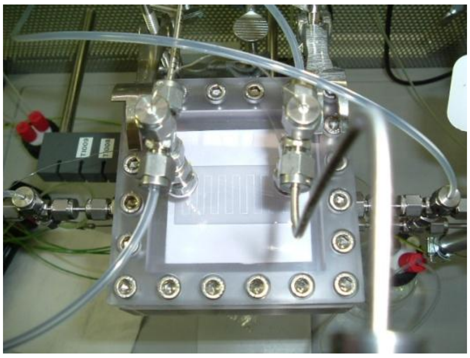
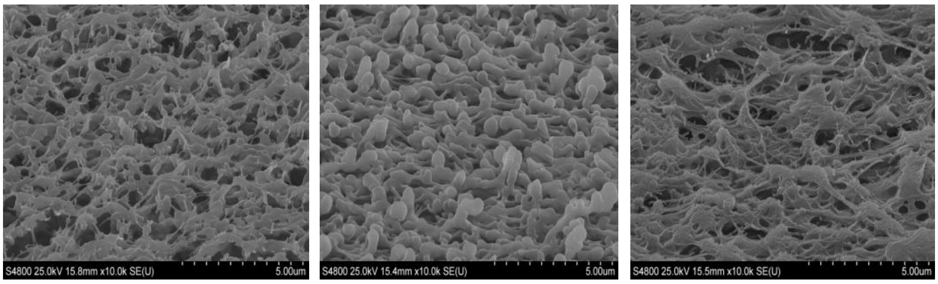


*Figure 1: Micro-separator*

In the previous work (Adiche and Sundmacher, 2010), the separation performance of the device was investigated for different membranes by varying the relevant operating parameters of the process. For all performed experiments, the separation feasibility has been proved. Particularly, the influence of the membrane characteristics on the reduction of the temperature polarization effects and thus on the enhancement of the distillate flux has been established. The following discussion will focus on the influence of membrane material, surface chemistry and characteristics on the distillate quality (separation factor).

## 2. Membrane contactors

The membrane contactors tested in this work are three commercial types of flat sheet polymeric porous membranes commonly used for microfiltration or ultrafiltration and for venting applications (Nunes and Peinemann, 2001). They are made of different materials: (a) polyvinylidene fluoride membrane (PVDF) provided by Millipore (USA), (a) polyethersulfone polymer cast on a non-woven polyester support membrane (PESS) provided by Pall (USA), and an (c) expanded polytetrafluoroethylene membrane on a non-woven polyester support (ePTFE) supplied by Gore (USA). All three membranes are hydrophobic and hence water repellent. Additionally as compared to PVDF membrane, the membranes PESS and ePTFE are oleophobic and are characterized by lower free surface energy obtained by appropriate surface modification techniques. That results in a greater contact angle between these membranes and low surface tension liquids, preventing thereby membrane wetting with such liquids. The tested membranes are characterized among other parameter by their thickness, pore size, porosity (volume fraction void), and water intrusion pressure $LEP_w$. The latter measures the pressure required to wet-out the membrane and constitutes therefore the main constraint for process operation. 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 (LEP) of the membrane for given feed temperature and composition (Garcia-Payo et al., 2000). Fig. 2 illustrates the SEM pictures of the three membrane surfaces obtained by a scanning electron microscope DHS 942 Zeiss (Germany).



*(a)* *(b)* *(c)*

*Figure 2: SEM pictures for: (a) PVDF, (b) PESS, and (c) ePTFE*

Table 1 summarizes the characteristics of the three membranes. The porosity of both PESS and ePTFE membranes was determined by Hg intrusion porosimetry technique whereas all other characteristics were provided by the manufacturer.

*Table 1 Characteristics of the membrane contactors*

| Membrane | Pore size $D_p$ (µm) | Thickness $\delta_m$ (µm) | Porosity $\varepsilon$ (vol %) | $LEP_w$ (bar) |
|---|---|---|---|---|
| PVDF | 0.22 | 114 | 0.75 | 2.0 |
| PESS | 0.20 | 166 | 0.55 | 1.37 |
| ePTFE | 0.45 | 250 | 0.91 | 1.3 |

## 3. Experimental results and discussion

Three sets of experiments have been performed to investigate the influence of the three tested membranes on the separation performance of the micro-separator. For this purpose the experimental set-up developed previously (Adiche and Sundmacher, 2010) has been used to separate a mixture of methanol and water. The distillate flux was determined by measuring the weight of the collected permeate, the membrane effective area, and the time period of the experiment after reaching steady state conditions. The methanol content in the distillate was obtained by means of a gas chromatograph (Agilent 6890 series) using helium as carrier gas and equipped with a flame ionization detector (FID), a thermal conductivity detector (TCD), and a HP Innowax column (L=30 m, ID = 0.25 mm). For all performed experiments, inert gas nitrogen was introduced at room temperature with a flow rate G of 49 ml STP / min (at standard conditions). The feed mixture was introduced at a temperature $T_F$ of 60°C with a flow rate $L_F$ of 8 ml / min. For each membrane, the feed composition has been varied and its operating range corresponding to a dry-state membrane was established by inspecting visually the wettability limit of the latter. The maximum feed composition that could be reached for both ePTFE and PESS was found to be 0.70 kg/kg, whereas that for PVDF could not exceed 0.20 kg/kg. This behaviour can be well explained by the olephobic character of both first membranes as compared to the hydrophobic character of the third one.

