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OPTIMAL CONFIGURATION, DESIGN AND OPERATION OF CONTINUOUS HYBRID DISTILLATION/ PERVAPORATION PROCESSES

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This paper considers the simultaneous optimisation of configuration, design and operation of continuous hybrid distillation/pervaporation processes by considering all possible process structures. The overall problem is formulated as a mixed integer optimisation (MIO) problem. The optimisation strategy comprises of an overall economics index that encompasses capital investment, operating costs and production revenues. Furthermore, rigorous dynamic models developed from first principles for distillation and pervaporation are used. A case study for the separation of a tangent-pinch (acetone-water) mixture is presented. It is found that a fully integrated hybrid configuration is economically favourable compared to normal distillation.

KEYWORDS: hybrid distillation/pervaporation, optimisation, GA

INTRODUCTION

Distillation is the most commonly used technique for separating liquid mixtures within the chemical industries despite being an energy and capital intensive process. Many mixtures commonly encountered in the fine chemical and pharmaceutical industries are, however, difficult or impossible to separate by normal distillation due to azeotropic behaviour, tangent pinch or low relative volatilities. Pervaporation has been hailed as an alternative to distillation for such mixtures as the separation mechanism is different, relying on differences in solubility and diffusivity between the components in the mixture and not vapour-liquid equilibrium as in distillation. Recently, hybrid processes have been proposed where a distillation column unit and a pervaporation unit are integrated into one process. In such a process, the shortcomings of one method are outweighed by the benefits of the other, allowing for significant savings in terms of energy consumption and cost. Recently, the simultaneous optimisation of configuration, design, and operation of *batch* hybrid distillation/pervaporation has been explored (Barakat and Sorensen, 2005). In this work, the methodology is applied to *continuous* hybrid distillation/pervaporation processes.

The two units can be integrated in different ways; the pervaporation unit can be positioned before the distillation column, after the column, or fully integrated. Depending on the particular separation task, the configuration, design and operation of a hybrid should be optimised to achieve the most suitable performance. Adding a pervaporation unit to the system, either before, after or fully integrated, adds complexity to the system but also more

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degrees of freedom which, if properly chosen, can further increase the profitability of the system, particularly for difficult separations such as that of azeotropic mixtures (Van Hoof *et al.*, 2004).

Eliceche *et al.* (2002) carried out optimisation studies of operating conditions for a continuous hybrid distillation/pervaporation system consisting of an azetropic distillation column connected via a side stream to a pervaporation unit. They solved the optimisation problem by minimising the operating costs, however, they did not consider the design or configuration of the hybrid system. Szitkai *et al.* (2002) optimised the design and operation of a continuous hybrid dehydration system using an MINLP model to minimise the annual operating costs of a single, post-distillation, hybrid configuration. Recently, Kookos (2003) proposed a methodology for structural and parametric optimisation of continuous hybrid separation systems. He described the superstructure of the hybrid process using a simplified steady-state model where it was assumed that all streams taken from, or returned to, the distillation column were vapour streams. The methodology is therefore not suitable for other membrane processes, such as pervaporation, or for dynamic systems.

The design engineer is faced with a difficult task: to determine not only the best design and operation of the separation process, but also which separation technique to use and, if considering a hybrid system, how the two units should be combined. The objective of this work is to propose an optimal process synthesis procedure that allows the optimal determination of the process type, its configuration, design and operation for a given separation duty. This procedure can be extended to include any number of separation process alternatives, but the discussion in this work will be limited to distillation, pervaporation and hybrid distillation/pervaporation processes. In the next section, the separation synthesis problem described in terms of a process superstructure is presented, followed by the objective function formulation and optimisation problem definition. The mathematical models used in this study are then presented together with an overview of the optimisation strategy. Finally, the solution strategy is applied to a case study for the separation of a tangent-pinch mixture (acetone-water).

THE SEPARATION SYNTHESIS PROBLEM

PROBLEM DEFINITION

The objective of the synthesis procedure is to determine the optimal separation process which results in the most economical benefit when processing a given separation task. To achieve this objective, optimal configuration, design and operation must be considered *simultaneously* based on an objective function that encapsulates capital investment, operating costs and production revenues.

There is a trade-off between capital investment in terms of equipment and performance and also between operational decisions and performance. When considering a distillation column for instance, it is possible to design the column with a low number of trays operating at high reflux ratio, or alternatively, to design the column with more trays and operating at lower reflux ratio and still achieve the same separation requirements. The decision will, however, clearly have an impact on the profitability of the process.

