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## RESEARCH ARTICLE

### HYBRID PERVAPORATION/DISTILLATION PROCESS FOR ETHANOL - WATER SEPARATION, EFFECT OF DISTILLATION COLUMN SIDE STREAM

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#### ABSTRACT

Separation operation is the crucial part of the chemical industry. With rising energy prices and increased focus on efficiency and sustainability, the development of alternative separation processes to conventional separation processes constitute major areas of opportunity in the chemical industry. Distillation is one of the most used separation processes in industry and is widely used for the separation of liquid mixtures. Energy requirement for distillation is more and a separation involving close boiling point mixture is not feasible in conventional units. Hybrid separation processes combine different unit operations and constitute a promising design option for the separation of complex mixtures. Hybrid separation processes are energetically more efficient compared to conventional distillation. This is demonstrated using ethanol-water as the model system and separation by Distillation-Pervaporation (D + PV) hybrid unit. A simple, user friendly tool has been developed to study different pervaporation and hybrid configurations. Hybrid process configuration with distillation column side stream is compared with simple hybrid process configuration. It is observed from that for same feed, energy requirement for process with side stream with drawl requires 9 % less energy as compared to process without side stream with drawl.

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#### INTRODUCTION

Distillation is one of the most used separation processes in industry and is widely used for the separation of liquid mixtures. However, it is difficult to separate the azeotropic or constant boiling mixtures with conventional distillation. As energy costs rise, hybrid separation strategies that combine one or more separation techniques with distillation are attracting attention as a means of saving energy. Pervaporation is one of the most promising membrane technologies offering solution to dehydration of organic compounds, recovery of organic compounds from aqueous solutions, separation of organic mixtures etc (Lipnizki *et al.*, 1999). The annual treatment cost of pervaporation is related to the membrane area and pervaporation alone is often unable to economically provide high purity retentate and permeate (Seader and Ernest, 2001; Nangare Doulat *et al.*, 2017). Combined process consisting of distillation and pervaporation/vapor permeation offers economically attractive alternatives as they can simplify the process structure, reduce the energy consumption and avoid the entrainers for the separation of azeotropes and close boiling solutions (Gonzalez and Ortiz, 2002).

The optimization and design strategies for various pervaporation/distillation configurations for the production of Methyl Tertiary Butyl Ether (MTBE) were explored by Hommerich and Rautenbach (Hommerich and Rautenbach, 1998). The analysis of hybrid pervaporation-distillation for acetic acid water separation revealed improved energy efficiency (Serval *et al.*, 2014). Hybrid pervaporation-distillation processes are expected to lead to decrease in energy consumption compared to conventional processes (Khebudkar *et al.*, 2017). In this paper we present a comparison of hybrid process, pervaporation and distillation with/without side stream using a friendly Visual Basic simulation tool. McCabe-Thiele method (Treybal, 1968) has been used for distillation tray calculations while Rautenbach model (Baker, 2004; Heintz and Stephan, 1994) based on solution diffusion theory has been used for pervaporation calculations.

#### Model Development

A distillation unit comprising with column, total condenser and partial reboiler was modeled using mass and energy balances at each tray, coupled with vapor liquid equilibrium data. UNIQUAC and Virial equations of state (Reid *et al.*, 1977), have been used to generate the vapor liquid equilibrium

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(VLE) of the mixture. Among the diverse pervaporation models described in literature, the solution diffusion model provides an accurate description of the behavior of membranes and the dependence of membrane transport on pressure, concentration etc. and hence is widely used (Heintz and Stephan, 1994; Wijmans and Baker, 1995). The Rautenbach solution diffusion model uses the fugacity gradient instead of the chemical potential gradient between the two sides of the membrane. The model works with transport coefficient instead of diffusion coefficient because its concentration dependence is considered negligible (Baker, 2004).

