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# Experimental and optimization studies of diabatic membrane-based distillation columns

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#### ABSTRACT

Membrane-based distillation column (MDC) which utilizes hollow fibers as structured packings provides the advantages of large and fixed interfacial area and independent control of fluid flow rate without the constraints of flooding and loading. Diabatic distillation columns can approach the thermodynamic reversible operation, which is energy-efficient. MDC can be easily transformed into diabatic columns (D-MDC), *i.e.* with internal heat exchange, by adding a jacket as the flow channel for heating or cooling media. For the methanol-water system, the experimental results show noteworthy reduction of HTU (height of a transfer unit) under diabatic operation when compared to adiabatic operation. For the benzenetoluene system, the optimization of the internal heat exchange rates for D-MDC was conducted by a nested loop optimization algorithm, which incorporates the equipartition of entropy production (EoEP), equipartition of heat exchange rate (EoQ) or linear distribution of heat exchange rate (LQ) approximation, and an evolutionary approach. The optimal solutions of EoEP, EoQ and LQ provide significant improvements in specific exergy loss, exergy loss per NTU (number of transfer unit) and variance of driving force to adiabatic columns. The optimal solutions obtained from nested loop algorithm are superior to the solution from evolutionary approach.

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#### 1. Introduction

Distillation is one of the most widely used separation technology in chemical process industry. Distillation demands large inputs of energy, and even small improvements can save large amounts of energy. Petlyuk column [1], divided-wall column (DWC) [2], internally heat-integrated distillation column (HIDiC) [3] and diabatic column [4] are well known designs for the reduction of energy demand in the distillation columns.

In thermodynamics, the reversible operation of a process corresponds to the operation by means of infinitesimal driving force and sets the ultimate energy performance of the process. The reversible distillation column requires infinite number of stages with diabatic operation, which means heat supply along the stripping section and heat removal along the rectifying section. In practice, only finite number of stages can be used in distillation columns. According to Rivero [5], minimum exergy loss is obtained with an operating line being largely parallel to the equilibrium line in the McCabe-Thiele diagram as illustrated in Fig. 1. For practical diabatic tray columns with finite number of stages, an immense amount of work has been carried out to determine the profile of

\* Corresponding author. Tel.: +886 2 26232094; fax: +886 2 26209887. *E-mail address:* nhchang@mail.tku.edu.tw (H. Chang). internal heat exchange rate which minimizes the entropy production. Based on the hypothesis [6] that various theorems [7,8] are good approximations to the state of minimum entropy production in a system that have sufficient freedom, researchers have compared the internal profiles determined from numerical optimization with that from various asymptotical concepts, such as equipartition of force (EoF) [9] and equipartition of entropy production (EoEP) [10]. Despite the in-depth research work, the practical application of diabatic columns is difficult because of the heat exchange inside the conventional columns of either tray type or packing type. The optimization problem has also been studied by Rivero using a revolutionary approach [11].

Membrane-based distillation column (MDC) using hollow fiber membrane contactors as structured distillation packing has been first proposed by Zhang and Cussler [12,13]. The membrane is microporous, non-selective, and with or without a thin layer of nonporous coating to prevent convection flow across the membrane [12,14]. Because of the small diameter fibers and the vapor and liquid flows are not in direct contact, such columns have the advantages of large and fixed interfacial area, free of the fluid mechanics problems of loading and flooding, and large turndown ratio operations. Possible concerns of such columns include the extra mass transfer resistance of the membrane, higher pressure drop in small fibers, and the membrane materials must stand the chemical and

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Nomenclature
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В	flow rate of bottom product (kmol/hr)
С	concentration (kmol/m <sup>3</sup> )
D	flow rate of distillate product (kmol/hr)
Ex	exergy of stream (W)
Ex <sub>loss</sub>	exergy loss (W)
ex <sub>loss</sub>	specific exergy loss
F	flow rate of feed stream (kmol/hr)
f	objective function
Н	molar enthalpy (J/kmol)
h	heat transfer coefficient (W/m <sup>2</sup> K)
HTU	height of a transfer unit (m)
k	mass transfer coefficient (m/s)
L	effective length of the membrane module (m)
NTU	number of transfer units
Ν	transmembrane mass flux (kmol/m <sup>2</sup> s)
N <sub>data</sub>	number of data points
т	flow rate of bulk fluid (kmol/hr)
Р	pressure (bar)
Q <sub>h</sub>	convective heat flux (W/m <sup>2</sup> )
$Q_{l}$	liquid side heat exchange rate (W)
Qv	vapor side heat exchange rate (W)
$\frac{\mathbf{R}}{T}$	gas constant (J/kmol K)
Т	temperature (°C)
w <sub>m</sub>	total circumvent length of membrane tubes (m)
х	mole fraction in liquid phase or decision variable
у	mole fraction in vapor phase or dependent variable
$y_*$	mole fraction of vapor in equilibrium with liquid
Ζ	height of the column or coordinate location in the
2	flow direction (m)
$\sigma^2$	variance of driving force
Subscript	
<b>P</b> 7	benzene

BZ	benzene
bl	bulk liquid
bv	bulk vapor

С	cooling fluid
F	feed stream

- *i* component i
- in inlet
- L liquid

mem	membrane
ml	membrane-bulk liquid interface
mv	membrane-bulk vapor interface
out	outlet
TOL	toluene
V	vapor

thermal challenges. Many researchers have experimentally demonstrated the feasibility and the performance of MDCs which employ polymeric or ceramic membranes for the separation of various mixtures, such as isopropanol-water, methanol-ethanol, propylenepropane, iso-/n-butane and benzene-toluene [12–17].

Hollow fiber membrane modules (HFM) are similar to packing columns, which are not staged but continuous devices. The shelland-tube configuration of HFM makes they can be easily modified into a diabatic MDC by adding extra flow channels for heating or cooling media. In this study, a simple design of adding a jacket outside of the shell, as shown in Fig. 2(a), is proposed. The heating or cooling media flows in the jacket can provide continuous internal heat exchange for the MDC.

This paper presents the experimental results of operating a MDC using commercial poly-propylene membrane for the separation of methanol-water mixture in both adiabatic (A-MDC) and diabatic (D-MDC) modes to demonstrate the enhancement of separation with internal heat exchange. The optimization of internal heat exchange rate for a D-MDC using a ceramic membrane module reported in [14] for the separation of benzene-toluene mixture is studied using a theoretical model of the membrane module. Two approaches are employed for the optimization task. The first one uses a nested loop searching procedure which incorporates one of the three approximations, including equipartition of heat exchange rate (EoQ), equipartition of entropy production (EoEP) and linear distribution of heat exchange rate (LQ). The second one uses an evolutionary approach.

#### 2. Experimental

#### 2.1. Chemicals and hollow fiber modules

Experiments were conducted using 20–40 mol% of aqueous methanol solutions made by blending distilled water and pure methanol. Methanol (99.8%) was purchased from Shimakyu Co., Ltd. (Japan).

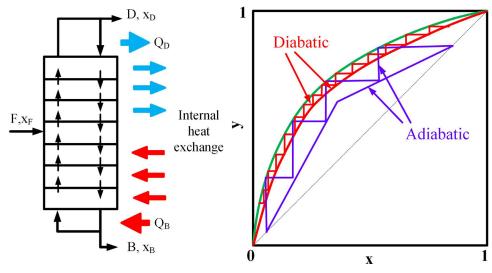


Fig. 1. Diabatic binary distillation column.

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