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New horizon for the membrane separation: Combination of organophilic and hydrophilic pervaporations

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ABSTRACT

The work is motivated by an industrial separation problem, that is, isobutanol removal from aqueous mixture. To complete this goal a hybrid separation system of organophilic–hydrophilic pervaporation system is designed applying Sulzer PERVAP™ 4060 and 1510 membranes and investigated to obtain information about the separation of isobutanol–water mixture. The aim of research is to rigorously model and optimize this novel hybrid process. Permeation fluxes, permeances, selectivities and separation factors are experimentally determined for this hybrid system. The pervaporation separation index (PSI) and selectivity data are compared with those of other membranes published in the literature and it is found that PERVAP™ 4060 has the highest PSI value and its other parameters are also among the best. Our experimental data are evaluated with the pervaporation model of our improvement and it is found that the model can be applied also for both organophilic and hydrophilic pervaporations. The hybrid separation system is rigorously modelled with ChemCAD and optimized with the dynamic programming optimization method. The objective function of the hybrid system is the total annual cost but its energy consumption is also investigated. It can be determined that this hybrid process, that is, the combination of organophilic and hydrophilic pervaporations is capable for the separation of isobutanol and water and it can become the alternative of distillation based separation. Considering our results it can be assumed that the combination of the organophilic and hydrophilic pervaporations opens new horizons for the membrane processes.

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1. Introduction

Pervaporation (PV) is a chemical unit operation where the liquid mixture to be separated is vaporized at low pressure on the downstream/permeate side of the membranes and the separation of the mixtures takes place by preferential sorption and diffusion of the desired component through the membrane [1]. A solution to achieve the difference in the partial pressures is to maintain a low vapour pressure using a vacuum pump on the permeate side [2,3]. Pervaporation shows good features such as special separation effect, no-extra material addition and energy-saving which are difficult to obtain by other conventional methods [4]. The pervaporation process can be used for the dehydration of organic solvents [5–9] or for the removal of low concentration organics from aqueous mixtures [10–16] depending on if it is about a hydrophilic or organophilic pervaporation. The pervaporation based separation of organic–organic mixtures is also a research subject [16–18].

The removal of organics from aqueous solutions is of particular interest of water recycling processes like fermentation, treatment of wastewater [19–22].

Depending on the permeating component two main areas of pervaporation can be identified: hydrophilic and organophilic pervaporation [23–25]. Baker [26] prognoses that pervaporation/vapour permeation based dehydration of bio alcohols has the largest potential for commercial application.

The aim of this work is to examine the isobutanol (IBU) water separation with pervaporation. The organophilic–hydrophilic pervaporation process is modelled and optimized for the separation of isobutanol–water mixture. IBU forms heteroazeotrope with water [27–29], therefore this mixture cannot be separated with conventional distillation. Valentynyi and Mizsey [30] have reported a hybrid distillation/hydrophilic pervaporation separation technology for water removal isobutanol.

At first IBU content is dehydrated into limited solubility region with organophilic pervaporation (OPV), and then the top phase of the isobutanol–water mixture is purified further with hydrophilic pervaporation (HPV). The aim of the second membrane module is to produce isobutanol with a purity of min. 99.0 m/m%.

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Nomenclature

| | |
|--------------|---------------------------------------------------------------------------------------------------------|
| A | membrane transfer area (m ²) |
| B | constant in Model II (–) |
| \bar{D}_i | transport coefficient of component i (kmol/(m ² h)) |
| F | feed |
| i | component number |
| j | component number |
| J_{total} | total flux (kmol/(m ² h)) |
| J_i | partial flux (kmol/(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 vapour phase membrane side (bar) |
| p_3 | pressure on the permeate side (bar) |
| P_i/δ | permeance of component i (kg/(m ² hbar)) |
| Q | head duty (MJ/h) |
| Q_0 | permeability of the porous support layer of the membrane (kmol/(m ² hbar)) |
| R | retentate |
| R^2 | R-squared value (–) |
| t | time (h) |
| T | temperature (°C) |
| x_F | feed isobutanol weight fraction in $y-x$ vapour–liquid equilibrium (VLE) diagram (Fig. 1 and 5) (–) |
| x_{i1} | concentration of component i in the feed (m/m%) |
| y | permeate isobutanol weight fraction in $y-x$ vapour–liquid equilibrium (VLE) diagram (Fig. 1 and 5) (–) |