The molar flux of component k through the membrane can be calculated using Eq.(1)

$$J_K = \frac{1}{1 + \frac{D_K}{Q_0 P_{K0} \bar{V}_K}} \cdot \frac{\bar{D}_K}{\bar{V}_K} \cdot \frac{(P_{K1} - P_{K3})}{P_{K0}} \dots\dots\dots (1)$$

Where  $\bar{D}_K$  is the concentration independent transport coefficient of component k,  $Q_0$  is permeability of non supportive porous layer,  $P_{K0}$  is vapor pressure of component at feed temperature,  $P_{K1}$  and  $P_{K3}$  are the partial pressure of component k in the feed and permeate respectively  $\bar{V}_K$  is the average activity coefficient of component k. For high water concentration azeotropes like isobutanol–water separation, this equation needs modification (Serval *et al.*, 2014). Pervaporation is strongly temperature dependent as can be seen in Eq. (2). Transport coefficient depends on the temperature in an Arrhenius type exponential way.

$$\bar{D}_K = \bar{D}_K^* \exp \left( \frac{E_K}{R} \left( \frac{1}{T^*} - \frac{1}{T} \right) \right) \dots\dots\dots (2)$$

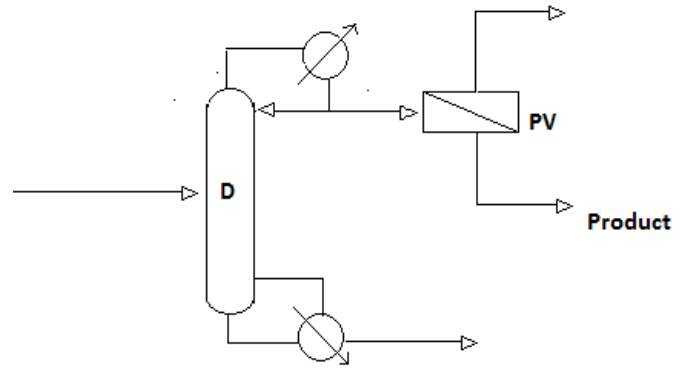
$E_K$  represents the activation energy for component k and is associated with the transport coefficient,  $T^*$  is the reference temperature, equal to 293 K. The activity coefficients are calculated with the Wilson equation (Serval *et al.*, 2014) in this work, but can be calculated with other equation of state models as well. The Wilson parameters were derived from the database of Chemsep software.

**Process simulation software**

A user friendly Visual Basic Simulation program (VBS) was developed, for PV and D+PV model calculations. The VBS tool developed is capable of calculation of Number of Trays, minimum reflux ratio, energy requirement for distillation column, membrane area, energy requirement for different product specifications etc. PV calculations require pervaporation parameters like component diffusivities, activation energies and permeance. Experimental data reported by Lovasz and coworkers (Lovasz *et al.*, 2007) has been used in this study for calculating the component flux through the membrane.

Lovasz used commercial membrane PERVAP 2210 (by SULZER Chemtech GmbH) for pervaporation. Data reported for this membrane at 80 °C and 2.7 mbar permeate vacuum has been used in our work. PERVAP 2210 is hydrophilic PVA/PAN (0.5-2 micro meter thick PVA as permselective and polyacrylonitrile as micro-porous support layer) membrane. The calculation flux was found to be in good agreement with the reported experimental data.