SUPERSTRUCTURE

The optimal synthesis of the superstructure is presented next. The superstructure incorporates three separation processes: distillation, pervaporation and hybrid distillation/pervaporation processes and it is applied for a continuous operation mode. The superstructure proposed here allows not only the most economical process configuration to be selected, but also its optimal design and operation which will carry out the required separation duty optimally. Similar work on hybrid superstructures has been proposed by Kookos (2003) but his work only allows for the optimisation of the hybrid process and exploring either distillation or pervaporation as a potential separation process, is therefore not possible.

The membrane separation stage used in this superstructure, as shown in Figure 1, consists of a number of identical pervaporation membrane modules (N_m) connected in parallel (Marriott and Sørensen, 2003). The membrane stage feed stream is assumed to be distributed evenly between the membrane modules and therefore a single mathematical model can be used to describe the modules.

A rigorous distillation column tray model is employed. Each tray is modelled to accommodate for three extra potential streams in addition to the regular vapour and liquid inlet/outlet to the neighbouring trays. The first stream is the feed stream inlet if the tray is selected as a distillation feed tray. The second is a side draw stream *to* the pervaporation unit in a hybrid configuration if the tray is selected as a membrane feed tray. The third stream is an inlet stream *from* the pervaporation unit in a hybrid configuration if the tray is selected as a retentate recycle tray.

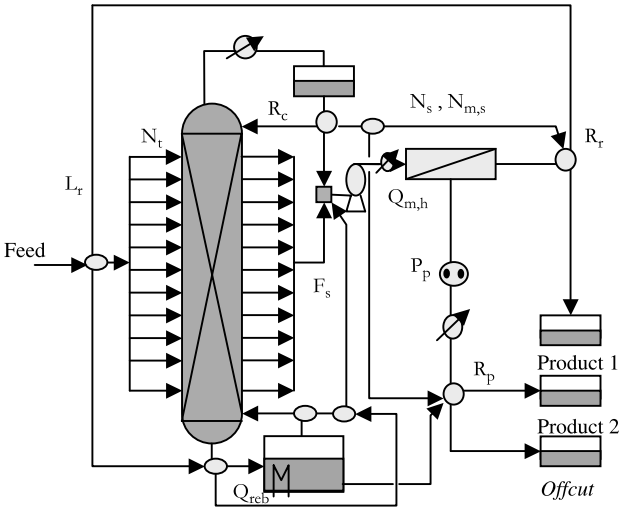

Figure 1. Separation system superstructure

OBJECTIVE FUNCTION

The optimal design and operation of continuous separation processes as it is considered in this paper, is determined as the most economical process design and corresponding operating policy that will satisfy all specified separation requirements and constraints. The optimal solution is a trade-off between capital and operating costs versus production revenue, and is reflected in the formulation of the objective function as shown below:

$$P_A = \left(\frac{\sum_{i=1}^{Nc} C_i M_{i,f} - C_{feed} M_{feed}}{t_f + t_s} \right) \times T_A - ACC - AOC \tag{1}$$

The annualised capital costs and operating costs for the distillation column based on Low and Sørensen (2003) is given by:

$$ACC_c = K_1 N_t^{0.802} V^{0.533} + K_2 V^{0.65} \tag{2}$$

$$AOC_c = C_{ut} \times (Q_{reb} + Q_{c,cond}) \tag{3}$$

The annualised capital costs and operating costs for the membrane process is given by:

$$ACC_m = ACC_m + ACC_{m,anc} \tag{4}$$

$$AOC_m = C_{ut} \times (Q_{m,h} + Q_{m,cond}) + AOC_{m,p} + AOC_{m,t} \tag{5}$$

The annualised capital costs and operating costs for the hybrid distillation column is given by:

$$ACC_{hyb} = ACC_c + ACC_m \tag{6}$$

$$AOC_{hyb} = AOC_c + AOC_m \tag{7}$$

OPTIMISATION PROBLEM FORMULATION

The objective of the synthesis procedure is to maximise the profitability defined by the objective function above, subject to process type, process model equations and all separation duty constraints. The optimisation problem is therefore:

Given a mixture M_{feed} with number of components N_C to be separated, minimum product purities $x_i^{\min}$, minimum product recoveries $M_{i,f}$, price structure of feed and products, C_{feed} & C_i, total production time available per annum T_A; determine the optimum set of design variables u_d, and the optimum set of operation variables u_o, to achieve the maximum objective function value P_A(equation 1):

$$Max_{u_d,u_o} P_A$$

subject to:

$$f(\dot{x},x,t,u_d,u_o) = 0 \tag{8}$$

$$x_i(t_f) \geq x_i^{\min} \quad \forall i = 1,\ldots,N_C \tag{9}$$

$$u_d^{\min} \leq u_d \leq u_d^{\max} \tag{10}$$

$$u_o^{\min} \leq u_o \leq u_o^{\max} \tag{11}$$

Equation (8) represents the mathematical process model of the continuous separation process; x is a vector of process state variables, u_d and u_o denote the vectors of design and operating control variables, respectively. Equation (9) represents the product purity constraints imposed which must be satisfied. Equations (10) and (11) represent the physical and optimisation bounds of the design and operating control variables, respectively.

