



Separation and purification of isobutanol from dilute aqueous solutions by a hybrid hydrophobic/hydrophilic pervaporation process



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ABSTRACT

In this study, a hybrid hydrophobic/hydrophilic pervaporation process was employed to separate and purify isobutanol from its dilute aqueous solutions. For this purpose, composite polydimethylsiloxane membranes were initially used for the recovery of isobutanol by hydrophobic pervaporation. Then the hydrophilic pervaporation with a composite polyvinyl alcohol membrane was utilized to separate water from the organic phase of the permeate stream of the hydrophobic pervaporation. The effect of feed flow rate on the performance of pervaporation was investigated. The resistance in series model was also applied to calculate the transport resistances through the composite membranes. It was observed that an enhancement in the feed flow rate led to higher permeation flux and selectivity of the more permeable component, while the flux of the less permeable component was almost constant. Also, the ratio of liquid boundary layer resistance to membrane layer resistance decreased by an increase in the feed flow rate. The isobutanol with a purity of higher than 99 wt.% was produced by the hybrid hydrophobic/hydrophilic pervaporation technique from a 2 wt.% aqueous isobutanol solution.

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1. Introduction

Biofuels are a suitable replacement for fuels gets from sources are not renewable or their renewal period is too long. Depletion of natural or petroleum products and rising prices of raw materials result in a search for renewable energy sources and biofuels [1]. Among biofuels, bioethanol is widely produced and used in different industries. Biobutanol is another biofuel that has properties which are more attractive in comparison with bioethanol. Biobutanol as opposed to bioethanol is non-hygroscopic and corrosion, and has a higher calorific value due to its greater energy content. Also, butanol is combinable with gasoline in any composition and its mixture with gasoline has a lower vapor pressure in comparison with ethanol-gasoline that leads to less fuel vaporization and destruction [1]. Isobutanol is produced by the carbonylation of propylene. It is also produced naturally during the fermentation of carbohydrates [2]. Isobutanol can also be produced by some engineered microorganisms such as corynebacterium [3]. Isobutanol produced from the biological method has a low concentration

and needs to be concentrated and purified for use in different applications [1,4]. Separation processes such as distillation, liquid-liquid extraction, adsorption, gas stripping and pervaporation have been used to separate alcohols from their aqueous solutions [5–7]. Compared to traditional processes, the pervaporation process has many advantages such as no heat damage to heat-sensitive compounds, low energy consumption, no additional separation treatment for added solvents or absorbents and minimum loss of alcohol. Therefore, pervaporation is an economical and useful technique for extraction of isobutanol from fermentation broth and its aqueous solutions [8,9].

Pervaporation is a membrane process for the separation of liquid mixtures by partial vaporization through a non-porous membrane. There are a large number of studies examining the performance of the pervaporation process for the separation of various alcohols such as methanol [10–12], ethanol [13–18], isopropanol [19–21], n-butanol [8,22–26] and isobutanol [22,27,28] from their mixtures with water. In these studies, a hydrophilic membrane was employed to separate water from the alcohol/water mixtures or a hydrophobic membrane was used to recover alcohol from its aqueous solutions, and further purification was not performed on the products of the pervaporation process. Furthermore, pervaporation-based hybrid processes have employed for the recovery and separation of alcohols from their aqueous solutions [29–31].

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List of symbols*Nomenclature*

A	constant in Eq. (13)
a	constant of Sherwood number
B	constant in Eq. (13)
b	constant of Sherwood number
c	constant of Sherwood number
d	constant of Sherwood number
D_i	diffusion coefficient of component i into the membrane (m^2/s)
d_h	hydraulic diameter of the membrane module (m)
J_i	flux of component i ($\text{g}/\text{m}^2 \text{ s}$)
K_L	mass transfer coefficient of the boundary layer (m/s)
L	length of the membrane module (m)
N	number of elements
P_i	permeability coefficient of component i ($\text{cm}^3 \text{ cm}/\text{cm}^2 \text{ s cmHg}$)
p_i	partial pressure of component i (cmHg)
$p_{o,i}$	equilibrium vapor pressure of component i (cmHg)
p_p	permeate pressure (cmHg)
Re	Reynolds number
R_L	mass transfer resistance of the liquid boundary layer (s/m)
R_M	mass transfer resistance of the membrane layer (s/m)
R_t	total mass transfer resistance (s/m)
S	area of the membrane (m^2)
Sc	Schmitt number
Sh	Sherwood number
t	time duration of the experiment (s)
u	feed velocity (m/s)
W	weight of the collected permeate (g)
x_i	mole fraction of component i

