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Experimental study and cost evaluation for ethanol separation from fermentation broth using pervaporation

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ABSTRACT

In this work, the separation of two types of mixtures, ethanol–water mixture and fermented sweet sorghum, was investigated using cellulose acetate supported polyvinyl alcohol composite membranes. The pervaporation (PV) performances of the two mixtures under different operating parameters (ethanol concentrations, operating times and temperatures) were studied. PV performances using sweet sorghum fermentation broth, under all operating parameters, were significantly lower compared to separating ethanol from binary mixtures. Preliminary economic analysis shows that cost of producing 1 l of ethanol from the broth is about 0.9 $/l which is about 1.1 times higher than from the pure binary system.

Keywords: Ethanol–water mixture; Sweet sorghum juice; Pervaporation; PVA; Membrane unit; Economic assessment

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1. Introduction

Sweet sorghum, as a biomass source to produce bioethanol, is gaining increasing interest because it can be cultivated in almost all temperate and tropical climate areas and is the only crop providing grain and stem that can be used as substrates for the production of sugar, alcohol, syrup, fodder, fuel, bedding, roofing, fencing and paper [1]. The conversion of sweet sorghum to bioethanol through fermentation is a common practice in which the recovery and purification of ethanol from fermentation broth generally involves distillation which is energy intensive and complex. It has been estimated to consume more than half of the total energy in the production of alcohol by fermentation [2].

For bioethanol processes to be competitive with fossil energy resources, production costs must be reduced. As a low-energy consumption, moderate cost, compact, unlimited by relative volatility of the components and modular design process, pervaporation (PV) has attracted intensive attentions and has been used as an alternative technique for in situ ethanol recovery from fermentation [2–5]. PV is a membrane separation process based on the difference in solubility and diffusivity of the components to be separated through a dense membrane [6,7].

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The complexity of fermentation broths, which are multicomponent mixtures that contain a variety of byproducts, results in several anecdotal observations regarding its impact, both positive and negative, on the PV performances [8–12]. To complicate matters, several studies have indicated no significant effect of fermentation broth on PV performance [11,13]. To the authors' knowledge, there has been no work undertaken to elucidate the effect of sweet sorghum fermentation broth on PV performances using composite membranes although ethanol production from these biomass is ever increasing.

The main objective of this study was to investigate the effect of sweet sorghum fermentation broth on the PV performances using commercial cellulose acetate coated with PVA membranes. Poly(vinyl alcohol) is chosen as the material of choice since it has good film-forming properties and is an industrialized candidate for the separation of ethanol–water mixtures [14]. PV studies were carried out using ethanol–water mixture as the control study prior to separating ethanol from fermented sweet sorghum broth The effects of operating temperature, ethanol concentrations and PV period were also investigated and evaluated in terms of permeate flux, separation factor and pervaporation separation index (PSI).

2. Material and methods

2.1. Preparation of ethanol–water and fermented sweet sorghum juice mixtures

All chemicals used in this study were of analytical grade (SC Scientific and Merck Chemical, Bangkok, Thailand). The first ethanol–water mixture was prepared from pure 99.9% ethanol (analytical grade) and mixed with deionized water to the desired ethanol concentrations (10, 15, 20 and 30%) whilst the second mixture of fermented sweet sorghum juice was obtained after 3 days of batch fermentation of sweet sorghum juice using a pure culture of *Saccharomyces cerevisiae*.

2.2. Experimental setup and procedure

A 500 mL benchscale Pyrex membrane unit was assembled with the PV system. The PV system consisted of a membrane unit on a magnetic and hot plate stirrer, air tube, vacuum unit, condenser and the vacuum pump was connected in the permeate side of the system (see Fig. 1). Flat sheet cellulose acetate membrane as the support, kindly received from Research Centre of Nanomaterial, Faculty of Science, Khon Kaen University, Thailand, was dip-coated (onto the feed side only) with PVA and placed in a circular plate and frame permeation cell. The dip-coating process was performed at room temperature in a clean-room environment. The membranes were 3.6 cm in diameter and had a surface area of 10.2 cm^2. The nominal pore size of the membranes was 0.45 μm and the membrane thickness was approximately 300 μm.

