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Modelling of Hybrid Method for VOC Removal from Process Wastewater: Distillation and Hydrophilic Pervaporation

Botond Szilágyi[1], Do Thi Huyen Trang[1], Dániel Fózer[1], Asmaa Selim[1,2], Enikő Haáz[1],
András József Tóth[1,3*]

[1] Environmental and Process Engineering Research Group, Department of Chemical and Environmental Process Engineering, Faculty of Chemical Technology and Biotechnology, Budapest University of Technology and Economics, H-1521 Budapest, P. O. B. 91, Hungary
[2] Department of Chemical Engineering and Pilot Plant, National Research Centre, 33 El Buhouth Street, 12622 Cairo, Egypt
[3] Institute of Chemistry, University of Miskolc, H-3513 Miskolc, Egyetemváros, P. O. B. 21, Hungary
[*] Corresponding author, e-mail: ajtoth@envproceng.eu

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Abstract

The study is motivated by the industrial problem from pharmaceutical industry, which is ethanol and methanol removal from process wastewater. To complete this goal hybrid method is investigated and optimized. Two distillation columns are sufficient for separation of alcohol-water mixture. Suitable water can be purified as bottom product of first column. Ethanol and methanol purification is achieved with combination of second distillation column and pervaporation. The target of this research is to rigorously model and optimize the separation of water-ethanol-methanol ternary mixture in professional flowsheet simulator environment. The minimal sufficient membrane transfers area and number of minimal theoretical stages of the columns are determined. Cost estimation is also investigated according to Douglas methodology. Considering the simulation and economic results it can be determined that, the hybrid configuration is suitable for separation of ternary mixture in 99.5 weight percent purity.

Keywords

ethanol and methanol removal, process wastewater, hydrophilic pervaporation, mathematical modelling, computer simulation

1 Introduction

Separation of water and alcohol mixtures can be considered well-known example of hydrophilic pervaporation operation in chemical and related industries [1, 2]. Although more references reported that Pervaporation (PV) method is applied for separating ethanol (EtOH) / water and methanol (MeOH) / water binary mixtures, but there is nearly no study and widespread technology for separating water-ethanol-methanol ternary mixture. Ethanol and methanol can be considered as Volatile Organic Compound (VOC).

The removal of organic compounds from aqueous solutions is particular interest for fermentation, treatment of wastewater water and recycling processes [3–9].

The target of this study is to optimize the separation of water-ethanol-methanol ternary mixture with combination of distillation and hydrophilic pervaporation method in professional flowsheet environment.

Ethanol forms minimal boiling azeotropic mixture with water. EtOH content above 96 weight% cannot be achieved with conventional distillation techniques [7, 10]. In contrasts, methanol and water is zeotropic mixture. Hybrid method of distillation and hydrophilic pervaporation has proven to be economically viable for separation of binary azeotropic mixtures [11]. If the azeotropic composition can be approached with distillation, then the distillate product (D) can be further purified applying PV. Hydrophilic PV proved to effective solution for separation of ethanol/water binary mixture in contrast to distillation [7].

Pervaporation is a relatively new technology, where the mixture to be treated is vaporized at low pressure on the permeate side of the membranes and the separation of the mixtures progresses by preferential sorption and diffusion phenomenon of the desired component through the dense membranes [7]. Vacuum pump on the permeate side can maintain the low vapor pressure [7, 12, 13]. Pervaporation is capable for the separation of many organic aqueous systems [14–17].

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The unit operation is mainly used for dehydration of organic compounds from its aqueous mixtures [18–20], removal of low concentration organics from water [21–23] and organic-organic separation [24–26]. Depending on the main permeating compound two main areas of pervaporation process can be classified: hydrophilic and organophilic pervaporation [27–30].

This unit operation has the specialties such as no-pollution and energy-saving, simply actualization and high separation which are difficult to obtain by other conventional technologies [7].

The pervaporation measurements can be achieved in continuous and batch configuration [31]. Table 1 shows the comparison of both solution.

Pervaporation can have advantages over distillation method because of the capability to separate azeotropic mixtures and its lower energy demand. Generally, distillation can be used to remove VOCs from water and pervaporation is also suitable for this problem [32]. Furthermore, the pervaporation operation has usually lower operating temperatures and the separation does not require an extra added component (such e. g. azeotropic and/or extractive distillation) [33–40]. As it can be determined, pervaporation is considered as the competitive separation alternative of distillation [7].

