DESIGN TECHNICAL REPORT – EXAMPLE 2

An example where a list of publications and two papers (one on "Optimal synthesis and design of hybrid batch separation processes" and the other on "The optimal design of membrane systems") have been provided to meet IChemE's design requirement.

B. Refereed journal and conference articles

1. Sørensen, E. and S. Skogestad (1994), "Control strategies for reactive batch distillation", *Journal of Process Control*, **4**(4), 205-217.
2. Sørensen Leversund, E., S. Macchietto, G. Stuart and S. Skogestad (1994), "Optimal control and on-line operation of reactive batch distillation", *Computers Chem. Engng.*, **18**(Suppl), 391-395.
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13. Rotstein, G., V.T. Voudouris, E. Sørensen, N. Shah, S. Macchietto, R.A. Weiss and H. Dempster (1997), "Process Concept Studio: An experiment on academic/industrial collaboration", *Computers Chem. Engng.*, **21**(Suppl), 319-324.
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15. J.I. Marriott, E. Sørensen, I.D.L. Bogle (1999), "Rigorous optimal design of a pervaporation plant", In *Proc. FOCAPD*, 415-418.
16. Sørensen, E. (1999), "A cyclic operating policy for batch distillation - Theory and practice", *Computers Chem. Engng.*, **23**, 533-542.
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19. Marriott, J.I., E. Sørensen and I.D.L. Bogle (2000), "Detailed mathematical modelling of membrane modules", In *Proc. ESCAPE'10*, 523-528, Florence.
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Optimal synthesis and design of hybrid batch separation processes

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ABSTRACT

This paper proposes a procedure for the optimal determination of process synthesis and design for batch separation of mixtures that are difficult to separate by conventional batch distillation. The procedure allows the determination of the optimal batch process, its configuration and its design to perform a given separation duty from available process alternatives. In this work, batch distillation, batch pervaporation and batch hybrid distillation/pervaporation processes are considered. Since a batch process is inherently dynamic, and the configurational decisions are integer in nature, the overall problem is formulated as a mixed integer dynamic optimisation (MIDO) program. The optimisation strategy takes into account an overall economic index that encompasses capital investment, operating costs and production revenues. Furthermore, rigorous mathematical models developed from first principles for distillation and pervaporation are used. Two studies for the separation of acetone-water mixtures of different feed specifications are presented and it was found that batch hybrid distillation/pervaporation is the optimal synthesis solution in both cases.

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. Pervaporation is still in general a more costly process than distillation, however, for some separations, the costs are comparable or in favour of pervaporation. Recently, hybrid processes have been proposed where a distillation column unit and a pervaporation unit are integrated into one process (Lipnizki *et al.,* 1999). 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. Guerreri (1992) and Van Hoof *et al.* (2003) reported up to 60% reduction in energy consumption in their studies of separation of azeotropic mixtures using hybrid systems.

The two units can be integrated in different ways; the pervaporation unit can be positioned before the column, after the column or fully integrated (Fig. 1). One example of a fully integrated system is to position the pervaporation unit after the column reflux drum outlet and recycle the unit retentate stream back to an appropriate location in the column (see Fig. 1). Further examples of how the two units can be integrated are found in Lipnizki *et al.* (1999).

Most separations within the fine chemical or pharmaceutical industries are run batch-wise. The optimal design and operation of batch distillation columns has received considerable interest in recent years, particularly, in terms of novel column configurations such as inverted, middle vessel and multivessel column configurations (Furlonge *et al.* (1999) and Low and Sørensen (2002, 2003)). Adding a pervaporation unit to the system, either before, after or fully integrated, adds complexity to the system but also more 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.*, 2003).

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The design engineer is thus 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 determination of the optimal process type, its configuration and design for a given separation duty. This procedure can be extended for any number of separation process alternatives, but the discussion in this work will be limited to batch distillation, batch pervaporation and batch hybrid distillation processes. In the next section, the batch separation synthesis problem is presented, followed by the objective function formulation and optimisation problem definition. The mathematical models used in this study are then presented as well as an overview of the optimisation strategy. Finally, the batch separation process synthesis problem and its solution strategy are applied to two case studies: the separation of a tangent-pinch mixture (acetone-water) of two different feed specifications.

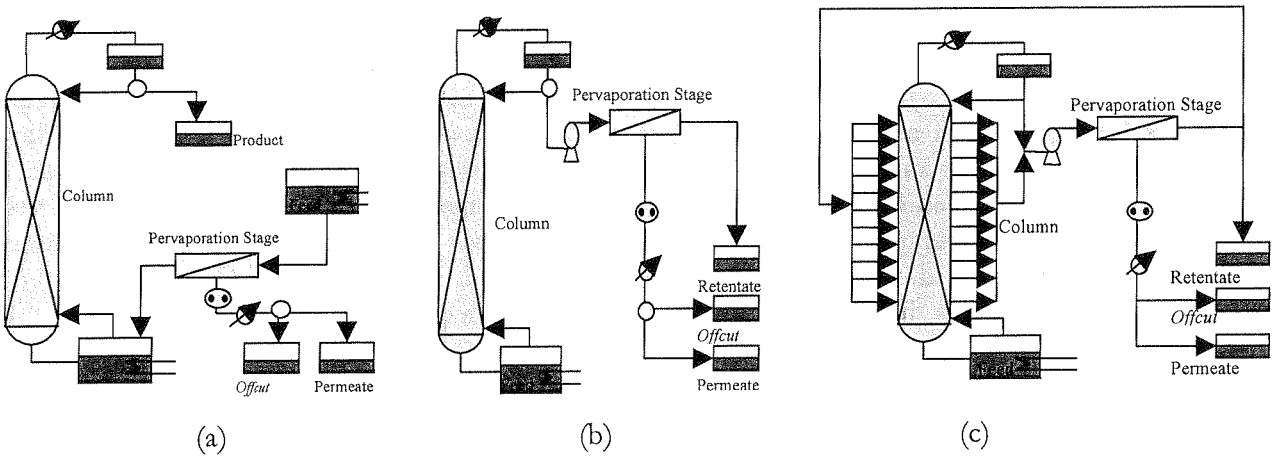

Figure 1, (a) pre-distillation hybrid, (b) post-distillation hybrid, (c) example of integrated hybrid system

THE BATCH SEPARATION SYNTHESIS PROBLEM

Problem Definition

The objective of batch separation process synthesis 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 a unified objective function that encapsulates capital investment, operating costs and production revenues. Unified production constraints must also be applied to ensure a fair comparison between process alternatives. Synthesis of batch separation processes is complex due to the interdependency exhibited between design and operational issues that need to be considered simultaneously (Fig. 2).

There is a trade-off between capital investment in terms of equipment and performance and also between operational decisions and performance. When considering a batch distillation column for instance, it is possible to design the column with a low number of trays operating at high reflux ratio, or alternatively, 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. The synthesis problem is further complicated when the process is designed for multiple separation duties. This is commonly the case in industry as batch processes are utilised as multipurpose units because of their flexibility to accommodate changes in separation duty. In this case, the optimal product synthesis tends to be biased toward the separation duties that are performed more frequently than others. High product recovery generally leads to longer batch times than does low recovery. However, high recovery operation generally incurs higher operating costs and thus the balance between revenue and operating costs must be considered.

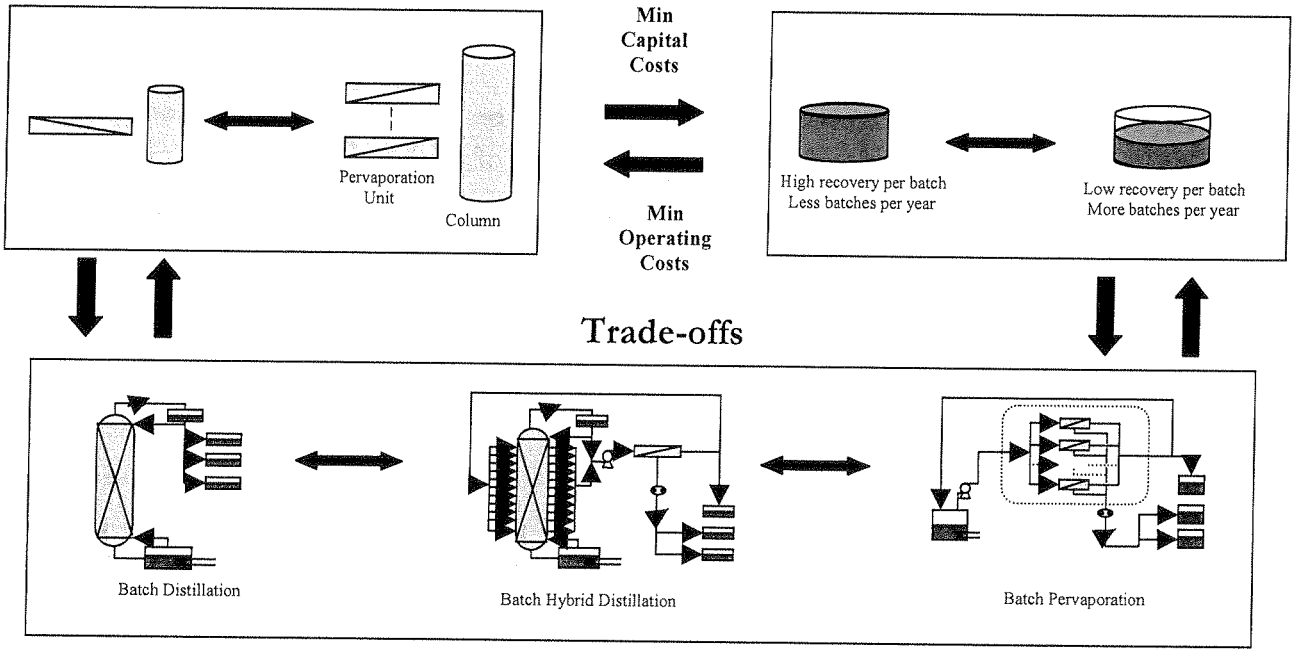

Figure 2, batch separation synthesis problem

Objective Function

The optimal design and operation of batch 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 batch distillation column (Low and Sørensen, 2003) is given by:

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

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

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

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

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

The annualised capital costs and operating costs for the hybrid batch 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 (eq 1):

$$Max_{u_d,u_o} P_A$$

subject to:

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

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

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

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

Equation (8) represents the mathematical process model of the batch 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 at the end of the batch. Equation (10) and (11) represent the physical and optimisation bounds of the design and operating control variables, respectively.