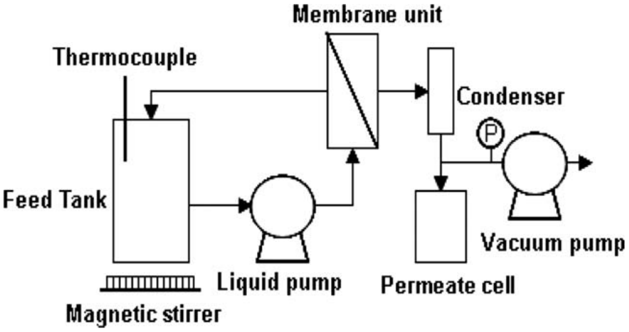

Fig. 1. Schematic of PV system.

The mixtures were pumped into the membrane from the stirred feed tank via a peristaltic pump. A vacuum pump was fixed to maintain a permeate pressure of 50.8 mm Hg. The temperature of the feed liquid mixture was heated and kept constant at the desired temperatures (50, 60 and 70°C) using a hot plate magnetic stirrer to heat the stirred feed tank. PV was carried out for periods of 15, 30, 45 and 60 min. Each PV experiment was repeated several times and the average value is reported here. All measurements show good reproducibility as the errors never exceed 5%.

2.3. Analytical methods

Permeate and feed concentrations were measured off-line before and after PV. The volumes of the mixtures were weighed using analytical balance (BP 2215, Sertorius, Germany) and the ethanol concentration was collected and analysed using gas chromatography (14 B-Shimadzu, Japan).

2.4. Evaluation of PV performances

PV performances were evaluated in terms of flux, separation factor (α), PSI and percent separation.

The flux, J, is calculated as

$$J = \frac{W}{A \times t} \tag{1}$$

where *W, A and t* are the mass feed pass through membrane (kg), surface area of membrane (m^2) and PV time (h), respectively.

The selectivity (α) of the membrane, or also termed as separation coefficient, for ethanol (E) relative to water (Wat) is defined as [15,16]

$$\alpha = \frac{Y_E/Y_{\mathrm{Wat}}}{X_E/X_{\mathrm{Wat}}} \tag{2}$$

where Y_i and X_i are the mass fractions of component i in the permeate vapour and feed liquid, respectively.

The effectiveness of separation is determined equally by the membrane selectivity and permeation properties. The parameter, which accounts for these two factors, is the so-called "pervaporation separation index"—calculated as the ratio of the total permeate flux (J) and the separation coefficient or selectivity (α) as

$$\mathrm{PSI} = J\alpha \tag{3}$$

The percent separation, also termed as separation efficiency %, was calculated as $100 \times \{$(EtOH in feed) − (EtOH in permeate)$\}/$(EtOH in feed).

The swelling measurements were carried out by immersing dried membrane in water–ethanol mixtures of different compositions. After 24 h, the membranes were removed, pressed between a tissue paper and weighed. This procedure was continued until no further weight increase was observed. The degree of swelling which is referred to herein as overall solubility, $\mathrm{Sol}_{\mathrm{overall}}$, is calculated from the weight of the swollen and the dry membrane sample and is expressed in units of grams of sorbed mixture per gram of dry membrane using the expression

$$\mathrm{Sol}_{\mathrm{overall}} = (m_{\mathrm{M}} - m_{\mathrm{D}})/m_{\mathrm{D}} \tag{4}$$

where m_{M} and m_{D} are the mass of the swollen membrane and mass of the dry membrane, both in grams, respectively.

3. Results and discussions

3.1. Effect of ethanol concentrations

Table 1 shows the influence of feed ethanol concentration on separation efficiency and permeate flux. The increase in ethanol concentration is accompanied by a decrease in the permeate flux but an increased separation efficiency at the highest ethanol concentration. The increased selectivity is due to the decreased swelling of the polymer in high ethanol concentrations whilst the decreased permeate flux can be explained in terms of the plasticizing effect of the water on membranes. With higher water concentration in the feed mixture, the amorphous regions of the membrane are more swollen and the polymer chains become more flexible. In swollen membrane, both ethanol molecules and water molecules pass across the membrane more easily, leading to the decrease of membrane selectivity and an increased in flux [17,18]. Diffusion behaviour of water and small molecules in polymer gel can be interpreted by the free volume theory as well as NMR spectroscopy [19,20]. In addition, the increase in the feed ethanol concentration will also impact the partial pressure driving force. The linear relationship between the flux to the partial pressure driving force is often used to explain the water flux data through PV membrane and is shown as follows [21],