2 Material and methods

In the pharmaceutical sector it is an important problem that methanol and ethanol should be separated from aqueous mixture. The investigated pharmaceutical process wastewater (PWW) had the following composition: 20 weight percent (wt%) methanol, 20 wt% ethanol and 60 wt% water. 1000 kg/h PWW must be treated and the product purities are 99.5 wt%. Continuous operation was used for separation because of the large amount of initial process wastewater and the products goal, which is maximal enrichment quality (see Table 1). ChemCAD professional flowsheet simulator was applied for the investigation of ternary mixture separation. UNIQUAC thermodynamic model was used in the case of SCDS distillation column. Table 2 shows the binary UNIQUAC parameters.

Table 1 Continuous versus batch configuration of pervaporation [31]

Continuous pervaporation	Batch pervaporation
Primary product goal: maximal enrichment/extract quality	Primary product goal: maximum recovery
No (significant) pre-treatment of feed required	Considerable pre-treatment of raw material required
Retentate holds full value during post-hydrophilic pervaporation processing	Retentate considered as low-value (by-)product
Abundant availability of (liquid) raw material	Restricted availability of (liquid) raw material

The fundamental (Model I) and the exponential Rautenbach model (Model II) were also applied for modelling of hydrophilic pervaporation [15]. Equation (1) shows the equation of fundamental Rautenbach model.

$$J_i = 1\Big/\left(1+\left[\bar{D}_i\Big/\left(Q_0\times p_{i0}\times\bar{\gamma}_i\right)\right]\right)\times\left(\bar{D}_i/\bar{\gamma}_i\right)\times\left(\left[p_{i1}-p_{i3}\right]\Big/p_{i0}\right) \tag{1}$$

The following equation introduces the improved Rautenbach model [41]:

$$\begin{aligned} J_i = 1\Big/&\left(1+\left\{\left[\bar{D}_i\times\exp\left(B\times x_{i1}\right)\right]\Big/\left(Q_0\times p_{i0}\times\bar{\gamma}_i\right)\right\}\right)\\ &\times\left[\bar{D}_i\times\exp\left(B\times x_{i1}\right)\right]\Big/\bar{\gamma}_i\times\left(\left(p_{i1}-p_{i3}\right)\Big/p_{i0}\right)\end{aligned} \tag{2}$$

The PV model verification can be taken with objective function (OF), which is minimized the difference of the modelled and the measured values [42].

$$\mathrm{OF}=\sum_{i=1}^{n}\left(\left(J_{i,measured}-J_{i,modelled}\right)\Big/J_{i,measured}\right)^2 \tag{3}$$

Table 3 and Table 4 show the estimated parameters of the mentioned semi-empirical models with Sulzer PERVAP™ 1510 membrane. The experimental conditions can be found in the paper of Valentínyi et al. [15].

It can be seen Model II is much more appropriate for description of pervaporation than Model I in both cases.

Hybrid method has been selected for separation of PWW. The flowsheet can be seen in Fig. 1.

The optimized parameters of complex separation processes were: reflux ratio, number of theoretical stages, feed tray number and effective membrane transfer area (*A*). The minimized Total Annual Cost (TAC) must be found as the objective function of the model optimization. The methodology of Tóth [7, 11] and Douglas equations [43] were applied for cost estimation with Marshall&Swift index of 2018 (M&S = 1638.2 [44]), while pump costs were determined by industrial data [7]. Membrane area-price function was determined on industrial data too and applied

Table 2 UNIQUAC parameters of investigated binary pairs

I	*J*	Sub Type	$U_{ij}-U_{jj}$	$U_{ji}-U_{ii}$	*p* range [kPa]	*T* range [°C]
Water	MeOH	VLE	−10.377	95.259	0.1 – 100	25 – 100
Water	EtOH	VLE	232.01	50.88	0.1 – 100	20 – 100
MeOH	EtOH	VLE	−181.286	247.378	0.5 – 100	25 – 100

Table 3 Estimated parameters and minimized objective functions in the case of Model I [15]

	Water	Ethanol
	binary mixture	
Transport coefficient [kmol/m^2 × h]	3.34E-04	1.89E-07
Activation energy [kJ/kmol]	80042	95414
OF [-]	0.93	5.03

Table 4 Estimated parameters and minimized objective functions in the case of Model II [15]

	Water	Ethanol
	binary mixture	
Transport coefficient [kmol/m^{2}h]	2.02E-04	1.93E-05
Activation energy [kJ/kmol]	77877	128572
Exponential parameter [-]	2.63	8.68
OF [-]	0.14	2.25

for the calculation of the investment costs of membrane modules [5, 45]. Two and half years were taken as membrane depreciation time, because membranes should be generally replaced in approximately every 2–5 years [7]. 10-year amortization of investment cost was assumed for the total cost estimation [45].