Fig. (3) illustrates the separation performance of the micro-separator for the three membranes which corresponds to a separation factor ranging between 2.5 and 5 over the investigated feed composition range (Adiche and Sundmacher, 2010).

It is shown that for a given feed composition, the distillate compositions for the three membranes are almost identical. This result indicates that neither the membrane material nor its physical characteristics (pore size, porosity, and thickness) have any influence on the separation factor. In other words that means that the diffusion in both the feed and gas channels is the rate limiting step for mass transfer in the investigated micro-separator.

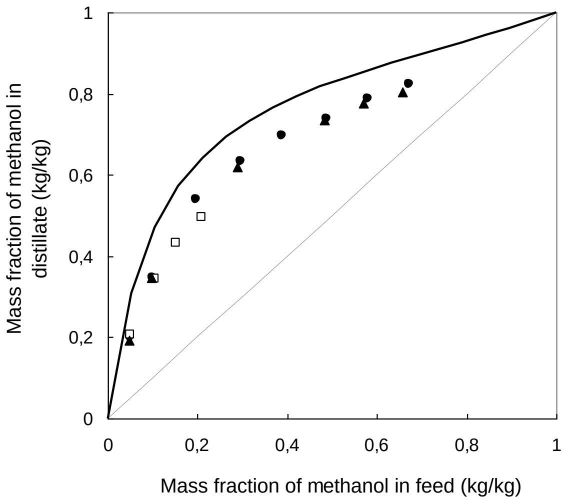



*Figure 3: Impact of a variation of the feed composition on the distillate composition, $T_F$= 60°C, $L_F$ =8 ml/min, G =49 ml STP/min, (●) PESS, (▲) ePTFE, (□) PVDF, continuous line: LVE isotherm of methanol-water mixture at 60 °C*

Fig. (4) shows that the partial distillate flux of methanol rises almost linearly with an increase of the mass fraction of methanol in feed. This behaviour is similar for the three tested membranes and can be explained by the effect of the methanol concentration on the respective partial vapor pressure resulting therefore in an enhancement of the driving forces across the membrane (Lawson and Lloyd, 1997). On the other hand it is shown in Fig. (5) that increasing the methanol content in the feed results in a slight decrease of the partial distillate flux of water for both PESS and ePTFE, while the value of partial distillate flux of water corresponding to PVDF increases almost exponentially within the allowed range of feed composition contributing therefore to a slightly smaller distillate composition (Fig. 3) as compared to the other membranes.

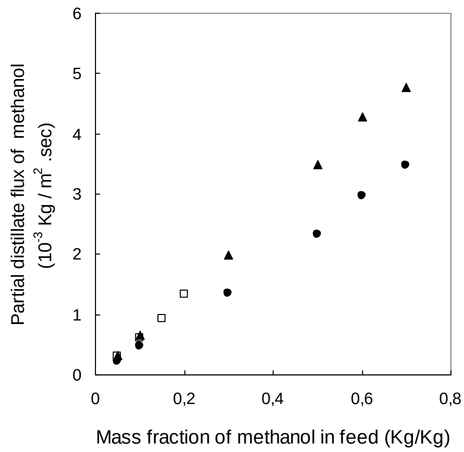



*Figure 4: Impact of a variation of the feed composition on the partial distillate flux of methanol, $T_F$ = 60ºC, $L_F$ =8 ml/min, G =49 ml STP/min, (•) PESS, (▲) ePTFE, (□) PVDF*

The different behaviour observed between PVDF and both PESS and ePTFE with respect to the partial distillate flux of water may be attributed to the difference of membrane surface chemistry.

On the other hand, ePTFE and PVDF present similar mass transfer performance (for both partial distillate fluxes) with a much higher distillate flux than that obtained with PESS. Accordingly a direct correlation between the total/partial distillate flux and the combination of the membrane characteristics $\frac{D_p.\varepsilon}{\delta_m}$ (Lawson and Lloyd, 1997) could be established.

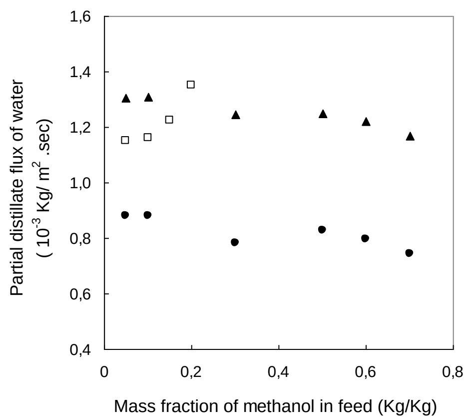



*Figure 5: Impact of a variation of the feed composition on the partial distillate flux of water, $T_F$ = 60 ºC, $L_F$ =8 ml/min, G =49 ml STP/min, legend of Figure 4*

## 4. Conclusions

1. As compared to hydrophobic membrane PVDF, oleophobic membranes PESS and ePTFE are more appropriate as non selective liquid-gas contactors for the separation of aqueous mixtures with low to high methanol concentration by using the developed micro-separator.
2. Independently of the membrane material and physical characteristics both oleophobic membranes PESS and ePTFE allowed the separation of aqueous mixtures with a methanol concentration ranging from 5 to 70 wt. %.
3. For the investigated micro-separator design, the experimental results showed that the membrane characteristics do not have any influence on the separation factor and that the concentration polarization effect is predominating.

Further investigations will focus on the determination of the influence of decreasing the feed/gas channel height on the separation factor in the developed micro-separator.

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