$$J_{\mathrm{w}} = K_{\mathrm{w}}(a_{\mathrm{w}}P^{\mathrm{s}}_{\mathrm{w}} - y_{\mathrm{w}}P_{\mathrm{p}}) \tag{5}$$

where J_{w}, K_{w}, a_{w}, P_{w}, y_{w} and P_{p} are the water flux, permeability constant of water, activity of water in feed, saturation pressure of water, mass fraction of water in permeate and permeate pressure.

Several workers [21,22] observe the association of low partial pressure driving force with high solvent concentrations in the feed mixture and this could also explain the observations found in the current study.

Table 1
Effect of ethanol concentrations on PV system at 50.8 mm Hg; 60°C; 30 min

Ethanol concentration (%)	J_{total} (kg/m^{2}h)	α	PSI (kg/m^{2}h)	Separation efficiency (%)	Overall solubility (weight fraction)
5	19.7	4.1	81.2	18.4	0.28
10	12.0	6.5	77.7	27.4	0.19
15	6.9	7.8	54.0	36.2	0.15
30	0.7	50.3	36.5	82.9	0.12

3.2. Effect of operating time

The membrane durability or long-term membrane operating stability, in commercial applications, is a critical factor and is therefore investigated in this study. The influence of operating time on the PV separation of a 15% ethanol–water solution at 60°C is shown in Table 2. As the PV period increased, both selectivity and permeate fluxes declined. From this table, it is apparent that after a period of 30 min, both permeate fluxes and selectivities are stable. This finding illustrates that the PVA-cellulose acetate composite membrane exhibited membrane durability during the PV separation at high operating temperature.

3.3. Effect of temperature

It is evident in Table 3 that the overall performance in terms of PSI increases with increasing temperature. The increase in PSI results from the increase in permeate fluxes and the decline in the separation factor. According to solution-diffusion mechanism [23], the increasing of temperature makes the solubility on the surface of membrane and the diffusion rate in the membrane increase. This is evidently the case since diffusion was found to increase and is reflected as a positive slope of the Arrhenius plot as discussed later. The free volume of the membranes increases under higher temperature, which makes the interspaces between the polymer chain bigger, and the kinetic energy of permeate is increasing, which makes the diffusion easier. Because of the coupling effect, the ethanol relative concentration is increased, and the separate selectivity is notably decreased with the temperature increasing. Arrhenius-type relationships can be used in describing the effect of temperature on flux as follows [24]

$$J_{\text{total}} = J_0 \exp(E_a/RT) \tag{6}$$

where J_{total}, J_0, E_a, R and T are the total permeate flux (g/(m^{2}h)), pre-exponential factor (g/(m^{2}h)), the activation energy (J/mol), the universal gas constant (J/(mol K)) and the absolute temperature in Kelvin (K), respectively. The temperature effect on the PV process was investigated. The curve $\ln(J) = f(1/T)$ is straight and the gradient from this plot following Eq. (6) gives the activation energy of PV. The activation energy is 52 kJ/mol and the positive activation energy value indicates that the permeation flux increases with increasing temperature. Corresponding activation energy of diffusion (E_D) is found to be 7 kJ/mol and is obtained from the slope of the Arrhenius plot of the apparent diffusion coefficients as a function of temperature i.e. $\ln(D) = f(1/T)$. The activation energy for diffusion is obtained from the temperature dependence of the apparent diffusion coefficient, which is described by the Arrhenius equation as

$$D_a = A \exp(E_{\text{diffusion}}/RT) \tag{7}$$

where D_a, A, $E_{\text{diffusion}}$, R and T are the apparent diffusion coefficient (m^2/s), constant, the activation energy of diffusion (J/mol), the universal gas constant (J/(mol K)) and the absolute temperature in Kelvin (K), respectively.

