THESIS APPROVAL

GRADUATE SCHOOL, KASETSART UNIVERSITY

Master of Engineering (Chemical Engineering)

DEGREE

Chemical Engineering	Chemical Engineering
FIELD	**DEPARTMENT**

TITLE: Design of Control Structure for Mass Exchanger Networks

NAME: Mr. Pattarasak Sattarattanakul

THIS THESIS HAS BEEN ACCEPTED BY

THESIS ADVISOR

(Associate Professor Thongchai Srinophakun, Ph.D.)

THESIS CO-ADVISOR

(Mr. Chanin Panjapornpon, Ph.D.)

DEPARTMENT HEAD

(Associate Professor Phungphai Phanawadee, D.Sc.)

APPROVED BY THE GRADUATE SCHOOL ON

DEAN

(Associate Professor Gunjana Theeragool, D.Agr.)

THESIS

DESIGN OF CONTROL STRUCTURE FOR MASS EXCHANGER NETWORKS

PATTARASAK SATTARATTANAKUL

A Thesis Submitted in Partial Fulfillment of
the Requirements for the Degree of
Master of Engineering (Chemical Engineering)
Graduate School, Kasetsart University
2009

Pattarasak Sattarattanakul 2009: Design of Control Structure for Mass Exchanger Networks. Master of Engineering (Chemical Engineering), Major Field: Chemical Engineering, Department of Chemical Engineering. Thesis Advisor: Associate Professor Thongchai Srinophakun, Ph.D. 94 pages.

This thesis aims to present a design of optimal operation for mass exchanger networks under the minimum total annual cost (TAC) and the optimal split-range control. The proposed strategy is consisted of four main steps. First, mass exchanger networks is formulated as a mixed integer nonlinear programming (MINLP) with the minimized total annual cost. Second, performing flexibility test to ensure the network is feasible for a large number of uncertain parameters that are generated randomly within the operating ranges. Next, the parametric programming is used to find the active constraint regions and the integer linear programming (ILP) is used to formulate an optimal split-range control structure. Finally, the optimal operations of control structure are verified with the proposed dynamic behaviors. The validation of the proposed method of design an optimal split-range control structure for mass exchanger networks is performed by three case studies; El-Halwagi and Manousiouthakis (1989), (1990a) and (1990b). The result of all case study are not guarantee the global optimization but all cases are checked through flexibility testing, dynamics testing and they demonstrate that the obtained control structures can keep all target mass fractions at the desired values even under the saturation of some manipulated variables when system of MENs are disturbed by reducing 10, 20 and 30% mass fraction of rich streams and lean streams.

_________________________ _________________________ ___ / ___ / ___

Student's signature Thesis Advisor's signature

ACKNOWLEDGEMENTS

There are many people giving contribution either directly or indirectly to this research. First, I would like to express my sincere thank to Assoc. Prof. Thongchai Srinophakun, the research advisor, for this good research setup. He always shared his invaluable ideas, suggestions, and recommendations, especially in slide reviewing and report proof-reading. Next, I would like to thank Assoc. Prof. Thumrongrut Mungcharoen and Dr. Chanin Panjapornpon, my thesis committees, for their comments and suggestions that are very useful for my research work. I would like to acknowledge Dr. Veerayut Lersbamrungsuk for helping and suggestions. This thesis was supported by National Center of Excellence for Petroleum, Petrochemicals and Advanced Materials, S&T Postgraduate Education and Research Development Office (PERDO).

Very big thank to my friends in Kasetsart and CPCO team for their encouragements and helps, especially Khun Wipawan Noicham. Thank you for all your help. Finally, I would like to thank my family who always supported me throughout this research.

Pattarasak Sattarattanakul

February 2009

TABLE OF CONTENTS

LIST OF TABLES

LIST OF TABLES (CONTINUED)

LIST OF FIGURES

LIST OF FIGURES (CONTINUED)

Appendix Figures **Page**

LIST OF ABBREVIATIONS

CV	=	controlled variable
DOF(s)	=	degree(s) of freedom
MEN(s)	=	mass exchanger network(s)
ILP	=	integer linear programming
LMTD	=	logarithmic mean temperature difference
NLP	=	nonlinear programming
MINLP	=	mixed-integer linear programming
MPT	=	multiparametric toolbox
MV	=	manipulated variable
NLP	=	nonlinear programming
RI	=	resilience index
TAC	=	total annual cost
A515	=	Carbon Steel Plates for pressure vessels for intermediate and higher temperature service)

Indices

i	=	rich process stream
j	=	lean process stream or mass separation agent (MSA)
k	=	index for stage, 1, ..., NS, and composition location, 1, ..., NS +1

Sets

RP	=	$\{ i \mid i$ is a rich process stream, $i = 1, ..., NR \}$
LP	=	$\{ j \mid j$ is a lean process stream or MSA, $j = 1, ..., NL \}$
ST	=	$\{ k \mid k$ is a stage, $k = 1, ..., NS \}$

LIST OF ABBREVIATIONS (Continued)