For the distillation process, the set of operating variables u_o include vapour boilup rate and column reflux ratio profile, *i.e.* $u_o^c = \{V, R_C\}$. The vapour boilup rate can subsequently be used to determine the diameter of the column (*e.g.* using Guthrie's correlation, $D \propto \sqrt{V}$) as well as the reboiler and condenser heat loads. Design variables u_d include the optimal number of trays N_t and location of the feed stream F_t *i.e.* $u_d^c = \{N_t, F_t\}$. For the pervaporation process, the set of operating variables u_o include retentate recycle ratio R_r, permeate pressure P_p and feed tank heat load $Q_{m,h}$, *i.e.* $u_o^m = \{R_r, R_p, Q_{m,h}\}$. The set of design variables u_d include number of membrane modules N_m, *i.e.* $u_d^m = \{N_m\}$. For the hybrid distillation/pervaporation process, the set of operating variables and design variables are a combination of the previous two processes with an additional design variable for the retentate recycle location L_r and the sidedraw location F_s.

PROCESS MODELS

The distillation model is based on the approach of Low and Sørensen (2002) which disposes of some of the common modelling assumptions, such as negligible tray holdup and constant molal overflow that may otherwise have a significant impact on the optimal solution. The main features of the model are: dynamic mass and energy balances and rigorous thermodynamics through the use of liquid and vapour fugacities. The assumptions retained in this work include no entrainment effects, no downcomer dynamics, adiabatic column operation, phase equilibrium and perfect mixing.

The mathematical model used in this study to describe the performance of hollow fibre pervaporation membrane modules is similar to that of Marriott and Sørensen (2003). The model features a 1-D plug flow pattern through the membrane fibres and module shell. Furthermore, dynamic mass and energy balances, as well as rigorous thermodynamics have been included. The membrane characterisation equations are from Tsuyumoto *et al.* (1997). Concentration variation perpendicular to the bulk flow direction is neglected and prefect mixing throughout is also assumed.

The mathematical model of the hybrid distillation/pervaporation is a combination of the distillation and pervaporation models outlined above. It should be noted that the approach outlined in the following can be used with models of any modelling complexity although the confidence in the results will depend on the accuracy of the models.

SOLUTION METHODOLOGY

The simultaneous consideration of optimal design and operation of continuous separation processes as outlined above translates into an optimisation problem with both discrete (*e.g.* number of trays and number of membrane modules) and continuous variables (*e.g.* reflux and recycle ratios). Furthermore, the nonlinear dynamic models used here, as well as the nonlinear objective function defined, transforms the problem into a complex mixed integer optimisation (MIO) problem. These problems are difficult to solve using conventional optimisation techniques, due to the high nonconvexity and complex search space topography of these problems and the combination of integer and continuous variables, and there is much ongoing research on developing robust and practical solution algorithms. In this work, the proposed superstructure is solved using a genetic algorithm (GA) optimisation framework that works through the conventional genetic algorithm operators (further details can be found in Goldberg,1989).

In this work, a given solution set consisting of all decision variables are represented in the genome as direct real values instead of converted binary bits and mapping which has been found to be less efficient (Coley, 1999). The initial population of 100 genomes is created randomly. The objective and constraints of each individual in this population are in this work evaluated using the *gPROMS* simulation software (Process Systems Enterprise Ltd., 2005). A penalty function procedure is applied as described by Low & Sorensen (2003) when necessary to encourage the GA to drive the population towards feasibility. Solutions are assigned a fitness score based on the annual profitability of each genome.

The GA procedure uses a roulette selection scheme, 75% replacement rate, 75% crossover rate, 10% mutation rate and a stopping criterion based on the maximum number of generations of 150. A steady-state population strategy is employed as described in Low & Sorensen (2003). The procedure has been implemented using the GALib genetic algorithm library (Wall, 1999).

RESULTS AND DISCUSSION

The optimal process synthesis procedure developed in this work is demonstrated by considering the separation of a tangent-pinch mixture of acetone and water. The separation process specifications are shown in Table 1.