For the batch distillation process, the set of operating variables 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 include the optimal number of trays N_t, *i.e.* $u_d^c = \{N_t\}$.

For the batch pervaporation process, the set of operating variables 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 include number of membrane modules N_m, *i.e.* $u_d^m = \{N_m\}$. For the hybrid batch distillation 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.

Process Models

Following the approach of Low and Sørensen (2002), a similar mathematical model is in this paper used to describe the batch distillation systems. The model 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 as demonstrated by Furlonge *et al.* (1999) and Low and Sørensen (2002). The main features of the model are:

- Dynamic mass and energy balance to dispose of the usual assumption of negligible tray holdup and constant molal overflow, respectively.
- Rigorous thermodynamics through the use of liquid and vapour fugacities that are functions of temperature and pressure, this disposes of the common assumption of constant relative volatility.

The batch distillation model 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 *et al.* (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 batch hybrid distillation is a combination of the batch 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

Simultaneous consideration of optimal design and operation of batch separation processes 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 dynamic optimisation (MIDO) problem. MIDO problems are difficult to solve using conventional optimisation techniques due to the high nonconvexity and complex search space topography nature of these problems and the combination of integer and continuous variables, and there is much ongoing research on developing robust and practical solution algorithms (Low and Sørensen, 2004). The case studies presented in this work were solved using the *gPROMS* simulation and optimisation software (Process Systems Enterprise Ltd., 2004). *gPROMS* implements a backward differentiation (*BD*) technique to solve the model set of nonlinear differential and algebraic equations (*DAEs*). The optimisation algorithm is based on decomposing the main problem into primal dynamic optimisation and master mixed-integer linear programming subproblems, details of this algorithm can be found in Bansal *et al.* (2003).

CASE STUDIES

In this section, two case studies for the separation of acetone-water mixtures of different feed concentrations are presented to demonstrate the procedure outlined in the previous section. The objective is to determine the optimal design and operation of the three processes under consideration (batch distillation, pervaporation or a hybrid of the two), and then to compare these optimal solutions (based on annual profit measures given by the objective function in equation 1) to determine the most economically favourable process for the given separation task. Note that only a single separation stage is considered in this work although multistage batch pervaporation processes are possible and may have higher economical potential.

Case Study I

For the first case study, the separation of an 85:15 mole% acetone-water mixture is considered. The objective is to determine the most appropriate process for the separation of the mixture assuming product constraints on both products. Tables 1 and 2 summarise unit specifications and operation conditions and the optimisation decision variable bounds used, respectively.

The batch distillation operation is split into two intervals, an acetone product collection period and an offcut period for water product purification. The optimal solution will have an appropriate balance between the time intervals as well as determine the total batch time which is the sum of these intervals.

For the batch pervaporation process, a polyion complex membrane type reported by Tsuyumoto *et al.* (1997) is used. The membrane preferentially permeates water and the module is constructed as a highly porous (75%) hollow-fibre bundle, further specification of these modules are found in table 3. Membrane characterisation equations reported by Tsuyumoto *et al.* are used in the modelling of the pervaporation membrane modules described in the pervious section. Although Tsuyumoto *et al.* (1997) only described the pervaporative separation of an ethanol-water mixture, it is considered a reasonable assumption to use these equations to describe the separation of the acetone-water mixture. The batch pervaporation process operation is separated into three intervals, a water product collection period to

purify the acetone product, followed by a possible offcut period and a final acetone product dumping period (into the retentate product tank) when acetone reaches the required product purity. During the second period, the existence of an offcut is decided.

The hybrid distillation operation is also separated into two task intervals, the first period is for water product collection while the acetone product is purified to the required purity, followed by an acetone product collection period.

Optimal Solution

The optimum solution set is shown in table 4. For batch distillation, the optimal number of trays and reboiler vapour load are found to be *27* & *1.97 mol/s*, respectively. The batch time is found to be *4.8 minutes* with interval split of 4.7 & 0.1 *minutes* and corresponding constant reflux ratio values of *0.70* & *0.97*, respectively.

The batch pervaporation process has an optimal number of one membrane module. The batch time is lower than that of batch distillation due to the low water contents on the feed side. The permeate pressure is found to be *300 Pa* and the feed flowrate is *5 mol/s*. As expected, the optimal permeate pressure is at its lower bound while the feed flowrate is at its upper bound allowing for the maximum possible flux through the membrane and thus purification of the acetone retentate stream in the shortest possible time.

For hybrid distillation, the optimal number of trays and membrane modules are found to be *11* & *1*, respectively. Optimal batch time is found to be *6.7 minutes* with interval split of *3.4* & *3.3 minutes*. Corresponding reflux and recycle ratios are found in table 4. Optimal reboiler vapour load is found to be *1.31 mol/*s and the optimal membrane permeate side pressure is *374 Pa*.

The optimal design and operation of batch hybrid distillation is found to be the most profitable processes alternative that meets all separation requirements for this case study (see table 1), with an estimated profit of £1,002,453 per annum. The batch distillation column configuration is found to be the least profitable to perform this separation (£749,231). The batch pervaporation process configuration is found to give an annual profit figure of £776,759. This figure is slightly higher than that of batch distillation due to the high efficiency of membrane separation for low water contents feed streams. Batch hybrid distillation configuration is therefore selected as the most suitable process to perform this separation. The optimal hybrid process flowsheet for this case is shown in Figure 3.

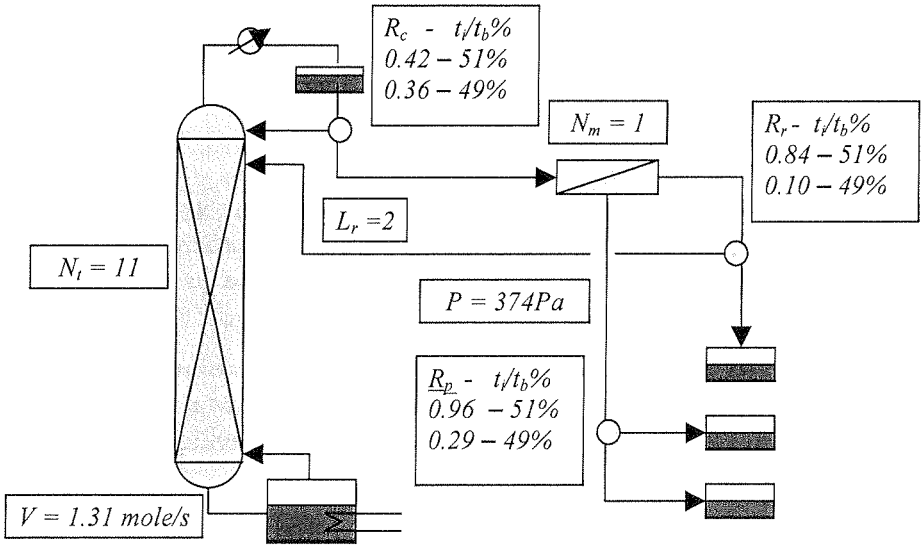

Figure 3, flowsheet of optimal hybrid process for case I.

Property	Case I/II
Feed composition, $x_{i,feed}$ *(mol fraction)*	
Acetone	0.85/0.50
Water	0.15/0.50
Batch size, M_{feed} *(mol)*	200
Tray/Stage holdup *(mol)*	0.1
Available production time T_A *(hr)*	8760
Batch setup time, t_s *(min)*	30
Cost, C_i *(£/mol)*	
Acetone	0.606
Water	0.040
Feed	0.250/0.150
Offcut	0.010
Utility *(£/MJ)*	0.019
Product purity specifications, $x_{i,f}$ *(mol frac.)*	
Acetone	0.97
Water	0.97
Product recoveries, $M_{i,f}$	>70%
ACC_m *(£/yr)*	500
$ACC_{m,anc}$ *(£/yr)*	250

Table 1, unit specifications and operating conditions

Decision variable	Bounds
N_t	[10,30]
N_m	[1,10]
L_r	[1,5]
t_f, t_i *(min)*	[0.1,1000]
R_r, R_c, R_p	[0,1]
V*(mole/s)*	[0.5,10]
P_p *(Pa)*	[300,4000]
F_{feed} *(mole/s)*	[0.5,5]

Table 2, optimisation variable bounds

Property	Value
Membrane material	Polyion complex
Support material	Polyacrylonitrile
Module radius, *(m)*	97.4 x 10^{-3}
Fibre inner radius, *(m)*	0.251 x 10^{-3}
Fibre outer radius, *(m)*	0.395 x 10^{-3}
Membrane thickness, *(m)*	0.015 x 10^{-3}
Fibre length, *(m)*	1.000
Number of fibres	3800

Table 3, hollow fibre module specifications (Tsuyumoto, et al., 1997)

Distillation	Pervaporation	Hybrid Distillation
Optimisation Variables,		
$u_d = \{N_t\}$	$u_d = \{N_m\}$	$u_d = \{N_t, N_m, L_r\}$
$u_o = \{t_f, t_i, R_c, V\}$	$u_o = \{t_f, t_i, R_r, R_p, P_p, F_{feed}\}$	$u_o = \{t_f, t_i, R_r, R_c, R_p, P_p, V\}$
Optimum Set,		
$u_d = \{27\}$	$u_d = \{1\}$	$u_d = \{11,1,2\}$
$u_o = \left\{4.8, \begin{bmatrix}4.7\\0.1\end{bmatrix}, \begin{bmatrix}0.70\\0.97\end{bmatrix}, 1.97\right\}$	$u_o = \left\{4.4, \begin{bmatrix}1.7\\2.1\\0.6\end{bmatrix}, \begin{bmatrix}1\\1\\0\end{bmatrix}, \begin{bmatrix}0.2\\0.6\\0.5\end{bmatrix}, 300, 5\right\}$	$u_O = \left\{6.7, \begin{bmatrix}3.4\\3.3\end{bmatrix}, \begin{bmatrix}0.84\\0.10\end{bmatrix}, \begin{bmatrix}0.42\\0.36\end{bmatrix}, \begin{bmatrix}0.96\\0.29\end{bmatrix}, 374, 1.31\right\}$
Annual Profit (/year),		
£749,231	**£776,759**	**£1,002,453**

Table 4, optimal solutions sets for case study I

Case Study II

For this case study, the separation of a 50:50 mole% acetone-water mixture is considered. The objective is again to determine the most appropriate process for the separation of the mixture considered. All other specifications are the same as for case study I. Unit specifications and operating conditions used are found in table 1.