First step, the initial process wastewater was entered into the Column I in the case of hybrid method, where suitable treated water can be got as bottom product (*W*). The alcohol-rich intermediate distillate (*D*) was purified further in Column II. Sufficient ethanol content can be reached using hydrophilic pervaporation. Retentate product (*R*) contains ethanol in 99.5 wt% and the permeate stream (*P*) was mixed into the feed stream of distillation Column II. The sufficient methanol concentration (99.5 wt%) can be received in distillate product of Column II.

Permeate and feed pressures were the following, 0.008 bar and 3 bar. The used feed temperature in membrane modules was 70 °C. Additional apparatuses were also needed for pervaporation process [7, 45]. The pressure and temperature had to be increased for the operational level prior to the first membrane unit with pump and heat exchanger, because the feed (*F*) had atmospheric conditions, 1 bar and 20 °C. Retentate stream was reheated after each membrane unit by further heat exchangers [46], except for the last module. Permeate flows leaving the pervaporation apparatuses were collected and condensed with cooler. At last, post coolers and valves decreased again in atmospheric pressure and temperature of Water, Ethanol, and Methanol products [36].

Before computer simulation, the distillation solution of Column I was experimentally verified. The separation process was examined with laboratory distillation column.

The main parameters of the column were the followings: structured packing with diameters of 0.4 m. The laboratory column had 10 number of theoretical stages based on measurement carried out by binary methanol-water mixture. Fig. 2 shows the laboratory experimental column.

The mixture was entered into the middle of the tower. The apparatus heating was controlled with a 300 W heating basket. The alcohol concentration of the feed and products were measured with Shimadzu GC2010Plus+AOC-20 autosampler gas chromatograph with a CP-SIL-5CB column connected to a flame ionization detector. EGB HS 600 Headspace apparatus was applied for sample preparation. The water concentration was measured with Hanna HI 904 coulometric Karl Fischer titrator [47–49].