OPTIMAL SOLUTION

The optimum solution sets of the superstructure, and that of a comparative distillation case, are shown in Table 2. A fully integrated hybrid distillation/pervaporation process

Table 1. Unit specifications and operating conditions

Property	Value	Property	Value
Feed conc., $_{xi,feed}$ (*mol fraction*)		Production time T_A (*hrs/year*)	7920
Acetone	0.50	Setup time, t_s(*min*)	30
Water	0.50	Products purities, $_{xi,f}$ (*mol frac.*)	$\geq$0.97
Feed rate, M_{feed} (*mol/hr*)	18,000	Product recoveries, $M_{i,f}$	$\geq$0.70
Tray/cond. holdup (*mol*)	0.1		

was found to be the optimal synthesis solution. The optimal number of trays (where tray 1 is the top tray of the distillation column) and membrane modules are found to be 21 & 1, respectively. The optimal feed stream location is found to be at tray 20. Optimal reboiler vapour load is found to be 2.44 mol/s with optimal sidedraw flowrate of 4.44 mol/s. The optimal membrane inlet heater temperature is found to be 330 K and permeate side pressure is 500 Pa. Optimal sidedraw location is found to be at tray 1, with optimal retentate return location to the same tray of the hybrid column.

The optimal design and operation of the hybrid distillation/pervaporation is found to be the most profitable process alternative that meets all separation requirements for this case study (see Table 2), with an estimated profit of 21.946 M£ per annum. The optimum process is found to be 7% more profitable than the comparative optimised distillation configuration as also shown in Table 2.

CONCLUSIONS

In this work, the optimal synthesis of continuous separation processes has been considered. The synthesis problem is solved through simultaneous consideration of optimal

Table 2. Optimal solutions sets (*: on upper bound)

Distillation	Hybrid distillation
Optimisation variables	
$u_d = \{N_t, F_t\}$	$u_d = \{N_t,N_m,F_s,L_r,F_t\}$
$u_o = \{M_{feed}, R_C, V\}$	$u_o = \{M_{feed},R_c,R_r,R_p,P_p,T_o,V,F_{side}\}$
Optimum set	
$u_d = \{23,22\}$	$u_d = \{21,1,1,1,20\}$
$u_o = \{5.0,0.75,5.43\}$	$u_o = \{5.0,1.0^*,0.76,0.42,500,330^*,2.44,4.44\}$
Annual profit (£/yr)	
£20,426,000	**£21,946,000**

configuration, design and corresponding operating policy of all process alternatives described through a process superstructure. The optimal solution is then determined as the most economical process configuration, design and operation that achieves all separation requirements. The problem objective function reflects the various trade-offs between design and operation decision variables versus production revenue, as well as that of capital investments versus operating costs.

A hybrid distillation/pervaporation configuration was found to be the optimal synthesis solution for the separation of the equimolar tangent-pinch acetone-water case considered, this was further verified by comparison with an optimised distillation process. The proposed methodology can be extended to allow for the synthesis of any number of separation alternatives by incorporating them into a single process superstructure. However, as alternatives increase, the required computational time to solve such a superstructure will also increase significantly.

NOMENCLATURE

ACC	Annualised equipment capital cost (£/yr)
AOC	Annualised equipment operating costs (£/yr)
C_i	Selling price of product i (£/mol)
C_{feed}	Cost price of feed (£/mol)
C_{ut}	Utilities cost (£/MJ)
F_t	Location of the feed stream
F_s	Location of the column sidedraw
F_{side}	Flowrate of the sidedraw stream
K_1	Guthrie's correlation coeff. for column shell cost
K_2	Guthrie's correlation coeff. for exchangers cost
L_r	Retentate recycle location
M_{feed}	Feed rate (mol/hr)
$M_{i,f}$	Final product i recovery
N_c	Number of components
N_m	Number of membrane modules
N_t	Number of column trays
P_A	Annual profit (£/yr)
P_p	Permeate pressure (Pa)
Q	Heat load (kW)
$Q_{m,h}$	Pervaporation heat load (kW)
R_c	Column internal reflux ratio
R_p	Permeate offcut ratio
R_r	Retentate recycle ratio
t	Time (min)
t_f	Total processing time (min)
t_s	Setup time (min)
T_A	Total production time available per annum

u_d	Vector of design variables
u_o	Vector of operation variables
V	Column boilup rate (mol/sec)
x	Vector of state variables
x_i	Composition of component i in mixture
x_i^{min}	Minimum composition of component i in mixture

SUPERSCRIPTS

c	Column
m	Membrane

SUBSCRIPTS

anc	Ancillary
c	Column
cond	Condenser
m	Membrane
reb	Reboiler
m,h	Pervaporation membrane system feed heater
m,t	Pervaporation membrane system turbine
m,p	Pervaporation membrane system feed pump
hyb	Hybrid system
a	Stage

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