A Capillary Module for Membrane Distillation Process*

M. GRYTA, M. TOMASZEWSKA, and A. W. MORAWSKI

Department of Water Technology and Environmental Engineering, Institute of Chemical Technology and Environmental Engineering, Technical University of Szczecin, PL-70-322 Szczecin, Poland e-mail: margryta@mailbox.tuniv.szczecin.pl

Received 19 May 2000

The influence of a capillary module design and its operating parameters on the efficiency of the membrane distillation (MD) process has been studied. It was demonstrated that a uniform arrangement of the capillary membranes over the cross-section of module at 0.33 membrane packing density ensured appropriate conditions of membranes cooling. This allowed to obtain a high value of the driving force for the mass transfer, hence the MD process proceeded with a high efficiency. The most favourable operating conditions of MD module were obtained with membranes arranged in a form of braided capillaries. The performance of membranes arranged parallel with their distribution alongside module controlled by sieve baffles was also satisfactory.

Several membrane processes such as reverse osmosis (RO), nanofiltration (NF), ultrafiltration (UF), microfiltration (MF), electrodialysis (ED), and pervaporation (PV) have been successfully applied in the industrial processes [1]. The implementation of membrane distillation for the production of ultrapure water and treatment of saline wastewater on a commercial scale is a subject of ongoing research [2—5].

Membrane distillation (MD) is an evaporation process of volatile components from solution (feed) across the pores of nonwetted hydrophobic membrane (Fig. 1). The driving force for the mass transfer is a vapour pressure difference occurring on both sides of the membrane, which depends on both the temperature (T_1, T_2) and the composition of solutions (c_1, c_2) in the layers adjacent to the membrane. The mass flux of the stream of permeate flowing through the membrane is described by the equation [4, 5]

$$N = \frac{\varepsilon M D_{\rm E} P}{\chi s R T_{\rm m}} \ln \frac{P - p_{\rm D}}{P - p_{\rm F}} \tag{1}$$

where $p_{\rm F}$ and $p_{\rm D}$ are the partial pressures of the water vapour at temperatures T_1 and T_2 , respectively. Owing to the mass transfer and heat conduction across the membrane material, the temperature of boundary layer on the feed side is lower, whereas on the distillate side it is higher than that of the bulk. This phenomenon is called the temperature polarization, which diminishes the magnitude of the mass stream flowing through the membrane. The temperatures T_1 and T_2 essentially depend on the values of the convective heat transfer coefficients α in MD module [5]

$$T_{1} = \frac{\frac{\lambda_{\rm m}}{s} \left(T_{\rm D} + \frac{\alpha_{\rm F}}{\alpha_{\rm D}} T_{\rm F} \right) + \alpha_{\rm F} T_{\rm F} - N H_{T_{\rm m}}}{\frac{\lambda_{\rm m}}{s} + \alpha_{\rm F} \left(1 + \frac{\lambda_{\rm m}}{\alpha_{\rm D} s} \right)} \qquad (2)$$

$$T_{2} = \frac{\frac{\lambda_{\rm m}}{s} \left(T_{\rm F} + \frac{\alpha_{\rm D}}{\alpha_{\rm F}} T_{\rm D} \right) + \alpha_{\rm D} T_{\rm D} + N H_{T_{\rm m}}}{\frac{\lambda_{\rm m}}{s} + \alpha_{\rm D} \left(1 + \frac{\lambda_{\rm m}}{\alpha_{\rm F} s} \right)} \qquad (3)$$

The magnitude of the α coefficients is significantly affected by the design of membrane module and its operating parameters (temperatures and flow rates of feed and distillate).

The capillary membrane module is the most appropriate configuration in the applications on a commercial scale since it is both compact and easily manufactured. The capillary membranes are generally arranged in a parallel form. A primary limitation of this design arises from difficulties encountered in the arrangement of capillaries alongside the module. The capillaries themselves align inside the housing in a rather random mode, what creates regions with different packing density. Therefore, the outside surface of capillaries is cooled by the distillate in a nonuniform manner. The local temperature T_2 increases thus the driving force for the mass transfer decreases and

^{*}Presented at the 27th International Conference of the Slovak Society of Chemical Engineering, Tatranské Matliare, 22—26 May 2000.