Optimal Solution

The optimum solution set is shown in table 5. For batch distillation, the optimal number of trays and reboiler vapour load are found to be *23* & *0.65 mol/s*, respectively. The batch time is found to be *8.8 minutes* with interval split of *8.6* & *0.2 minutes* and corresponding reflux ratio values of *0.70* & *0.71*, respectively. Comparing this to case study I, the optimal batch time has doubled due to the decreased reboiler vapour load from *1.97* to *0.65 mol/s*. The number of trays, however has decreased to *23*.

For the batch pervaporation process, a single membrane module is found to perform the separation duty optimally. The batch time is higher than that of case study I, which is due to a higher water amounts in the feed mixture and thus slower purifications of the acetone retentate stream. The permeate pressure is found to be *369 Pa* and the feed flowrate is *2.3 mol/s*.

For hybrid distillation, the optimal number of trays and membrane modules are found to be *17* & *1*, respectively. Optimal batch time is found to be *9.8 minutes* with interval split of *3.3* & *6.5 minutes*, corresponding reflux and recycle ratios are found in table 5. Optimal reboiler vapour load is found to be *0.65 mol/*s and the optimal membrane permeate side pressure is *585 Pa.*

The optimal design and operation of batch hybrid distillation is found to be the most profitable processes alternative that meets all separation requirements for this case study (see table 1), with an estimated profit of £428,237 per annum. The batch distillation column configuration is found to give slightly lower annual profit figure of £393,567. The batch pervaporation process configuration is found to be the least profitable process (£226,410) when compared to batch distillation and hybrid configuration. Batch hybrid distillation configuration is therefore the most suitable process to perform this separation.

Distillation	**Pervaporation**	**Hybrid Distillation**
Optimisation Variables,		
$u_d = \{N_t\}$	$u_d = \{N_m\}$	$u_d = \{N_t, N_m, L_r\}$
$u_o = \{t_f, t_i, R_c, V\}$	$u_o = \{t_f, t_i, R_r, R_p, P_p, F_{feed}\}$	$u_o = \{t_f, t_i, R_r, R_c, R_p, P_p, V\}$
Optimum Set,		
$u_d = \{23\}$	$u_d = \{1\}$	$u_d = \{17,1,2\}$
$u_o = \left\{8.8, \begin{bmatrix}8.6\\0.2\end{bmatrix}, \begin{bmatrix}0.70\\0.71\end{bmatrix}, 0.65\right\}$	$u_o = \left\{17.3, \begin{bmatrix}8.3\\8.3\\0.7\end{bmatrix}, \begin{bmatrix}1\\1\\0\end{bmatrix}, \begin{bmatrix}0.1\\0.9\\0.9\end{bmatrix}, 369, 1.97\right\}$	$u_O = \left\{9.8, \begin{bmatrix}3.3\\6.5\end{bmatrix}, \begin{bmatrix}1.0\\0.0\end{bmatrix}, \begin{bmatrix}0.2\\0.6\end{bmatrix}, \begin{bmatrix}1.0\\0.9\end{bmatrix}, 585, 0.65\right\}$
Annual Profit (/year),		
£393,567	£226,410	£428,237

Table 5, optimal solutions sets for case study II

CONCLUSIONS

In this work, the optimal synthesis of batch separation processes has been considered. The synthesis problem is solved through simultaneous consideration of optimal design and corresponding operating policy of all process alternatives. These optimal solutions are then compared in order to determine the most economical process 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.

Hybrid batch distillation configuration was found to be the optimal synthesis solution for both cases considered. It was found that for the low purity feed specification (50:50 acetone-water), a batch distillation process was more effective and thus more profitable, than a batch pervaporation process. This was not the case, however, for the higher purity feed specification (85:15 acetone-water), when the batch pervaporation process was more profitable than a distillation process.

NOMENCLATURE

ACC	Auualised equipment capital costs (£/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_{feed}	Membrane stage feed rate (mol/s)
K_1	Guthrie's correlation coefficient for column shell cost
K_2	Guthrie's correlation coefficient for column exchangers cost
L_r	Retentate recycle location
M_{feed}	Batch feed size (mol)
$M_{i,f}$	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_f	Total batch processing time (min)
t_i	Task interval time (min)
t_s	Batch setup time (min)
T_A	Total production time available per annum
u	Vector of control variables
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
h	Pervaporation membrane system feed heater
m	Membrane
reb	Reboiler
m,t	Pervaporation membrane system turbine
m,p	Pervaporation membrane system feed pump
hyb	Hybrid system

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The optimal design of membrane systems

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Abstract

This paper introduces a novel optimal design strategy for membrane separation systems. This strategy is characterised by two main features: firstly, detailed models are used. This is essential if sub-optimal and inaccurate solutions are to be avoided. Secondly, an optimisation technique based on genetic algorithms is implemented. The feasibility of the optimal design strategy is investigated using a pervaporation case study. To this end, an existing pervaporation plant is evaluated and a significantly improved design is found.
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1. Introduction

During the last 30 years, the search for viable alternatives to traditional energy intensive methods such as distillation, has led to the introduction of separation processes based on membranes. Membrane technology often offers cheaper capital and utility costs and has displaced conventional separation techniques in many areas. Wider use is expected in the future (Ho & Sirkar, 1992). The increasing market for membrane technology has motivated interest in the development of reliable design strategies that aim to optimise the economic performance of membrane separation systems (El-Halwagi, 1992; Qi & Henson, 2000).

In many areas of chemical process design and operation, the use of mathematical models for process synthesis, optimisation and for control studies has shown significant benefit. These methods are beginning to be applied to the design of membrane processes (El-Halwagi, 1992; Qi & Henson, 2000) and are explored further in this paper.

To realise the full potential of membrane technology, it is important that all available degrees of freedom are explored when a separation system is designed. Although simulation studies are useful for this purpose, they are unable to account for the interacting nature of all the design choices. Therefore, in this research, optimisation techniques are employed to determine the best way to design membrane separation systems.

Conventionally, chemical engineering optimisation problems are solved using gradient-based solution techniques. These return a single, sometimes sub-optimal, solution and provide little insight into the design problem. In reality, design engineers are often faced with a number of qualitative decisions that cannot easily be described mathematically. This research establishes a solution technique which provides multiple solutions at the end of the optimisation allowing the user to interpret the results and make an informed decision.

Although simple unit models have previously been used to optimise the operating conditions of a given membrane system (e.g. Ji, Hilaly, Sikdar, & Hwang, 1994; Tessendorf, Gani, & Michelsen, 1999), full structural optimisation has only been carried out using approximate models (e.g. El-Halwagi, 1992; Srinivas & El-Halwagi, 1993; Qi & Henson, 2000). Unfortunately, inaccuracies in modelling the membrane modules will lead to the development of sub-optimal plant designs with the possible over (or under) prediction of plant performance and a lack of generality due to implicit assumptions. In this work, the use of rigorous mathematical models (verified against experimental data) for the optimal design of membrane systems is explored.

2. Optimisation methods

The performance of a membrane process is limited by the magnitude of the chemical potential driving force for

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mass transfer. This driving force can be maximised by manipulating the temperature or pressure of the feed stream as well as the pressure of the permeate outlet. In a pervaporation plant, it is usual to control the permeate pressure and feed temperature; whereas for reverse osmosis and gas separation plants, it is the feed pressure which is generally controlled.

The manipulation of stream pressures and temperatures can demand significant energy and is costly. When designing a membrane separation system, this expenditure must be balanced against other costs, such as the cost of the membranes and other capital outlay. The interactive nature of all these choices is complex and, consequently, formal optimisation techniques are required to determine the best values for the various design parameters.

To consider a design problem in its entirety, a formulation that enables all decision choices to be considered simultaneously must be produced. This is done by generating a *superstructure* of design alternatives.

2.1. Solution of superstructure optimisation problems

A process can be described by a set of individual unit models connected in the correct sequence by information streams. Therefore, a *process superstructure* can be rigorously generated by considering all possible units and allowing stream connections between all of these units. Generally, both discrete and continuous variables are required: discrete variables may describe whether or not a unit exists in a process, and continuous variables may describe unit operation and stream flow rates.

An optimisation problem requires that the value of a given objective function be maximised or minimised, subject to equalities such as those derived from mass and energy balances, and to inequalities such as product specifications. The objective of a design problem is usually to select the process flowsheet with the minimum annualised plant cost. The annualised plant cost incorporates both capital and operating costs.

In general, such problems require the solution of equations containing both linear and non-linear functions in discrete and continuous variables. This is referred to as a mixed integer non-linear programming (or MINLP) problem. The solution of MINLP optimisation problems is complex. Conventional MINLP solution methods involve disassociating the integer and binary variables from the continuous variables and solving the resulting NLP sub-problems using a gradient-based approach. A variety of approaches have been developed for this purpose, these include: enumeration of all possible integer values; branch and bound methods; generalised benders decomposition and outer-approximation methods. All these approaches have been well documented in the literature over the last 15 years—Biegler, Grossmann, and Westerberg (1997) serves as a very good general reference.