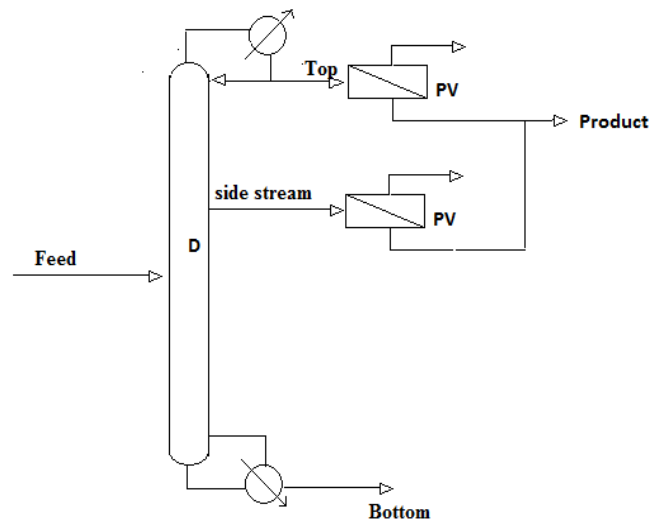
**Hybrid process (D+PV)**



**Figure 1. Configuration of a hybrid process coupling distillation and pervaporation (D+PV)**

Hybrid process configuration D+PV coupling pervaporation and distillation column with side stream studied in current work is presented in Figure 1. Ethanol rich distillate is condensed, partly refluxed and the remaining is passed through a pervaporation unit PV. As the membrane is hydrophilic, water permeates through, whereas ethanol remains as the retentate. Final product from this configuration is the combined retentate streams from pervaporation units placed at the top.

**Hybrid process (D+PV) with side withdrawl from distillation column**



**Figure 2. Configuration of a hybrid process coupling distillation and pervaporation (D+PV) with distillation column side stream**

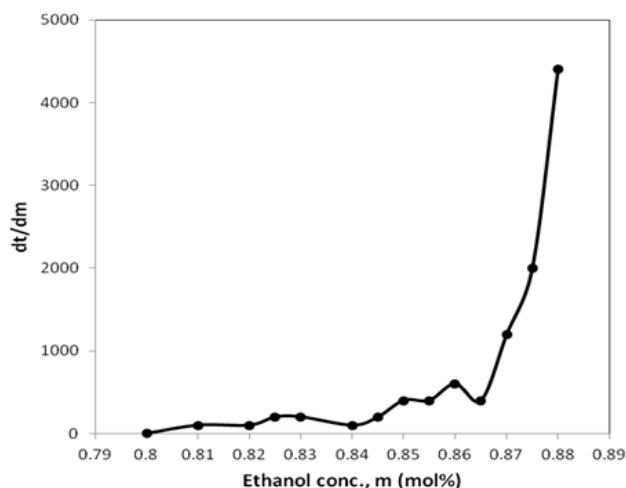
Hybrid process configuration D+PV coupling pervaporation and distillation column with side stream studied in current work is presented in Figure 1. Ethanol rich distillate is condensed, partly refluxed and the remaining is passed through a pervaporation unit PV1. In addition to distillate a side stream (saturated liquid) with 65 mol% ethanol composition is withdrawn from distillation column. Side stream is taken from the distillation column and fed to the pervaporation unit PV2. Final product from this configuration is the combined retentate streams from pervaporation units placed at the top and at the side stream.

**Table 1. Comparison of Hybrid process (D+PV) without side stream withdrawal from distillation column with Hybrid process (D+PV1+PV2) with side stream withdrawal from distillation column for production of 98 mol% ethanol. (Feed 1000 kmol/h, 20 mol% ethanol) (Top product distillate composition: 86 mol% ethanol)**

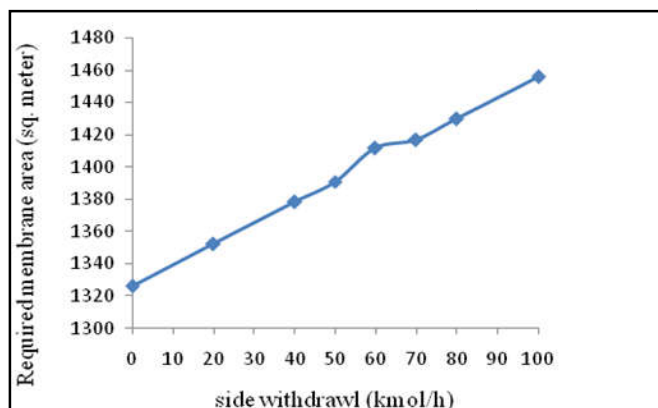
Side stream (65 mol% ethanol) (kmol/h)	No. of Trays	Hybrid process product (kmol/h)	Required Membrane area (m <sup>2</sup> )	Required energy (kW/h)
0	21	187.78	1271.43	23543.01
40	22	195.87	1378.04	21329.51