Greek letters

α	selectivity
δ_M	membrane thickness (m)
ΔC	concentration difference (g/m^3)
ν	kinematic viscosity (m^2/s)

Subscripts and superscripts

i	component index
f	feed
p	permeate

In the present study, a hybrid hydrophobic/hydrophilic pervaporation process is used for the recovery and purification of isobutanol from aqueous solution. At first, isobutanol is separated from its dilute aqueous solutions through the hydrophobic pervaporation process with a composite polydimethylsiloxane membrane. Since isobutanol and water dissolve in each other only to a limited extent, the permeate stream of the hydrophobic pervaporation separates into two insoluble liquid phases, an isobutanol-rich phase and a water-rich one. The water-rich liquid phase is returned to the feed solution of the hydrophobic pervaporation process. Then the hydrophilic pervaporation with a composite polyvinyl alcohol membrane is utilized to separate water from the organic phase of the permeate stream of the hydrophobic pervaporation. Furthermore, the resistance in series model is applied to analyze the mass transport resistances and study the transport mechanisms in the composite membrane.

2. Theory

The flux of component i varies linearly with the gradient in partial pressure according to [32]:

$$J_i = \frac{P_i}{\delta_M} (p_{f,i} - p_{p,i}) \quad (1)$$

where $p_{f,i}$ and $p_{p,i}$ are the partial pressure of component i in the feed mixture and vapor permeate, respectively, δ_M is the membrane thickness and P_i is the permeability coefficient. The partial pressure of component i in the feed is:

$$p_{f,i} = \gamma_i x_{f,i} p_{o,i} \quad (2)$$

where γ_i is the activity coefficient, $x_{f,i}$ is the mole fraction in the feed and $p_{o,i}$ is the equilibrium vapor pressure of component i . According to Dalton's law, the partial pressure of a component in the permeate can be expressed as:

$$p_{p,i} = x_{p,i} p_p \quad (3)$$

where $x_{p,i}$ is the mole fraction of the component in the permeate and p_p is the permeate pressure. Due to a very low total permeate pressure, the partial pressure of the permeate is very small in comparison with the partial pressure of the feed, so it can be neglected. The flux can thus be assumed to vary linearly with the equilibrium vapor pressure of the pure component:

$$J_i = \frac{P_i}{\delta_M} \gamma_i x_{f,i} p_{o,i} \quad (4)$$

Furthermore, according to resistance in series model, the transport of components from the feed solution through the composite membrane occurs by the following steps [33]: (i) diffusion through the liquid boundary layer, (ii) sorption into the membrane active layer, (iii) diffusion of liquid through the membrane active layer, (iv) desorption out of the active layer, and (v) transport of vapors through the porous support. Among these resistances, the liquid boundary layer and membrane active layer resistances control the mass transport in the pervaporation, and other transport resistances are low and negligible [14,16]. Therefore, under the steady state conditions, the flux of component i across the membrane can be expressed as a ratio of the driving force over the total mass transfer resistance (R_t), as follows:

$$J_i = \frac{\Delta C_i}{R_t} \quad (5)$$

where

$$R_t = R_L + R_M \quad (6)$$

and

$$R_L = \frac{1}{K_L} \quad (7)$$

$$R_M = \frac{\delta_M}{D_i} \quad (8)$$

where K_L and D_i are the mass transfer coefficient of boundary layer and diffusion coefficient of component i in the membrane, respectively.

The Sherwood correlation has been used to determine the mass transfer coefficient (K_L) through the boundary layer [16,34]:

$$Sh = \frac{K_L L}{D} = a Re^b Sc^c \left(\frac{d_h}{L} \right)^d \quad (9)$$

The constants a , b , c and d for the Sherwood correlation are given in Table 1.

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