The NLP sub-problems are usually solved using successive quadratic programming methods or reduced gradient methods (Biegler et al., 1997), for which many commercial algorithms are available. In many cases, the NLP problems involve non-convexities that may give rise to a number of local optima. Currently, whilst there have been considerable recent advances (Grossmann & Daichendt, 1996; Adjiman, Androulakis, & Floudas, 1997), a method that guarantees a global optimum solution is not yet available. Therefore, several optimisations at different starting points must be carried out to determine whether the global optimum has indeed been identified.

Unfortunately, many design problems are highly non-convex and gradient-based methods will only converge to the closest local optimum. Furthermore, gradient-based methods are unable to handle discontinuities correctly and can be computationally expensive as they require derivative information (Androulakis & Venkatasubramanian, 1991).

The application of alternate stochastic methods has been explored by a number of researchers: in particular the use of genetic algorithms for the design of process plants has been found to be beneficial in several cases (Androulakis & Venkatasubramanian, 1991; Fraga & Senos Matias, 1996; Garrard & Fraga, 1998). However, the use of genetic algorithms for the design of membrane systems has not previously been considered. Whilst generally slower than gradient-based solution techniques when used for solving simple optimisation problems, they potentially offer a number of important advantages (Garrard & Fraga, 1998):

- They do not require a smooth search space—i.e. discontinuous functions can easily be handled as derivative information is not required.
- They perform a global search and are less likely to get trapped in local optima.
- They can cope with both continuous and discrete variables.
- Multiple solutions are available at the end of the optimisation.

For these reasons, we investigate the use of genetic algorithms for the optimal design of membrane systems in this paper.

2.2. Genetic algorithms

Genetic algorithms are used for a range of purposes from solving practical optimisation problems to providing computational models of natural evolutionary systems (Mitchell, 1996). These were first investigated by Holland (1975) who devised methods to introduce the mechanisms of natural adaption into computer systems.

In a genetic algorithm (borrowing names by analogy from their biological counterparts), *genes* are used to encode optimisation decision variables. A *genome* is the set of genes which uniquely encodes a candidate solution to the

optimisation problem. The genes are of variable type, so they can hold continuous values, be selected from an allele set or even be complex data structures such as graphs.

A genetic algorithm is usually initialised by filling the population with randomly generated genomes. Each subsequent generation is populated by applying genetic operators, such as *mutation* and *crossover*, to the best individuals from the previous generation. Essentially, genetic operators stochastically adjust the value of one or more genes in order to generate a new set of optimisation decision variables. Mutation is used to introduce new information and crossover combines information from two existing solutions. The algorithm can continue evolving the population in this manner indefinitely. However, often stopping criteria such as a maximum number of generations or population convergence, are used.

The genetic algorithm uses the fitness of each individual to determine which individuals should survive, which should reproduce and which should die. The fitness is usually the objective function of the optimisation problem.

In the first part of this paper, we outline a novel strategy based on genetic algorithms for the optimal design of membrane separation systems. In the second part of the paper, the application of the method is demonstrated using a pervaporation case study.

3. Optimal design strategy for membrane separation systems

A membrane separation process consists of a large number of membrane units as well as ancillary equipment which may include heat exchangers, pumps and compressors. The purpose of the ancillary equipment is to control the transport of material through the membrane. This is done by changing the state (i.e. pressure or temperature) of the process streams.

A process superstructure of design alternatives is rigorously generated by considering all possible units and allowing stream connections between all of these units. Such an approach usually leads to very large problems. Therefore, it is necessary to formulate the problem carefully, in order to minimise computational expense.

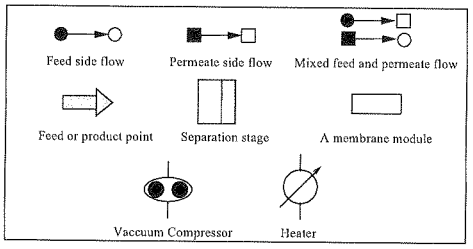
3.1. Membrane process superstructure

The membrane process is conceptualised using separation stages. A *separation stage* is defined simply as a number of identical membrane modules connected in parallel (discussed later).

The process superstructure is then built from three *unit* types: N^s separation stages, N^f feed points and N^p product points. The features of each unit type are shown in Table 1. Most membrane systems contain a single feed and two (permeate and retentate) product points. A superstructure for a process containing two separation stages is illustrated in Fig. 1—which also includes a key to all the figures in this paper.

Table 1
The unit types used to build a superstructure for a membrane separation process

Unit type	No.	No. of inlets per unit	No. of outlets per unit
Separation stage	N^s	1	2
Feed point	N^f	0	1
Product point	N^p	1	0



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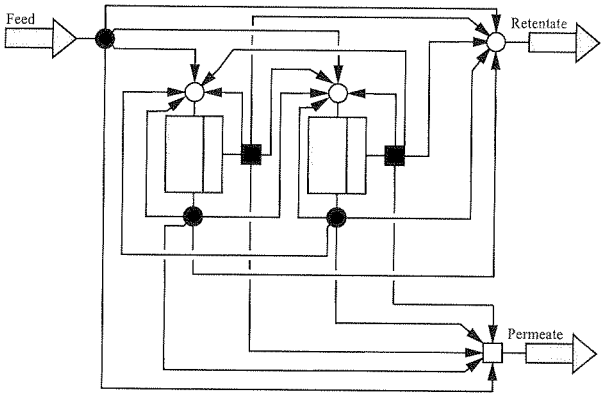

Fig. 1. A process superstructure for two separation stages.

To define the superstructure, we must allow for all possible network choices, this is done using streams. These allow a flow of material from a unit outlet to any unit inlet. For the example shown in Fig. 1, material from the feed outlet can potentially go to the retentate product, permeate product, and the two separation stages. The thermodynamic state, S, (temperature and/or pressure) of the fluid is manipulated to the conditions of the destination unit. The minimum cost (C) for this transformation is calculated using the ancillary equipment models (the appendix).

$$C^{\text{fixed}} = f_1(\Delta S), \tag{1}$$

$$C^{\text{op}} = f_2(\Delta S). \tag{2}$$

The functions can be in any form—this is specified by the ancillary equipment models. For example, the fixed cost may be a step function: 0 if $\Delta S = 0$ and 10 if $\Delta S > 0$. In this work, two types of ancillary equipment are considered:

Table 2
The main ancillary equipment requirements for different membrane processes

Separation process	Main driving force for mass transport	Main ancillary equipment
Gas separation	Pressure	Compressors
Reverse osmosis	Pressure	High-pressure pumps Energy recovery devices
Pervaporation	Concentration	Heaters Vacuum compressors

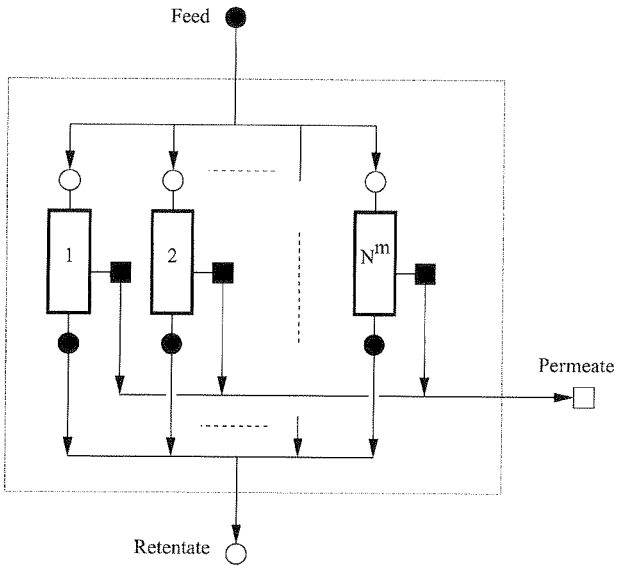

Fig. 2. A separation stage.

pressurisation/depressurisation devices; and heating/cooling devices—see Table 2.

This new approach concentrates on the interacting choices between the number of modules and the driving force for transport across the membrane. As the ancillary equipment models are embedded in the stream model, the number of potential stream connections that are required is minimised. Furthermore, it is not necessary to introduce constraints to forbid two manipulators operating on the same stream (as required in alternate approaches, see for example the state space approach used by El-Halwagi, 1992).

3.1.1. Separation stage

A separation stage consists of a number of identical membrane modules (N^m) connected in parallel. This is shown for a general system in Fig. 2. Assuming that the feed is distributed evenly between them, a single mathematical model can be used to describe the set of parallel membrane modules in each stage. This approach requires a significantly smaller computational effort than if a separate model was used to describe each module. For each stage, the state (pressure or temperature) of the feed stream and pressure of the permeate product must be specified. This enables the driving force across the membrane to be controlled.

Table 3
Number of optimisation decision variables for different superstructure sizes

System size	Number of stage variables	Number of network variables	
		Full	Simplified[a]
One stage	3	6	2
Two stages	6	15	6
Three stages	9	28	12
Four stages	12	45	20

[a]Simplified network assumes that the permeate and retentate streams cannot be mixed.

For each stage, the optimiser is free to manipulate the state (S) of the feed and permeate streams and the number of membrane modules (N^m). These can be optimised directly or selected from an allele set. Each choice may have associated capital and operating cost implications. For example, changing the pressure of a stream will introduce the unit cost of a pump as well as the cost of pressurising the fluid.