Abbreviations

| | |
|-------|--------------------------------------------------------|
| HPV | hydrophilic pervaporation |
| hydr | hydrophilic |
| IBU | isobutanol |
| LTA | Linde Type A |
| M & S | Marshall & Swift index |
| OF | objective function |
| OPV | organophilic pervaporation |
| org | organophilic |
| PAE | polyamide-imide |
| PAN | polyacrylonitrile |
| PEBA | polyether-block-amide |
| PEI | polyetherimide |
| PDMS | polydimethylsiloxane |
| PSI | Pervaporation Separation Index (kg/(m ² h)) |
| PUR | polyurethane |
| PVA | polyvinyl alcohol |
| PV | pervaporation |
| TAC | Total Annual Cost (1000\$/year) |
| TFN | thin film nanocomposite |
| VLE | vapour–liquid equilibrium |

Greek letters

| | |
|------------------|---------------------------------------------------|
| α | separation factor |
| β | selectivity |
| $\bar{\gamma}_i$ | average activity coefficient of component i |
| γ_{i1} | activity coefficient of component i in the feed |
| δ | membrane thickness (μm) |

Pervaporation can be characterized by certain quantities and factors. The flux is calculated using the following equation [31]:

$$J_i = \frac{P_i}{\Delta t \cdot A} \quad (1)$$

where P_i is the partial weight of component i in the permeate, Δt is the time of duration of experiment and A is the membrane area. Separation factor is calculated by the following equation:

$$\alpha = \frac{y_i(1 - x_i)}{x_{i1}(1 - y_i)} \quad (2)$$

where α is separation factor (dimensionless), x_i is weight fraction of isobutanol in feed and y_i is weight fraction of butanol of permeate. The pervaporation separation index (PSI) is defined:

$$\text{PSI} = J \cdot (\alpha - 1) \quad (3)$$

The performance of pervaporation membranes can be described by the permeance as component flux normalized for driving force the pressure difference-normalized flux [31,32]:

$$\frac{P_i}{\delta} = \frac{J_i}{\gamma_{i1} \cdot x_{i1} \cdot p_{i0} - y_i \cdot p_3} \quad (4)$$

The ideal membrane selectivity β is calculated as the ratio of permeances [32,33]:

$$\beta = \frac{P_i/\delta}{P_j/\delta} \quad (5)$$

1.1. Comparison of different membranes on the separation of isobutanol and water

Tables 1 and 2 show a comparison of available experimental data of pervaporation membranes for organophilic and hydrophilic PV of the isobutanol–water mixture. Using Eq. (3), PSI values can

be calculated and applied to characterize and rank membrane operations. For the sake of membrane characterization the permeance (Eq. (4)) can be also applied. The selectivity (Eq. (5)) is also a frequently applied tool of comparison and it characterizes the membrane in a clear and obvious way [32].

It can be seen that the separation factors (Eq. (2)) are significantly higher for the hydrophilic pervaporation than those of the organophilic ones. In the case of OPV, polydimethylsiloxane (PDMS) membranes have the highest PSI published in the literature. The zeolite membranes possess superior thermal, mechanical and chemical properties that may show advantage over polymer membranes [34]. This type of membranes have good separation capability in the case of hydrophilic, but in group of commercially available membranes, polyvinyl alcohol (PVA) membranes have the highest PSI values.

For the sake of comprehensive information, the features of the different kinds of pervaporation membranes are compared and ranked. The comparison can be explained that the pervaporation data found in the literature are usually reported only as simple measured data on a special kind of membrane and they include fluxes and separation factors. These data are only a function of the intrinsic properties of the membranes used but they also depend on the operating conditions e.g. feed concentration, permeate pressure and feed temperature.

Calculated selectivities of pervaporation of isobutanol–water mixtures are ranked in Table 3. More data cannot be extracted from Tables 1 and 2 because permeate pressures are not reported in the presented papers.

Following the idea already applied in [43–45], the $y-x$ vapour–liquid equilibrium (VLE) diagram is selected for such a comparison. Since pervaporation is always compared with distillation, this representation allows to get clear information about the features of the different membranes compared with a possible flash distillation [26,32]. Using Eq. (2) permeate isobutanol concentrations

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