Parameters

AC_j	=	annual operating cost of lean stream j
ACt	=	per stage annual cost of tray column
ACh	=	per height annual cost of packed column for i rich and j lean match
b_{ij}	=	intercept of equilibrium line in i rich and j lean match
G_i	=	flow rate of rich stream i
K_ya	=	overall mass transfer coefficient
$L^{(up)}{}_j$	=	upper bound on mass flow rate of lean stream j
m_{ij}	=	slope of equilibrium line in i rich and j lean match
S	=	cross-sectional area of an exchange unit
U	=	upper bound for mass exchange
Γ	=	upper bound for composition difference
$X^{(in)}{}_j$	=	inlet composition of lean stream j
$X^{(out)}{}_j$	=	outlet composition of lean stream j
$X^{s}{}_j$	=	supply compositions for each MSA
$X^{t}{}_j$	=	target compositions for each MSA
$Y^{(in)}{}_i$	=	inlet composition of rich stream i
$Y^{(out)}{}_i$	=	outlet composition of rich stream i
$Y^{s}{}_i$	=	supply compositions for each waste stream.
$Y^{t}{}_i$	=	outlet composition for each waste stream.
ε_{ij}	=	minimum composition difference between rich stream i and lean stream j

Variables

$dyxi^{p}_{ijk}$ = composition approach in the lean end of the mass exchanger between rich i and lean j in stage k

LIST OF ABBREVIATIONS (Continued)

$dyxo^{p}_{ijk}$ = composition approach in the rich end of the mass exchanger between rich i and lean j in stage k

N_{ijk} = number of trays in the tray column (i, j, k)

H_{ijk} = height of in the packed column (i, j, k)

HTU = height of an overall rich-phase transfer unit

NTU = number of overall rich-phase transfer units

gg_{ijk} = flow rate of rich i that is connected to lean j in stage k

ll_{ijk} = flow rate of lean j that is connected to rich i in stage k

L_j = flow rate of lean process stream j

M_{ijk} = mass exchanged between rich stream i and lean stream j in stage k

x_{jk} = composition for lean stream j in rich end of stage k

y_{ik} = composition for rich stream i in rich end of stage k

sx_{ijk} = composition for the part of lean stream j that is connected to rich i in the rich end of an exchanger in stage k

sy_{ijk} = composition for the part of rich stream i that is connected to lean j in the lean end of an exchanger in stage k

z_{ijk} = 1 denoting existence of match (i, j) in stage k; otherwise = 0

Binary variables

$x_{i,j}$ = existence of relationship between manipulated variable i and manipulated variable j

$z_{k,j}$ = existence of control pair between controlled variable k and manipulated variable j

DESIGN OF CONTROL STRUCTURE FOR MASS EXCHANGER NETWORKS

INTRODUCTION

Chemical processes commonly consist of many unit operations to achieve the target of the production. In the past, the typical approach is the design of control system for each individual unit. The chemical plants tend to be higher integrated and interconnected because of the results of strict environmental regulation and economic consideration. However the steady-state and dynamic behaviors of these interconnected units are significantly difference from each the individual units. Accordingly, the problem of plantwide control becomes the operation and control of these interconnected process units. Novel chemical processes have to face the problem of steady-state and dynamic interaction and, design engineers have to eliminate many surge tanks, increase recycle streams and introduce heat integration for both existing and new plants. Hence, the attraction in the study of dynamics in chemical plants with recycles and plantwide control design has increased significantly in the last four decades. Moreover, to achieve the best performance including cost effectiveness, the yield enhancement, energy efficiency, and pollution prevention of current processes in the chemical plants, those processes must be developed. Because the chemical process is an integrated system of interconnected units and streams, the concept of process integration that refers to process design, operation, and retrofitting is a powerful instrument in developing the current processes. The popular technique to integrate the process is the network synthesis e.g. Mass Exchanger Networks (MENs), Heat Exchanger Networks (HENs), and Combined Heat and Reactive Mass Exchange Networks (CHARMENs).

The chemical industry is challenged currently by the conflict between the growing amount of industrial waste, ever-tighter governmental emissions targets and cleaner processes. Many countries have made restrictive laws on pollution prevention, which prompted industry to seek various approaches to reduce environmental impact.

How to prevent pollution has become one of the most serious problems to be faced. To a great pollution from industrial processes is a direct result of inefficiencies in raw material and energy utilization. Improvements in process efficiency should, therefore, lead to a reduction in pollution. Now, the leading methods in the pollution prevention hierarchy are source reduction, recycling and reuse. Reuse systems are employed to recover the pollutants for reuse in the plant or for sale as feedstock for other chemical processes. The considerations about the practical situations require engineers to pay more attention to the modification of existing processes in order to achieve the cost-effective target and reduce the passive environmental impact of chemical processes by using conception of synthesizing mass exchange networks (MENs). The mass exchanger can be defined as direct-contact unit and counter-current unit that employ a mass-separating agent (MSA) to remove the certain constituent from a rich stream. The certain components generally represent the impurities in any process streams, pollutants, by products, or even valuable products themselves. Examples of mass exchangers are absorption, adsorption, extraction, ion exchange, leaching, and stripping. A mass exchanger is depicted in Figure 1.