3.1.2. Network choices

As each unit outlet can potentially send flow to any unit inlet, the total number of streams from a unit outlet is equal to the total number of unit inlets ($N^i = N^p + N^s$). If each unit outlet is assigned a number a and each unit inlet a number b, we can define the split fraction, $\sigma_{a,b}$ as the fraction of material leaving unit outlet a that goes to unit inlet, b. This is calculated

$$m_{a,b}^{\text{in}} = \sigma_{a,b} m_a^{\text{out}}. \tag{3}$$

However, as the mass balance must hold at each unit outlet, a, we can write

$$\sum_{b=0}^{N^i-1} \sigma_{a,b} = 1. \tag{4}$$

Therefore, we can impose the following constraint on the bounds of each split fraction to ensure that Eq. (4) is not violated:

$$0 \leqslant \sigma_{a,b} \leqslant \max\left(\left(1 - \sum_{b=0}^{b=N^i-2} \sigma_{a,b}\right), 0\right). \tag{5}$$

The structure of the system is fixed by specifying the split fractions (σ) for each unit outlet. It is clear that despite using separation stages, a large number of choices must be taken. This was illustrated earlier in Fig. 1. For a general superstructure $\{(N^f + 2N^s) \times (N^p + N^s - 1)\}$ network decisions are required. The number of optimisation decision variables for one-, two-, three-, and four-stage systems is shown in Table 3. Fortunately, in many cases, it is

Table 4
Genome encoding the decision variables for a two separation stage system

Decision variable	Lower bound	Upper bound
Stage decision variables		
Number of modules, stage 1	$n^{\min}$	$n_m^{\max}$
Normalised feed state, stage 1	0	1
Normalised permeate state, stage 1	0	1
Number of modules, stage 2	$n^{\min}$	$n_m^{\max}$
Normalised feed state, stage 2	0	1
Normalised permeate state, stage 2	0	1
Split fractions		
Feed to stage 1, $\sigma_{0,1}$	0	1
Feed to stage 2, $\sigma_{0,2}$	0	$1-\sigma_{0,1}$
Feed to retentate product, $\sigma_{0,3}$	0	$1-(\sigma_{0,1}+\sigma_{0,2})$
Stage 1 retentate to stage 1, $\sigma_{1,1}$	0	1
Stage 1 retentate to stage 2, $\sigma_{1,2}$	0	$1-\sigma_{1,1}$
Stage 1 retentate to retentate product, $\sigma_{1,3}$	0	$1-(\sigma_{1,1}+\sigma_{1,2})$
Stage 1 permeate to stage 1, $\sigma_{2,1}$	0	1
Stage 1 permeate to stage 2, $\sigma_{2,2}$	0	$1-\sigma_{2,1}$
Stage 1 permeate to permeate product, $\sigma_{2,3}$	0	$1-(\sigma_{2,1}+\sigma_{2,2})$
Stage 2 retentate to stage 1, $\sigma_{3,1}$	0	1
Stage 2 retentate to stage 2, $\sigma_{3,2}$	0	$1-\sigma_{3,1}$
Stage 2 retentate to retentate product, $\sigma_{3,3}$	0	$1-(\sigma_{3,1}+\sigma_{3,2})$
Stage 2 permeate to stage 1, $\sigma_{4,1}$	0	1
Stage 2 permeate to stage 2, $\sigma_{4,2}$	0	$1-\sigma_{4,1}$
Stage 2 permeate to permeate product, $\sigma_{4,3}$	0	$1-(\sigma_{4,1}+\sigma_{4,2})$

possible to reduce the number of network decisions by making a few reasonable assumptions: for example in pervaporation, the vapour permeate stream cannot be mixed with the liquid retentate stream. Simplifying the network by preventing permeate and retentate mixing is seen to greatly reduce the number of network choices.

3.1.3. Summary

A superstructure for the optimal design of membrane systems has been described. It provides a neat and efficient framework, yet, is able to encompass all design alternatives. This new methodology enables the number of modules, as well as the driving force for mass transport, to be optimised directly. The structure of the system can also be optimised by manipulating the stream flows leaving each unit.

The process superstructure is fixed by specifying the values of all the optimisation decision variables. To illustrate this, the list of decision variables for a two separation stage system is given in Table 4. The list of decision variables divides into two. The first part is concerned with the stage decision variables. These are the number of modules (discrete), the feed stream pressure or temperature (continuous) and the permeate stream pressure (continuous). Throughout this work, for neatness, the operating state variables (temperature or pressure) are normalised between 0 and 1—where 0 corresponds to the minimum allowed value of the variable, and 1 the maximum. The second part is concerned with the full structural layout of the membrane system and therefore holds the values of the network decision variables (the split fractions).

3.2. The design of membrane systems using genetic algorithms

Based on the problem formulation presented above, an optimisation strategy using genetic algorithms has been implemented in a C++ program. This uses a genome to encode the optimisation decision variables, with a single gene used to represent each decision variable. The program is based on the genetic algorithm library GAlib (version 2.4) which provides a set of genetic algorithm tools.

3.2.1. The genetic algorithm

All of the work in this paper is based on a steady-state genetic algorithm. This algorithm is initiated using a population of randomly generated genomes. It selects a new population of genomes based on the best individuals from the current population and applies the crossover and mutation operators to these genomes (discussed below). The probability of an operator being applied to any genome is determined by the crossover and mutation rates, and these are also discussed later.

Each new population is merged with the existing population and the worst individuals removed to keep the size of the population constant. This method ensures that the best solutions encountered are not lost from one generation to the next. In this case, the algorithm is terminated when the population converges to a single genome.

3.2.2. Genetic algorithm operators

The key characteristics of the genome are the *crossover* and *mutation* methods that the genetic algorithm uses to create the new generations. The crossover operator defines the procedure for generating two children from two parent genomes. In these studies, the operator simply randomly mixes the parent genes. Consequently, the two children are opposing mixtures of their parents' genes.

Mutation is used to introduce new genetic material or to move existing genetic material around the genome. The type of mutation that takes place will depend on the data type: a binary genome is usually mutated by randomly flipping some of the genes; whereas a Gaussian distribution around the current value is often used to mutate a real number gene. In this work, the genome is mutated in one of two ways, as described below.

- *Gaussian mutation*: Each gene can be individually mutated: the value of the gene is changed using a Gaussian distribution around the current value. If the gene can only take discrete values then it is rounded to the nearest feasible value. However, if the gene reaches either of its bounds then it is reset to that bound. It is important that the algorithm explores the boundary possibilities as this

enables units to be either deselected or operated at maximum power.

- *Stage mutation*: The genes for one stage are copied to genes that represent another stage. This enables information to be passed around the genome. So for example, if a certain stage property (e.g. feed temperature) is found to be beneficial for one stage this information can potentially be copied to other stages.

3.2.3. Fitness

The likelihood of selecting a member of the population is based on its fitness. The fitter the genome the more likely it is to be selected. There is no limit on the number of times a certain genome can be selected.

Each time the genetic algorithm makes a call to calculate the fitness of a genome, a simulation is executed. The current values of the optimisation decision variables are passed to the simulator. After the simulation has been executed, the value of the objective function is returned. The fitness is usually a simple (often linear) function of the objective function.

3.2.4. Genetic algorithm parameter values

There are four main parameters that control the performance of the genetic algorithm. These are: (1) the population size, (2) the percentage of the population to replace from one generation to the next, (3) the crossover rate and (4) the mutation rate. The choice of suitable parameter values is crucial to the success of a genetic algorithm, where both high-quality solutions and short computational times are desired. The best values for these parameter are now discussed. For further details refer to Garrard and Fraga (1998).

Population size: The size of the population should be large enough to provide sufficient diversity at the start of the evolution. Unfortunately, "sufficient diversity" is difficult to quantify. However, Lewin, Wang, and Shalev (1998) report that genetic algorithms are insensitive to the population size provided that it is not very small. In this work, a population size of 50 genomes will be used for small one-stage problems. This will be increased to 150 for larger problems.

Replacement percentage and crossover rate: The maximum number of genomes that are replaced in each generation (the replacement policy) and the rate of crossover must be specified. Garrard and Fraga (1998) found good results for mass exchanger synthesis using a 75% replacement policy and a crossover rate of 75%,[2] these values will also be used here.

Mutation rate: The mutation rate is the likelihood that a gene in a newly born individual will mutate. It is an important parameter used to control the rate of introduction of new genetic material into the population. A high value overrides the effect of crossover, but a value set too low forces the genetic algorithm to converge prematurely.

[2] A crossover rate of 75% indicates that 25% of children are clones of their parents (subject to any later mutation).

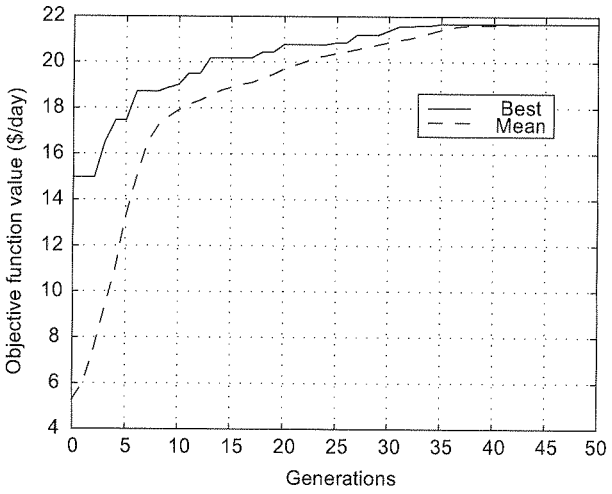

Fig. 3. Objective function profile at a mutation rate of 5%.

Table 5
The effect of mutation rate on genetic algorithm performance for a two-stage pervaporation system

Mutation rate, (%)	Best solution ($/day)	Generations required
0	19.4	37
1	20.8	33
5	21.7	46
15	21.7	37
20	22.1	81
40	21.8[a]	>300

[a]Forty per cent mutation rate run was incomplete and terminated after 300 generations.