Fig. 1. Heat and mass transfer in MD.

reduces the efficiency of the module. Membranes arranged in twisted or braided form in the housing improve the hydrodynamic conditions in the module, and as a consequence enhance its efficiency [3]. These results were obtained for large modules with the area of $50-100 \text{ m}^2$. The application of such a large module may be ineffective, particularly at the initial periods of implementation of this technology (pilot-scale installation). For that reason the studies were undertaken on the design of smaller MD modules.

EXPERIMENTAL

The studies of membrane distillation were carried out employing capillary modules made from polypropylene membranes with an inside/outside diameter of 1.8 mm/2.6 mm. The membranes were characterized by the pore size with a maximum and nominal diameters of 0.6 μ m and 0.2 μ m, respectively, and the porosity of 73 %. The membrane modules were assembled in the MD installation in a vertical position. The feed and distillate streams flow cocurrently from the bottom to the upper part of module. The feed circulates inside the capillary membranes. The distillate temperature at the entrance of the module was kept at a constant level of 293 K, whereas the feed temperature was varied in the range of 333—363 K. The flow rates within the range $0.05 - 1 \text{ m s}^{-1}$ were used during the measurements. An aqueous solution of NaCl with a mass fraction of 0.1 % was used as a feed.

RESULTS AND DISCUSSION

The application of a higher degree of capillary



Fig. 2. Dependence of the permeate flux on feed temperature for modules with different membrane packing density (mpd). Parallel capillary membranes – arrangement in a random way. Modules symbols: □ M1, 0 M2, ■ M3,
M4. Modules M1—M3 mpd = 0.4, module M4 mpd = 0.07. Process parameters: T_{Din} = 293 K, v_F = 0.11 m s⁻¹, v_D = 0.03 m s⁻¹ (M1—M3) and v_D = 0.01 m s⁻¹ (M4).

membrane packing results in a considerable resistance of distillate flow. Moreover, cooling of the membrane surfaces is interior. The packing density within the range of 0.4-0.5 is regarded as the optimum [3, 6]. The experimental results from MD studies using laboratory-scale modules equipped with the membranes arranged in a parallel mode with the working area of 0.014 m^2 are presented in Fig. 2. Different diameters of housings for the same number of capillaries in the module resulted in various degrees of packing density: 0.4 for M1-M3 modules and 0.07 for M4 module. In these modules the membranes were arranged in a random way. This implied differences in the spatial arrangement of membranes for M1-M3 modules, *i.e.* a part of the membranes was in the direct contact. The differences of the permeate flux (N) for modules M1—M3 shown in Fig. 2 indicate that this arrangement influences the magnitude of obtained stream. In the case of M4 module the membranes with a low packing density were arranged in a certain distance from each other, which enabled an adequate cooling of capillary surfaces. Therefore, the temperature polarization was diminished, and as a consequence, a significant increase of the module efficiency was achieved.

The adequate cooling of the membrane surfaces by the use of low packing density leads to excess increase of the MD module dimensions. However, the appropriate conditions of cooling in the module with a higher packing density can be achieved through the membranes arrangement in a controlled way.

Three modules (M5—M7) with an effective length of 1 m, a housing diameter of 0.025 m, and a packing



Fig. 3. Capillary modules with a different arrangement of membranes within the housing. 1. Housing; 2. membrane fixations; 3. capillary membrane; 4. sieve baffle; 5. braided membranes. M5 – module with braided membranes; M6 – module with parallel membranes fixed in sieve baffles; M7 – module with membranes arranged in a random way.