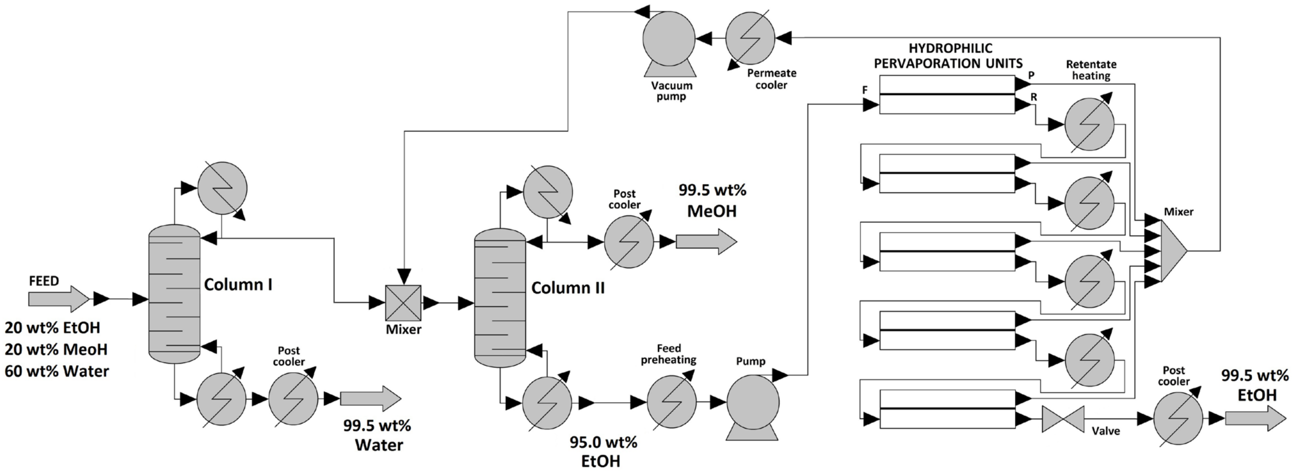

Fig. 1 Flowsheet of water-ethanol-methanol ternary mixture separation

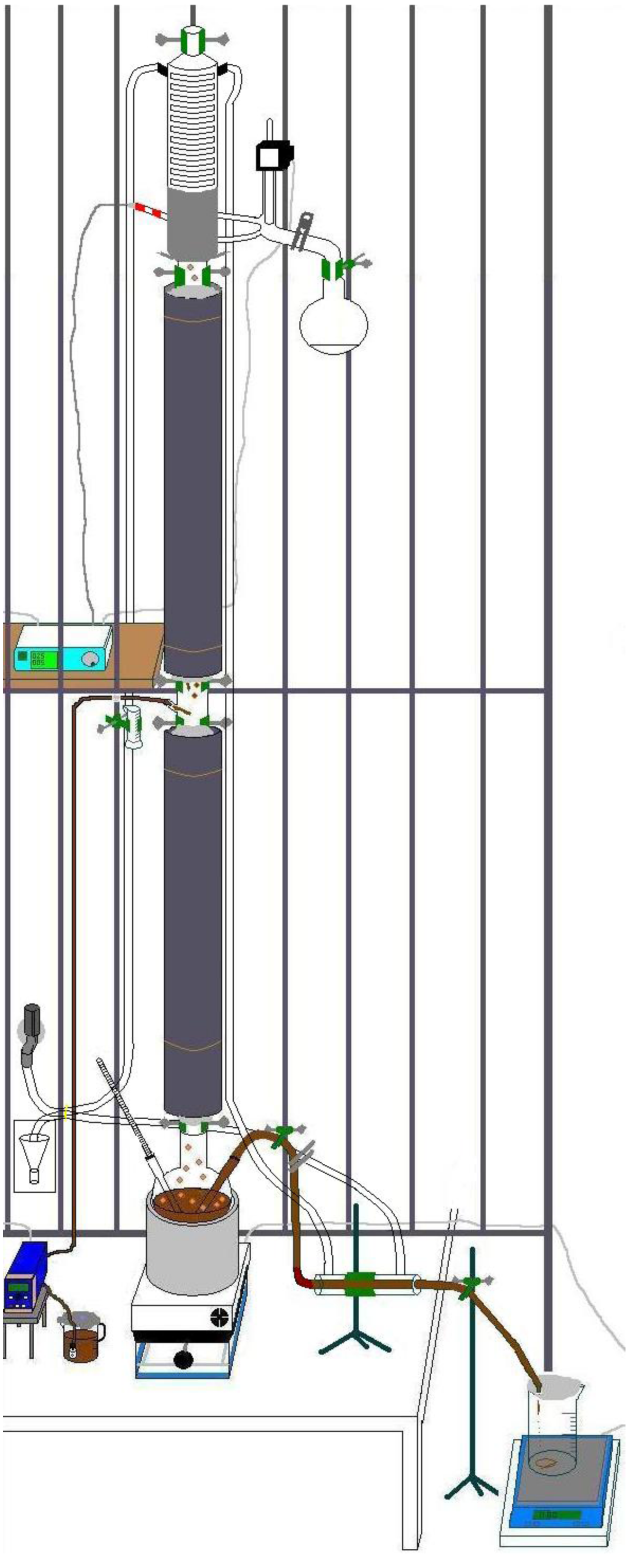

Fig. 2 Laboratory distillation column [11]

3 Results and discussion

3.1 Experimental verification of distillation separation

Table 5 shows the simulated and measured results of first distillation column (Column I). It can be seen the comparison presents the accuracy. The reflux ratio was 5.

Table 5 Comparison of modelling and experimental data for ternary mixture with Column I

	Mixture	Modelling results		Experimental results	
	feed	*D*	*W*	*D*	*W*
EtOH [wt%]	20	47.4	0.3	47.3	0.2
MeOH [wt%]	20	47.4	0.2	47.6	0.2
Water [wt%]	60	5.3	99.5	5.1	99.6
Stream [kg/h]	0.5	0.21	0.29	0.19	0.31
T [°C]	20	70.2	99.1	70.0	99.3

3.2 Modelling results of flowsheet simulator

The optimized results of simulations with distillation processes are listed in Table 6. It can be stated, the purity requirements (99.5 wt%) can be achieved using the hybrid method (see Fig. 1 too).