The best mutation rate is heavily dependent on the type of problem. A mutation rate of 10% is used by Garrard and Fraga (1998) for mass exchanger network synthesis. However, for heat exchanger networks, Lewin et al. (1998) use a value of 0.1% and Androulakis and Venkatasubramanian (1991) use 30%. Androulakis and Venkatasubramanian (1991) also investigate the use of variable *or generation-dependent* mutation rates.

To determine a suitable value for the type of problem studied in this paper, the effect of mutation rate on both solution quality and computation time has been investigated using a population of two-stage genomes and the pervaporation case study detailed in the following section. Fig. 3 shows the results for a mutation rate of 5%. It is seen that the mean objective function score and the best-of-generation score converge at generation 46. At this point, the population is filled with identical genomes and population convergence. This is used to terminate the genetic algorithm in these studies. Although random mutation may enable further improvement beyond this point, it is highly unlikely.

The mutation study results are presented in Table 5. The table shows the effect of different mutation rates on the best

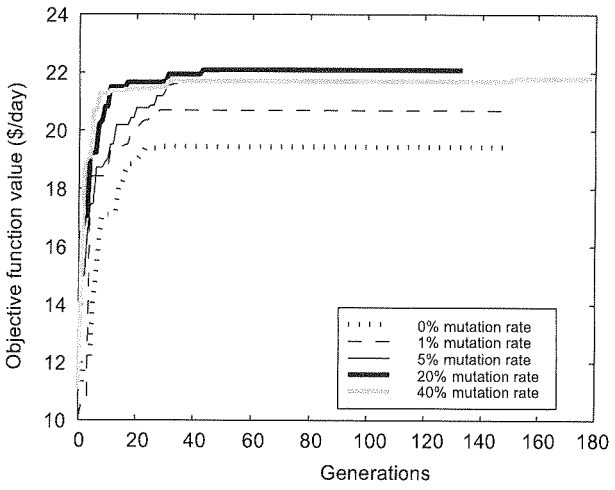

Fig. 4. Objective function profile for different mutation rates.

solution found by the genetic algorithm and the number of generations required for population convergence. It is seen that similar solutions are found when there is even a small amount of mutation. However, a high rate of about 20% is required to find what is believed to be the global optimum (22.1$/day) and prevent premature convergence to sub-optimal values—most apparent for the zero mutation case. Unfortunately, population convergence takes much longer at higher mutation rates, and at a rate of 40% population convergence did not occur within 300 generations. The results are also plotted in Fig. 4 which shows the effect of various mutation rates on population evolution.

Based on these experiences, throughout this work, a mutation rate of 20% is used in conjunction with a crossover rate of 75%; a replacement rate of 75%; and a population size from 50 to 150 (depending on the problem size).

3.2.5. Summary

An optimisation methodology that uses a genetic algorithm to solve a superstructure optimisation problem has now been outlined. The approach is based on the problem methodology described in Section 3.1 and can be applied to any membrane system. In the next part of this paper, the design method will be demonstrated by application to a pervaporation case study.

4. The optimal design of a pervaporation system

Many of the liquid mixtures that are encountered in the speciality chemical industries are difficult to separate as they are either close-boiling or azeotropic in nature. Extractive distillation is a common technique for separating such mixtures, however, it is energy intensive and requires the addition of a solvent to change the relative volatilities of the various components. The solvent is often toxic (e.g. benzene) and must subsequently be separated from the mixture. These drawbacks fuel continuous research into new and better separation techniques. The use of pervaporation as an alternative to distillation for difficult separations has been suggested for a number of cases. In particular, the use of pervaporation to separate azeotropic ethanol water mixtures is becoming increasingly important (Fleming & Slater, 1992, Chapter 7–10).

In pervaporation, the membrane forms a semi-permeable barrier between the liquid feed and a low-pressure gaseous product. Consequently, the heat of evaporation must be supplied to the permeating material—typically resulting in a feed stream temperature drop. However, in contrast to distillation, only the heat of evaporation for a small fraction of the mixture (the permeating material) must be provided. Thus, the energy requirements of a pervaporation plant are much lower.

The modular nature of membrane systems means that pervaporation is unlikely to compete with distillation for large-scale separations as it does not benefit from such great economies of scale. However, pervaporation offers a number of additional advantages (Fleming & Slater, 1992) that make it an attractive option for many separations: lower capital costs for small-scale systems, easier retrofitting and debottlenecking (modular design), and often superior separations as it is not constrained by thermodynamic azeotropes.

Pervaporation is inherently complex as large temperature and concentration changes are common inside the membrane modules. Therefore, the accurate simulation and optimisation of pervaporation processes can be computationally expensive. As a result, optimisation studies (e.g. Srinivas & El-Halwagi, 1993) have relied on approximate design models to describe the modules. For instance, non-isothermal conditions and constant physical and membrane properties have invariably been assumed. However, the use of approximate models for pervaporation is particularly inappropriate due to the high dependence of permeability on temperature which, if ignored, will cause significant inaccuracies. Therefore, in this paper, the application of detailed models to the optimal design of pervaporation systems is advocated. The mathematical model of a membrane module presented by Marriott and Sørensen (2003) will be used. This model is developed from rigorous mass, momentum and energy balances and disregards many common assumptions such as: plug flow; constant pressure; constant temperature; binary mixture; steady-state conditions; and constant physical properties.

4.1. Solution methodology

In pervaporation, in order to optimise the driving force across the membrane, the feed stream temperature and the permeate pressure are manipulated. Thus, in addition to membrane modules, pervaporation processes can contain ancillary equipment such as heaters and compressors. The individual roles of these items are given in Table 6.

Table 6
Ancillary equipment for pervaporation processes

Equipment	Use
Heaters	To provide the heat of evaporation to the liquid stream
Condenser	1. To condense permeate vapour 2. To control downstream permeate vacuum pressure
Booster compressors	To further reduce downstream vacuum pressure
Liquid pumps	To pump feed and recycle flows around the system

4.1.1. Separation stage decisions

A general membrane separation stage was illustrated earlier in Fig. 2. For a pervaporation stage, the number of modules, the feed temperature, and the permeate pressure for that stage must be determined.

The permeate stream pressure is usually maintained well below atmospheric conditions using a condenser (the condenser pressure is fixed in this study and hence not considered as part of the design problem). In some cases, however, booster compressors are used to further reduce the downstream pressure (e.g. Tsuyumoto, Teramoto, & Meares, 1997) and these will be considered here. Therefore, the pressure of each permeate stream is an optimisation decision variable. To enable stages to share a compressor, the permeate pressures will be selected from an *allele* set (this will be of size N^s). In which, the values of the pressures are also optimisation decision variables.

4.1.2. Network decisions

The structural layout of the plant is determined by fixing the values of the network decision variables. In pervaporation, each stage has an upstream (liquid) input and an upstream output as well as a downstream (vapour permeate) output. Therefore, the two output streams cannot be mixed. As discussed earlier, this is a significant advantage as it greatly reduces the number of network choices. In this study, we will further simplify the system by only allowing separation stages to be connected sequentially—this is reasonable as modules can be connected in parallel inside each stage.

The superstructure consists of N^s separation stages, and is generated by connecting the output from one stage to the input of the next. Consequently, the only network decision variable is the fraction of retentate product that is recycled at each stage. The simplified process superstructure for a two-stage pervaporation system is illustrated in Fig. 5.

4.1.3. Solution strategy

The best plant design will be determined by the solution of the superstructure optimisation problem. To select a design from the superstructure, the number of modules, heater temperature, recycle fraction, and the choice of permeate pressures must be specified for each separation stage. The permeate pressures in the allele set must also be determined.

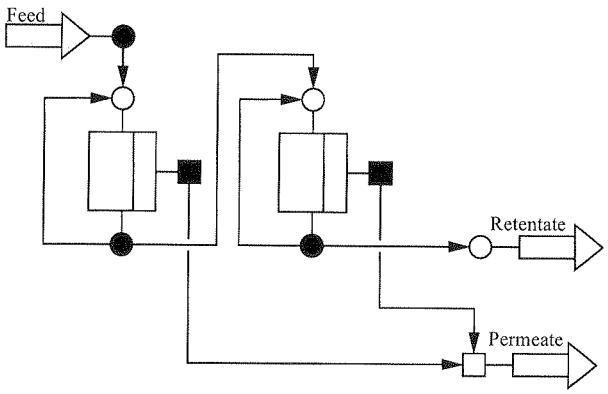

Fig. 5. A superstructure for a pervaporation process (two stages).

Table 7
Decision variables for a pervaporation system

Decision variable	Lower bound	Upper bound	Interval
The number of membrane modules, N^m	1	$n^{\max}$	1 (integer)
Normalised feed stream temperature, D^T	0	1	0.01
Compressor pressure choice, $D^{P'}$	0	N^s	1 (integer)
Normalised compressor pressure(s), D^P	0	1	0.0025
Stage recycle fraction, $\sigma_{a,a}$	0	0.75	0.05

This requires a total of five decision variables per stage as indicated in Table 7.

To enable the algorithm to distinguish between different designs and to help convergence to a distinct solution the continuous decision variables have been discretised.[3] The discretisation intervals for each variable are also shown in Table 7.

4.2. Description of the case study

The design and performance of a pervaporation pilot plant for the dehydration of azeotropic ethanol/water mixtures has been reported by Tsuyumoto et al. (1997). The plant, which is shown in Fig. 6, is based on nine hollow-fibre modules in seven separation stages, four heaters and two vacuum compressors. Models which describe the ancillary equipment have been developed and are given in the appendix. In this work, the optimal design of this pervaporation process will be determined to demonstrate the capabilities of the optimal design strategy proposed in this paper.

4.2.1. Mathematical model

Before consideration is given to the optimal design of the ethanol dehydration system, it is important to determine the

[3] It is worth pointing out that discretisation of the continuous decision variables is not a general requirement of the formulation proposed in this paper, but can be useful in helping the user distinguish between different designs.