density of 0.33 have been constructed (Fig. 3). In the module M5, the membranes were positioned in every second mesh of six sieve baffles, arranged across the housing within 0.15 m. A parallel bundle of braided capillaries (three membranes in a braid) was assembled in the M6 module. The braids with a membrane packing density of 0.33 filled the entire cross-section of housing, which ensured a uniform arrangement of membranes in the module. For a comparison, in the M7 module, the bundle of parallel membranes was arranged in a random way. This module construction resulted in a twofold lower efficiency than that of the M5 and M6 modules (Fig. 4). A uniform arrangement of the membranes in these modules allows to obtain the permeate flux at a level of 450 $dm^3 m^{-2} day^{-1}$ for the feed inlet temperature of 363 K and its flow rate of 0.31 m s^{-1} . Improved results were obtained with a module consisting of braided membranes since their shape acted as a static mixer. This resulted in the enhanced turbulence of distillate stream and increase of heat transfer coefficient. As a consequence of the temperature polarization, T_2 was closer to the T_D value, thus the driving force increased. It is noteworthy to mention that the particular membranes in the braid are in the form of curved tube. A fluid flowing in the curved tube is influenced by centrifugal force, and consequently the additional rotary current crosswise to the flow direction, called "Dean flow" is formed [7]. This phenomenon modifies the conditions of heat and mass transfer, the temperature polarization on the feed side is reduced, and as a consequence the efficiency of MD module increases [8].

The efficiency of the module increased with the



Fig. 4. The influence of feed temperature and the mode of membrane arrangement in a capillary module on the permeate flux. ■ M5 module with braided membranes;
O M6 module with parallel membranes fixed in sieve baffles; □ M7 module with parallel membranes arranged within the housing in a random way. T_{Din} = 293 K, v_D = 0.056 m s⁻¹, v_F = 0.31 m s⁻¹.

flow rate $v_{\rm F}$ (Fig. 5). For instance, the enhancement of $v_{\rm F}$ from 0.3 m s⁻¹ to 0.6 m s⁻¹ (at $v_{\rm D} = 0.72$ m s⁻¹) caused the increase in the efficiency from 405 dm³ m⁻² day⁻¹ to 730 dm³ m⁻² day⁻¹. A further increase of the flow rate of feed resulted in a small growth in the permeate flux. The observed dependence of permeate flux on the flow rate is essentially due to two reasons. Firstly, with an increase of flow rate, the values of the heat transfer coefficient α rise, and the negative



Fig. 5. The effect of the flow rate of streams in a module with braided membranes (M5) on the permeate flux. $T_{\rm Fin} = 353$ K, $T_{\rm Din} = 293$ K, $\blacksquare v_{\rm D} = 0.72$ m s⁻¹, $\bigcirc v_{\rm D} = 0.38$ m s⁻¹, $\Box v_{\rm D} = 0.26$ m s⁻¹.

influence of temperature polarization decreases. The value of α equal to 5000 W m⁻² K⁻¹ is considered a threshold value, above this value the effect of temperature polarization may be neglected [9]. This value was achieved at $v_{\rm F}$ of about 0.6 m s⁻¹. Secondly, the increase in the flow rate caused that the outlet temperatures of streams were closer to their inlet values (Fig. 6), therefore the driving force for mass transfer increased through the module. This effect was particularly large for the feed and slightly smaller for the distillate. At a high distillate flow rate (0.72 m s^{-1}) , the value of $T_{\rm Dout}$ increased only by 1.5 K for the feed flow rate growing from 0.3 m s^{-1} to 0.96 m s^{-1} . For a smaller distillate flow rate of 0.26 m s^{-1} , this change amounted to 4 K. Such insignificant variations of distillate temperature with the increase of v_{D} resulted in a slight increase of permeate flux (Fig. 5). The achieved optimum values of the flow rate in the studied MD modules are twofold higher than those reported in work [3]. Thus, it can be concluded that the optimum flow velocity should be determined individually for each type of MD module.