Table 2
Effect of operation time on PV at 50.8 mm Hg; 60°C; 15% ethanol–water

Time (min)	J_{total} (kg/m^{2}h)	α	PSI (kg/m^{2}h)	Separation efficiency (%)
15	10.7	9.4	100.6	38.5
30	6.9	7.8	53.8	36.2
45	6.3	7.9	49.7	37.5
60	6.1	7.6	46.4	36.3

Table 3
Effect of temperature PV at 50.8 mm Hg, 30 min, 15% ethanol–water

Temp (°C)	J_{total} (kg/m^{2}h)	Α	PSI (kg/m^{2}h)	Separation efficiency (%)
50	2.6	10.9	28.3	40.5
60	6.9	7.8	54.1	35.6
70	7.9	7.4	59.0	34.4

In this binary system, the activation energy and E_D for both ethanol and water were positive values which show that the activation energy of diffusion dominates over the activation energy of sorption since activation energy (E_a) is the sum of the activation energy of diffusion and the enthalpy of sorption (ΔH).

3.4. Effect of fermented sweet sorghum juice broth on PV performances

The concentration of ethanol within the fermentation broth is at 14.6%. Generally, there is significant deterioration in membrane flux and selectivity when PV is carried out using the fermentation broth, except for the instance at 50°C where flux of 3.8 kg/(m^{2}h) is found which is nearly 50% higher compared to the value for the binary ethanol–water mixture (Table 4). This anomaly may arise from the potential change to the composition of the fermentation broth as a result of the microorganisms which it contains. The slight change in the composition can lead to significant changes in the PV performances. Further work is required on the effect of temperature on fermentation broth composition to ascertain the reason for the anomaly. The general observation arises from the presence of high yeast cell concentrations and residual byproducts within the fermented ethanol. The yeast cell concentration in the broth is approximately 9.0 cells m/l whilst residual soluble solids of 50 g/l. This result is also observed with other workers [2,10] who attribute the decline in membrane flux to the presence of yeast cells as well as other byproducts within the fermented ethanol. Ethanol selectivity decreases in the presence of the broth. The PV performances in the fermentation broth follow the similar trend observed with ethanol–water mixture studies under the changing temperatures, although the changes in permeate fluxes and separation efficiencies are not as significant in the fermentation broth. This suggests that the transport of ethanol molecules in the fermentation broth system is less affected by temperature. It is well known for the PV process, the permeating components are first absorbed at the membrane surface, and the absorbed components then diffuse through the membrane. The results observed with fermentation broth system seem to suggest that the absorption of ethanol molecules at the membrane surface is hindered. Therefore, the effect of temperature on ethanol flux for the fermentation broth system is small, although the increase of temperature is beneficial for the diffusion of ethanol molecules due to the expansion of the free volume [23].

Table 4
Effect of various operating conditions on the PV performance in fermentation broth

Operating conditions (%)	J_{total} (kg/m^2 h)	α	PSI (kg/m^2 h)	Separation efficiency (%)
Time (min)				
15	5.2	8.6	44.7	39.1
30	4.1	7.8	31.6	36.2
45	3.8	7.3	27.7	37.5
60	3.6	7.0	24.8	36.3
Temperatures (°C)				
50	3.8	9.2	35.2	38.3
60	4.0	7.8	31.6	34.9
70	4.3	6.8	29.4	32.6

The apparent activation energies (E_a) of permeation are 3 and 13 kJ/mol for ethanol and water, respectively, whereas their corresponding activation energies of diffusion (E_D) are 2 and 10 kJ/mol. In this study, the positive E_D and E_a indicate that the membrane permeability coefficient increases with increasing temperature [25]. A higher E_a was obtained in the binary system (52 kJ/mol) when comparing to the E_a obtained in the fermentation broth system of 5 kJ/mol which indicates that PV on fermentation broth is not as sensitive to temperature changes as the binary system.

A preliminary sensitivity analysis of the PV performance is carried out following Pangarkar and Wasewar [26]. These authors carried out an economic analysis on the complete process of fermentation, PV and distillation. The cost analysis i.e. direct production costs presented in this study focuses only on the PV aspect of the whole process. The separation cost of ethanol per litre in the ethanol–water mixture as well as from sweet sorghum broth has been estimated based on PSI (see Fig. 2) based on the following assumptions listed in Table 5.