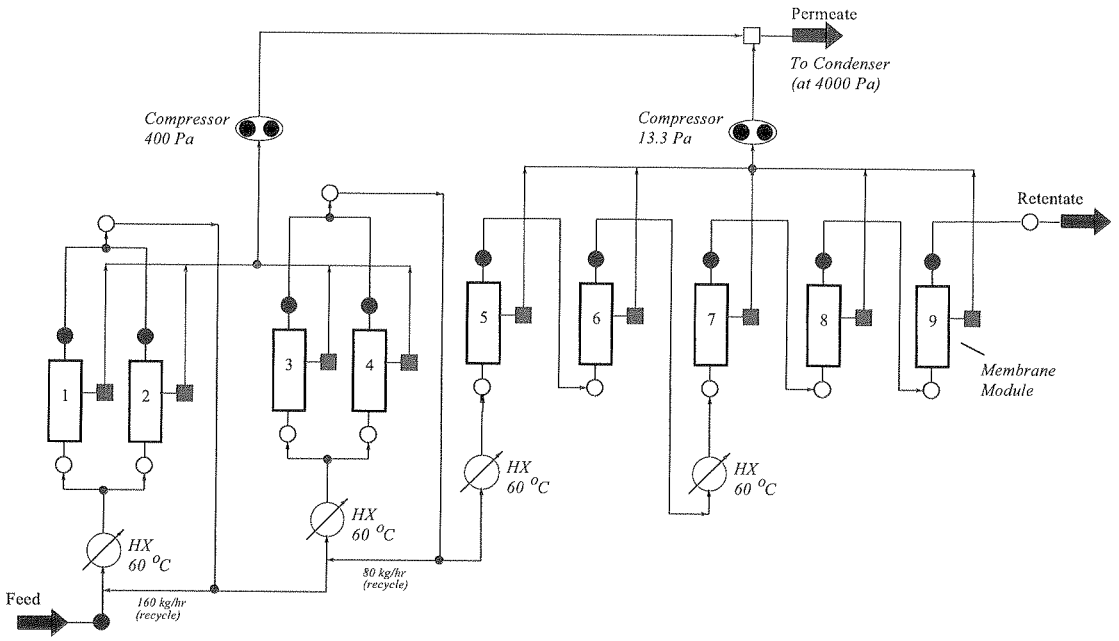

Fig. 6. Pervaporation separation process, as proposed by Tsuyumoto et al. (1997).

accuracy of the model used to describe the system. In earlier work (see Marriott & Sørensen, 2003), we introduced a detailed model of a membrane module. The model was developed from rigorous mass, momentum and energy balances enabling it to predict the spatial variation in fluid properties along the length of the membrane module. Critically for describing pervaporation, the model rigorously accounts for non-isothermal flow and concentration polarisation. The model was shown to provide excellent agreement with experimental data for the hollow-fibre modules described by Tsuyumoto et al. (1997). The model is solved using the gPROMS simulation software (Process Systems Enterprise Ltd, 2000) with physical properties provided by the *Multiflash* software package (Infochem Computer Services Ltd, 2000).

4.2.2. *Optimisation objective and constraints*

Economic criteria will be used to determine the optimal plant design. The objective function is, therefore, to minimise production costs (C) subject to the same constraints as those used for the Tsuyumoto et al. (1997) design (these are given in Table 8)

$$\min[Cf(\omega_r)g(m_r)]. \tag{6}$$

To ensure that the optimal solution meets the required production quality, it is necessary to penalise solutions that do not satisfy the product constraints. This is done using the penalty functions $f(\omega_r)$ and $g(m_r)$.

$f(\omega_r)$ is given by

if $\omega_r \geqslant \omega_r^{\min}$

$$f(\omega_r) = 1, \tag{7}$$

Table 8
Production and design constraints for the ethanol dehydration plant (Tsuyumoto et al., 1997)

Constraint	Symbol	Value
Feed rate (kg/hr)	m_f	100
Feed purity (wt% ethanol)	ω_f	94
Production rate (kg/h)	$m_r^{\min}$	94
Product purity (wt% ethanol)	$\omega_r^{\min}$	99.7
Maximum heater temperature (°C)	$T^{\max}$	70
Minimum permeate pressure (Pa)	$p^{\min}$	10
Permeate outlet pressure (Pa)	$p^{\max}$	4000

else

$$f(\omega_r) = \left(1 - \frac{\omega_r^{\min} - \omega_r}{\omega_r^{\min} - \omega_f}\right)^{-4} \tag{8}$$

and $g(m_r)$ is written as

if $m_r \geqslant m_r^{\min}$

$$g(m_r) = 1, \tag{9}$$

else

$$g(m_r) = \left(1 - \frac{m_r^{\min} - m_r}{m_r^{\min}}\right)^{-4}. \tag{10}$$

4.2.3. *Costing*

The estimated annualised capital charges for each unit and the utility costs (from Tsuyumoto et al., 1997) are given in Table 9. Unfortunately, due to the small scale of the separation system considered in this study (100 kg/h),

Table 9
Economic criteria for the ethanol dehydration plant

Annualised fixed cost of membrane modules ($/yr)	56
Annualised fixed cost of heaters ($/yr)	250
Annualised fixed cost of compressors ($/yr)	150
Steam cost ($/J)	1.316×10^{-3}
Electricity cost ($/J)	2.66×10^{-3}
Mechanical efficiency of compressors	0.75
Operational hours per year	8000

The annualised costs include replacement costs, labour and maintenance charges. The steam and electricity costs are taken from Tsuyumoto et al. (1997).

accurate capital cost algorithms are unavailable. Therefore, nominal size-independent capital (fixed) costs are used (Table 9).

4.2.4. Recycle flows

Theoretically, recycle flows can easily be included as part of the optimisation problem. Previous studies (Tsuyumoto et al., 1997) have shown that recycling some of the outlet flowrate back to the module inlet can improve product quality. However, for this system, only a marginal increase in product quality was found to be possible and this required a greater amount of heat to be supplied.

As size-independent capital costing is used for the heater in this study (due to the small scale of the separation system), introducing recycle flows will enable a much higher heat input without increasing equipment costs. The result of a structural optimisation would be an unrealistic plant design with very large recycle flows and a few heaters supplying all the heat. Therefore, recycle flows will be neglected for this optimisation study (i.e. $\sigma_{a,a} = 0$).

4.3. Optimisation results

The annualised cost of the ethanol dehydration system proposed by Tsuyumoto et al. (1997) is calculated as $6787 per year using the economic data given in Table 9. The optimal design strategy proposed in this paper is now used to determine the optimal design of the same system. The design strategy is also assessed in terms of computational requirements and then by comparison with an MINLP solution technique based on a branch and bound method (see Marriott, Sørensen, & Bogle, 1999).

4.3.1. Number of stages

A separation stage is defined as a number of membrane modules connected in parallel. The bounds on the number of modules for each separation stage must be fixed: for this case study a separation stage is specified as containing between 1 and 25 modules (i.e. $n^{\min} = 1$, $n^{\max} = 25$). The optimisation problem is solved for four superstructure sizes: containing from one to four separation stages. The summarised results for each case are presented in Table 10.

Initially, a superstructure for a single separation stage is generated (i.e. all the modules will be connected in parallel). The solution technique described earlier is then used to solve the optimisation problem. The best design (Table 10) found by the algorithm has an annualised cost of $6447 per year, this compares favourably with $6787 per year for the Tsuyumoto et al. (1997) system due to significantly reduced heating requirements.

To determine the effect of the number of separation stages, the optimisation is repeated for superstructures built from two, three and four stages (refer to Table 10). An improved annualised cost of $5970 per year is found by increasing

Table 10
Optimal pervaporation plant designs for different numbers of separation stages

		Number of separation stages			
Stage	Number of modules	One	Two	Three	Four
		16	2	1	1
1	Heater (°C)	70	70	70	70
	Permeate pressure (Pa)	60	630	1260	1300
		—	6	1	1
2	Heater (°C)	—	70	70	70
	Permeate pressure (Pa)	—	60	1260	1300
		—	—	5	1
3	Heater (°C)	—	—	70	None
	Permeate pressure (Pa)	—	—	120	90
		—	—	—	3
4	Heater (°C)	—	—	—	70
	Permeate pressure (Pa)	—	—	—	90
Number of compressors		1	2	2	2
Annualised cost ($/yr)		6447	5970	5863	5873

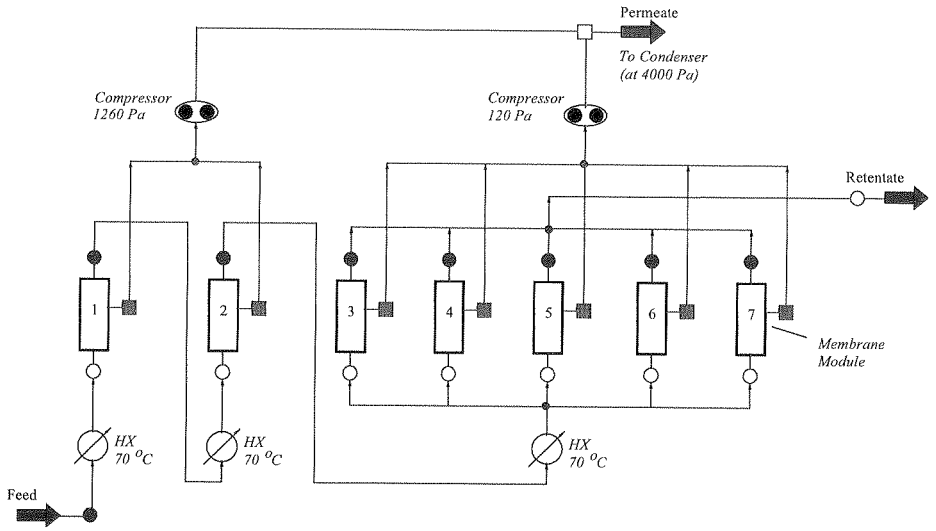

Fig. 7. Optimal plant design identified using the genetic algorithm.

the number of separation stages to two. A further slight improvement can be achieved by enabling three separation stages, \$5863 per year. However, forcing the algorithm to choose a design with four stages results in slightly increased costs, \$5873 per year, indicating that the optimum solution is a three-stage separation plant. Further details of all the designs are given in Table 10.