Table 7 introduces the optimized modelling results of hydrophilic pervaporation: membrane surface area, input and output streams of method. It can be concluded the basic Rautenbach model (Model I) underestimates the values in all cases.

It can be stated, the process design needs the evaluation of the heat demands at the different separation steps [7, 45]. Table 8 includes the calculated heat duties of the hybrid method. It can be concluded that the reboiler of distillation columns have the highest heating requirement of the method.

3.3 Cost estimation

The conceptual design of an industrial tasks takes a small part of the project costs but recommends a huge cost reduction opportunity for the whole project [7, 45], therefore the investigated method should be investigated also from an economic point of view. Table 9 shows the main cost elements of the method.

Table 6 Modelling results of distillation columns

	Column I	Column II
Feed stream [kg/h]	1000	420
EtOH conc. in the feed [wt%]	20	47.1
MeOH conc. in the feed [wt%]	20	47.4
Reflux ratio [-]	5	17
Number of total theoretical stages [-]	20	30
Feed tray number [-]	10	15
EtOH conc. in the distillate [wt%]	48.6	0.5
MeOH conc. in the distillate [wt%]	48.9	99.5
Distillate temperature [°C]	69.5	64.2

Table 7 Modelling results of hydrophilic pervaporation

		Model I	Model II	Deviation [%]
EtOH HPV	A_{PV} [m^2]	60	65	8.3
	F [kg/h]	218.4	220.2	0.8
	P [kg/h]	12.7	13.2	3.6
	R [kg/h]	204.4	207.0	1.3

Table 8 Calculated heat duties of hybrid separation method

Calculated heat duties		$Q_{Heating}$ [MJ/h]	$Q_{Cooling}$ [MJ/h]
Distillation	Reboiler	4150	
	Condenser		−4060
	Post cooler		−320
Pervaporation	Feed preheating	240	
	Retentate heating	330	
	Permeate cooler		−250
	Post cooler		−90

Table 9 Cost elements of hybrid separation method

	Investment cost		Operating cost		TAC
10 years amortization	1000$/ year	%	1000$/ year	%	1000$/ year
Distillation column	18.8	14	-	-	18.8
Heat exchangers	31.1	23	229.2	79	260.3
Membrane modules	81.2	60	23.2	8	104.4
Permeate cooling	4.1	3	37.4	13	41.5
Pumps	0.1	<0.5	0.3	<0.5	0.4
Total	135.4		290.1		425.5

It can be determined that the highest part of install cost is membrane modules and the utility cost of heat exchangers are the most significant part of total annual cost. This statement is consistent with other hybrid methods [7, 41].

4 Conclusions

The combination of distillation and pervaporation method is investigated in flowsheet environment. Semi-empirical models are used for modelling of pervaporation and the separation conditions of first distillation column are also verified with laboratory experiment. It can be concluded water-ethanol-methanol ternary mixture can be separated into pure components with the selected unit operations. The goal composition, which is 99.5 wt% in every product cases can be reached. The presented method is considered suitable for industrial applications.

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Nomenclature

A	Membrane transfer area [m^2]
B	Constant in Model II [-]
D	Distillation product
$\bar{D}_i$	Transport coefficient of component i [kmol/(m^2 × h)]
F	Feed
i	Component number
j	Component number
J_i	Partial flux [kg/(m^2 × h)]
P	Permeate
p_{i0}	Pure i component vapour pressure [bar]
p_{i1}	Partial pressure of component i on the liquid phase membrane side [bar]
p_{i3}	Partial pressure of component i on the vapor phase membrane side [bar]
Q	Heat duty [MJ/h]
Q_0	Permeability coefficient of the porous support layer of the membrane [kmol/(m^2 × h × bar)]
R	Retentate
T	Temperature [°C]
x_{i1}	Concentration of component i in the feed [m/(m%)]
W	Bottom product

Abbreviations

EtOH	Ethanol
HPV	Hydrophilic pervaporation
MeOH	Methanol
OF	Objective function
PV	Pervaporation
PWW	Process wastewater
TAC	Total Annual Cost [1000$/year]
VLE	Vapor-Liquid Equilibrium
VOC	Volatile organic compounds

Greek letters

$\bar{\gamma}_i$ Average activity coefficient of component i

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