The PSI was chosen as the overall determining PV parameter since it relates to both flux and separation factor. Generally, membrane having higher flux requires less PV membrane area and hence lowers production costs whilst membrane having higher separation factor gives higher concentration in permeates and requires less energy for further enrichment in the concentration and hence less production cost. Fig. 2 shows that as PSI increases the cost of producing ethanol decreases. The cost of producing 1l of ethanol from the broth is about 1.1 times higher than from the pure binary system. This implies that the cellulose acetate PVA composite membrane for the separation of ethanol from sweet sorghum fermentation broth is

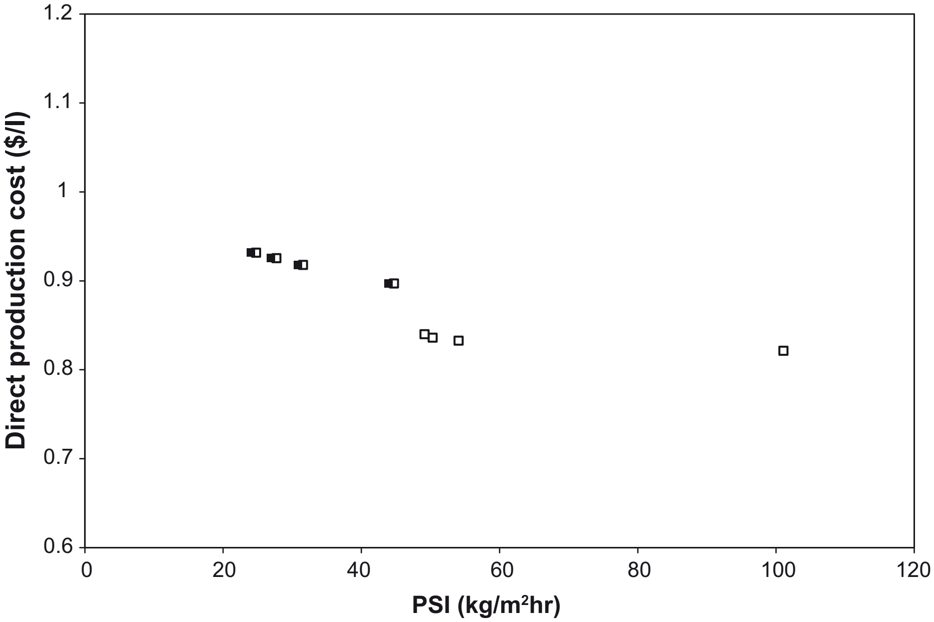

Fig. 2. Influence of PV performances on production costs (☐: binary feed mixture ethanol–water; ■: sweet sorghum fermentation broth).

Table 5
Data used for the calculation of the ethanol production cost attributable to PV (all costs are in US $)

Quantities of filtered ethanol	30 m^3/d
Capital cost—membrane cost	\$200 per m^2
Operational cost—utilities[a]	\$0.01/l
Direct production costs = raw material (membrane) + utilities	

[a] Utilities cost includes steam, electricity, air and cooling water and the cost was been adapted from Pangarkar and Wasewar [26].

not commercially viable and further investigations in the development of membranes for high flux and separation factor are required for the separation of ethanol from sweet sorghum fermentation broth.

4. Conclusion

The PV performances using PVA-cellulose acetate composite membranes under the various operating conditions using ethanol–water mixtures and subsequently, sweet sorghum fermentation broth were investigated. In the binary mixture, increasing ethanol concentrations and temperature resulted in higher selectivity but decreased fluxes. This is attributed to the decreased swelling of the membrane structure. Stable membrane flux and selectivity were observed after 30 min of operation. Direct comparisons of PV performances were made at 15% ethanol–water since it closely resembles the ethanol concentration found in the sweet sorghum fermentation broth. In the sweet sorghum fermentation broth, both permeate flux and selectivity decreased significantly compared to the binary mixture under all operating conditions which makes PV using PVA-cellulose acetate composite membrane undesirable.

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