4.3.2. Design comparison

The new design based on three stages is illustrated in Fig. 7. The main emphasis of the design is to keep the temperature high throughout the process by adding heat in the initial stages, where most of it is otherwise lost. The higher initial temperatures mean that less membrane area is necessary for the first two stages and that lower compressor duties are required than in the Tsuyumoto et al. (1997) design (Fig. 6). Furthermore, additional heaters are not required at the end of the process.

Summarised cost comparisons are given in Fig. 8. This shows that capital, membrane and utility costs are all lower for the optimised design with a total saving of 13.5%. The new design contains less modules and heaters leading to the reduction in capital costs. Operating costs are lower due to a similar total heat input but lower compressor duties. In fact, it is interesting to note that as all aspects of the design are cheaper then whatever costing algorithm is used, this design would provide a cost saving over the base design.

A disadvantage of the new design is its lack of flexibility—there is little scope to increase the purity of the product if required later, without additional capital investment. This is because the heaters are already operating at the maximum process temperature. To overcome this problem, it is relatively straightforward to initially over-design

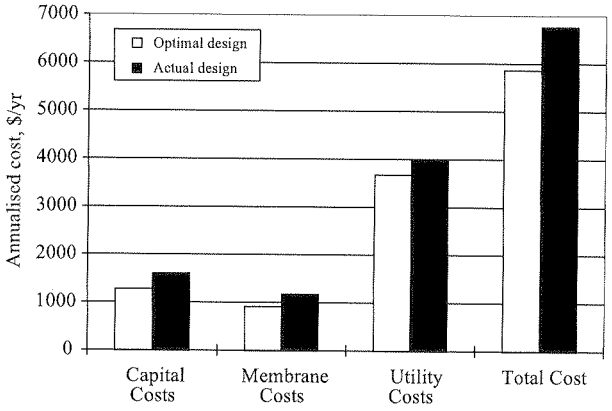

Fig. 8. Cost comparison between the optimal and Tsuyumoto et al. (1997) designs.

the plant by setting tighter product specifications than required. However, that is beyond the scope of this work.

4.4. Computational requirements

All of the calculations reported in this work were performed on an IBM RISC System/6000 workstation running under the AIX 4.3.2 operating system.

The optimisation results are summarised in Table 11. This demonstrates that the fraction of candidate solutions evaluated by the genetic algorithm is tiny—for the three-stage case, less than 5000 evaluations were required out of more than 3.5×10^{16} possibilities. Yet the genetic algorithm works remarkably well: the optimal solution is seen to offer a significant improvement over the actual plant design.

Typical gPROMS simulation (genome evaluation) times ranged from 2 CPUs for a single-stage to 10 CPUs for

Table 11
Summarised optimisation results for the pervaporation case study

Number of stages	Best solution ($/yr)	Total number of simulations	Total time (h)
One	6447	1200	0.7
Two	5970	3100	4.2
Three	5863	5000	9.6
Four	5873	13500	36.8
Tsuyumoto et al. (1997) design	6787		

a four-stage plant. The number of simulations required to determine the solution (Table 11) is seen to be relatively low and does not inflate exponentially as the problem size is increased. Therefore, optimisation times range from 1 h for the small one-stage problem, to just under 40 h for the largest problem.

It is possible to reduce solution times by using less decision variables and an approximate membrane model. Nevertheless, it is felt that restricting the range of decision variables would reduce the solution space and hide the true potential of pervaporation for liquid mixture separation. The use of an approximate design model cannot be justified in this case due to the highly non-isothermal system.

4.5. Solution using a NLP solver

A number of methods for the solution of MINLP optimisation problems were identified earlier in this paper. In order that the performance of the genetic algorithm can be assessed, the optimisation has been repeated using an MINLP solution technique based on a manual branch and bound method. Further details of this optimisation were presented in an earlier paper (Marriott et al., 1999).

With this method, approximately 40 NLP sub-problems were solved using the gOPT solver which is incorporated in the gPROMS software (Process Systems Enterprise Ltd, 2000). Each NLP optimisation took approximately 20 min; however, a guarantee of global optimality was not possible so each optimisation was repeated from two or three different initial conditions. Consequently, over 100 NLP optimisations were performed, taking a total of 35 h of computational time.

The solution found using this method ($5927 p.a.), is similar (although, not quite as good) to the best design found by the genetic algorithm—in a comparable computation time. However, it should be noted that the designs cannot be compared directly as a simpler module model was used in the MINLP case (Marriott et al., 1999). Nevertheless, the study was able to highlight two shortcomings of this solution technique.

- Only a single best solution was found: a significant advantage of genetic algorithms is that multiple solutions are available following the optimisation. This feature will be explored further in a future paper.
- It is difficult to automate the manual branch and bound technique, due to the subjective need to repeat each optimisation until the global optimum is found. This is usually necessary in order to prevent the search space being *pruned* prematurely and the best solutions being cut off. This problem is inherent to all current NLP solvers where a guarantee of global optimality is not possible.

Whilst potentially faster solution strategies based on NLP solvers exist (e.g. outer approximation methods), these suffer from the same disadvantages that were highlighted above: namely that only a single best solution is reported, convergence to local optima and, because of this, implementation is difficult. In contrast, genetic algorithms are not subject to such concerns and in many cases (such as those explored in this paper) can find equally good, if not better solutions, without significantly increasing computational expense.

5. Conclusions

A new optimal design methodology for membrane systems has been presented in this paper. The design methodology focuses directly on the balance between the number of membrane units and the driving forces for mass transport through the membranes. However, the structure of the system can also be optimised by manipulating the stream flows leaving each unit. In this approach, a genetic algorithm is used to solve a superstructure optimisation problem.

Using this strategy, a pervaporation system to concentrate 100 kg/h of 94 wt% ethanol to a purity in excess of 99.7 wt% ethanol has been designed. Reduced operating and capital charges of 13.5% are seen in comparison to the actual plant (Tsuyumoto et al., 1997). These substantially lower costs are a result of smaller equipment duties and a modified configuration with two fewer modules and one fewer heater.

Unlike previous optimisation studies found in the literature, this design was produced using accurate membrane unit models that have been assessed against experimental data for a range of different conditions. The unit models do not make assumptions as to the nature of the process and fully account for interacting decisions. The use of accurate models not only ensures that any proposed designs will meet product specifications but that similar designs can be compared fairly. The use of a rigorous model is essential for this case study as this process is characterised by large temperature changes which cannot accurately be described by a simple design model.

It has also been shown that genetic algorithms are an excellent basis for an effective and powerful tool for the optimal design of membrane systems. The computational

requirements of the method were relatively large but necessary, if all the degrees of freedom in the design are to be explored. The method compared favourably when contrasted to more conventional MINLP optimisation solution methods.

Notation

a	unit outlet number, dimensionless
b	unit inlet number, dimensionless
C^{Total}	total production cost, \$/yr
C^{fixed}	fixed unit cost, \$/yr
C^{op}	operating cost, \$/yr
C^{elec}	electricity cost, \$/J
C^{steam}	steam cost, \$/J
C_p	heat capacity, J/mol K
D^S	normalised state decision variable, dimensionless
D^T	normalised temperature, dimensionless
D^P	normalised pressure, dimensionless
$D^{P\prime}$	pressure selector variable, dimensionless
H	enthalpy, J/mol
m	mass feed rate, kg/s
m_a^{out}	total mass flow rate of material leaving unit a, kg/s
$m_{a,b}^{\text{in}}$	mass of material entering unit b from unit a, kg/s
m^r	mass flowrate of retentate product, kg/s
$n^{\max}$	maximum number of membrane modules per separation stage, dimensionless
$n^{\min}$	minimum number of membrane modules per separation stage, dimensionless
N^c	number of components, dimensionless
N^f	number of feed points, dimensionless
N^i	number of inlets, dimensionless
N^m	number of membrane modules per separation stage, dimensionless
N^p	number of product points, dimensionless
N^s	number of separation stages, dimensionless
P	pressure, Pa
q'	heater duty, W
S	fluid state (temperature or pressure), K or Pa
T	temperature, K
w	work, W

Greek letters

η	efficiency, dimensionless
v	volumetric flowrate, m^3/s
$\sigma_{a,b}$	stream split fraction from unit a outlet to unit b inlet, dimensionless
ω	weight fraction, dimensionless
ω_p	weight fraction of key component in permeate, dimensionless
ω_r	weight fraction of key component in retentate, dimensionless

Superscripts

in	value at system or unit inlet
max	maximum value
min	minimum value
out	value at system or unit outlet

Appendix.

Ancillary equipment models: In addition to the detailed mathematical model of a membrane unit presented by Marriott and Sørensen (2003), a number of other models were used in the optimal design case study investigated in this paper. Essentially, these models determine the energy and utility requirements of the membrane system.

Liquid pump: The power requirement is written

$$-w = \frac{v}{\eta}(P^{\text{out}} - P^{\text{in}}), \tag{A.1}$$

the operating cost is

$$C^{\text{op}} = -wC^{\text{elec}}. \tag{A.2}$$

Heater: The heat duty is defined

$$q' = m(H^{\text{in}} - H^{\text{out}}) \tag{A.3}$$

and the operating cost is

$$C^{\text{op}} = q'C^{\text{steam}}. \tag{A.4}$$

Compressor: The compressor work is calculated assuming isothermal compression

$$-w = \frac{v}{\eta}P^{\text{in}}\ln\left(\frac{P^{\text{out}}}{P^{\text{in}}}\right) \tag{A.5}$$

and again the operating cost is

$$C^{\text{op}} = -wC^{\text{elec}}. \tag{A.6}$$

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