## RESULTS AND DISCUSSION

For the distillation of azeotropes in a conventional distillation unit without entrainer, the number of trays required for distillation increases rapidly as the concentration approaches azeotrope composition. The Figure 3 shows that there is rapid increase in the number of trays beyond an outlet ethanol concentration of 86.5 % (mol). So it becomes important to judiciously choose the number of trays in the distillation column of the D+PV hybrid unit. The figure indicates that in D + PV hybrid separation unit, it is advantageous to introduce PV unit at a tray composition of about 86 % (mol) instead of the azeotrope point (89.2 % (mol) ethanol).



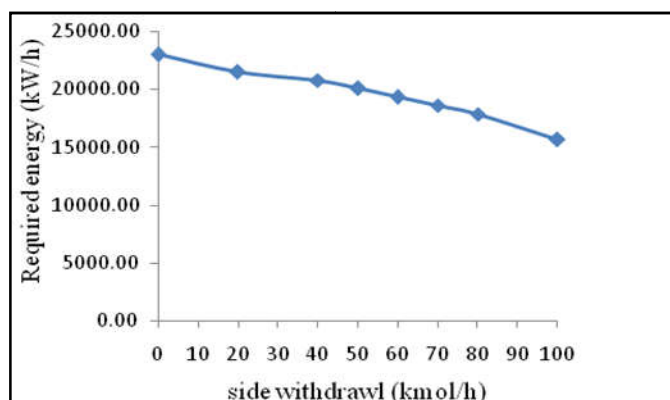
**Figure 3. Incremental variations in the number of trays with incremental increase in outlet ethanol concentration (dt/dm) as a function of mole fraction ethanol (m) at the outlet in a distillation tower for the distillation of ethanol-water system without any entrainer**



**Figure 4. Effect of side withdrawal from distillation column on hybrid process membrane area requirement**

A simulation has been carried out to study the effect of distillation column side stream withdrawal on the energy requirement for hybrid process. 1000 kmol/h Ethanol-Water

feed mixture (20 mole% Ethanol) is considered as feed for this simulation study. Top product composition from distillation column is fixed as 86 mol% ethanol (feed for PV1). Distillation column side stream withdrawal is increased from 0 to 100 kmol/h with 65 mol% ethanol composition (feed for PV2). The simulation study carried out is presented in figure 4 and figure 5. Comparison of Hybrid process (D+PV) without side stream withdrawal from distillation column and Hybrid process (D+PV1+PV2) with side stream withdrawal from distillation column for production of 98 mol% ethanol is studied and reported in Table 1.



**Figure 5. Effect of side withdrawal from distillation column on hybrid process energy requirement**

## Conclusion

While it is not possible to obtain ethanol purity above the azeotrope composition in a conventional distillation tower, by attaching a PV unit after the condenser in conventional distillation, it is possible to obtain very high purity ethanol as product. A comparison of hybrid separation unit comprising of pervaporation and distillation with /without sidestream has been presented in this paper. It is clear from both figures that for same top product composition and to produce same product composition, as side withdrawal from distillation column increases, membrane area required for separation increases and energy required for the separation decreases. For same feed energy requirement for process with side stream withdrawal requires 9 % less energy as compared to process without side stream withdrawal. However membrane area requirement is 8% more and product quantity is 4% more in case of hybrid process (D+PV1+PV2) with side stream withdrawal as compared to hybrid process (D+PV) without side stream withdrawal.

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