CONCLUSION

The efficiency of the MD capillary module is significantly affected by the mode of the membranes arrangement within the housing. A traditional construction based upon the fixation of a bundle of parallel membranes solely at their ends results in that the membranes arrange themselves in a random way. This creates unfavourable conditions of cooling of the membrane surface by the distillate, hence, the module efficiency is reduced due to enhancement of temperature polarization.

Arrangement of membranes in such a way to ensure



Fig. 6. The effect of the flow rate of feed and distillate streams on their outlet temperature for M5 module. $\circ T_{\text{Fout}}$ for $v_{\text{D}} = 0.26 \text{ m s}^{-1}$, $\Box T_{\text{Fout}}$ for $v_{\text{D}} = 0.72 \text{ m s}^{-1}$, \bullet T_{Dout} for $v_{\text{D}} = 0.26 \text{ m s}^{-1}$, $\blacksquare T_{\text{Dout}}$ for $v_{\text{D}} = 0.72 \text{ m}$ s^{-1} , $\triangle T_{\text{Fin}} = 353 \text{ K}$, $\blacktriangle T_{\text{Din}} = 293 \text{ K}$.

a uniform distribution over a cross-section of module (braided membranes or supported by sieve baffles alongside module) allowed to achieve over 100 % increase of efficiency. The module efficiency is also dependent on the flow rate. The optimal value of the flow rate for studied modules amounts to 0.6—0.8 m s⁻¹ and 0.4—0.7 m s⁻¹ for feed and distillate, respectively.

Acknowledgements. This work was supported by the Polish State Committee for Scientific Research.

SYMBOLS

с	concentration	$ m mol~m^{-3}$	
D_{E}	effective diffusion coefficient	$\mathrm{m}^2~\mathrm{s}^{-1}$	
$d_{ m h}$	hydraulic diameter	m	
Η	vapour enthalpy	${ m J~kg^{-1}}$	
L	flow channel length	m	
M	molar mass	$\rm kg \ mol^{-1}$	
N	permeate flux	${\rm kg} {\rm m}^{-2} {\rm s}^{-1}$	
P	total pressure	$ m N~m^{-2}$	
p	partial pressure of saturated vap	artial pressure of saturated vapour over the	
	solution	${ m N}~{ m m}^{-2}$	
Q	heat flow	W	
R	gas constant	$\rm J~mol^{-1}~K^{-1}$	
s	membrane thickness	m	
T	temperature	Κ	
v	flow rate	${ m m~s^{-1}}$	
α	convective heat transfer coefficie	$nt W m^{-2} K^{-1}$	
ε	membrane porosity		
λ	thermal conductivity coefficient	$\mathrm{W}~\mathrm{m}^{-1}~\mathrm{K}^{-1}$	
χ	membrane pore tortuosity		

Subscripts

1 polarization boundary layer on feed side

- 2 polarization boundary layer on distillate side
- D distillate
- F feed
- m membrane
- in input
- out output

REFERENCES

- 1. Scott, K., *Handbook of Industrial Membranes*. Elsevier Advanced Technology, Kidlington, UK, 1995.
- 2. Carlsson, L., Desalination 45, 221 (1983).
- Schneider, K., Hölz, W., Wollbeck, R., Ripperger, S., J. Membr. Sci. 39, 25 (1988).

- Gryta, M., Tomaszewska, M., and Morawski, A. W., Sep. Purif. Technol. 11, 93 (1997).
- Gryta, M. and Tomaszewska, M., J. Membr. Sci. 144, 211 (1998).
- Serra, C., Moulin, Ph., Rouch, J. C., Clifton, M. J., and Aptel, Ph., in *Proceedings of Euromembrane* '95, Vol. 2, p. II43. University of Bath, 18—20 September 1995.
- Ophoff, J., Vos, G. S., Racz, I. G., and Reith, T., in Proceedings of Euromembrane '97, p. 400. University of Twente, 23—27 June 1997.
- Gryta, M. and Tomaszewska, M., *Inż. Chem. Proc.* 20, 221 (1999).
- Schofield, R. W., Fane, A. G., and Fell, C. J. D., J. Membr. Sci. 53, 173 (1990).