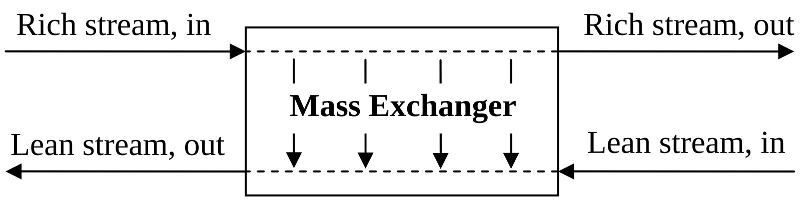


Figure 1 A mass exchanger

In some situations, there are a lot of mass exchangers, several rich streams which certain species have to be removed and many mass separating agents that can be used for removing the targeted species. Therefore, to obtain the best separation system from these various options, Mass Exchange Networks (MENs) would be considered. A Mass Exchange Networks is a network of one or more mass exchangers with the purpose of transferring a certain component or components from a set of rich stream to a set of lean streams. The schematic of MENs can be illustrated as shown in Figure 2

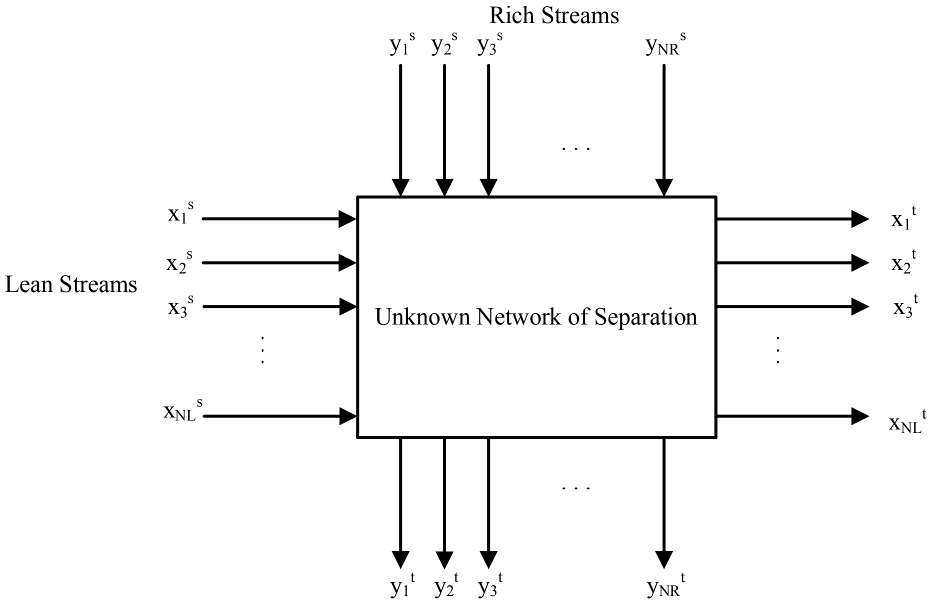


Figure 2 Mass integration schematic with source and target concentrations of rich and lean streams

Moreover, the changes of operating conditions, the optimal vertices could be affected and shifted. So the procedure based on split-range control technique is proposed in this work. It is a simple technique to find constraint of manipulated variables offered by Marlin (2000). Manipulated variables in split-range control are divided into two variables and they are used to adjust one controlled output. When primary manipulated variable is saturated, the secondary manipulated variable will take over task of the saturated one. However, the objective here is focusing on the application of split-range control to implement optimal operation. Glemmestad (1996) and Giovanini (2003) pointed out that in most cases optimal operation of HENs can be implemented using split-range control. A systematic procedure for finding an optimal split-range control structure of HENs can be found in Lersbamrungsuk *et al.* (2008).

OBJECTIVES

1. To design a control system for processes with the of mass exchanger networks by using split-range control.

2. To develop the systematic approach for designing the control system of mass exchanger networks.

3. To determine basic equipment design data and construction cost for mass exchanger networks.

Scope of work

1. Complex Mass Exchanger Networks that consist of many mass exchanger units, are focused in this work.

2. Aspen Dynamics version 2006.5 is used for process simulation.

3. GAMS (General Algebraic Modeling Systems) is used for solving the optimization problem.

4. Aspen Icarus version 2006.5 is used for determining basic equipment design data and construction cost for mass exchanger networks.

5. Using multi-parametric toolbox to solve set of active constraints for additional
information by using integer linear programming (ILP).

6. The network synthesis is targeted on the minimum total annual cost.

LITERATURE REVIEW

1. Multi-period Mass Exchanger Network design

This section describes the reviews of the synthesis of MINLP problem. By talking about the complexity of the separation process into account, the large number of equipment and flows and the highly nonlinear equations are involved. The general mathematical formulation generates a large and complex MINLP model, where even feasible solutions are difficult to find.

El-Halwagi and Manousiouthakis (1989) first introduced the procedure for optimal synthesis of Mass Exchanger Networks (MENs). They first applied pinch analysis to MENs and considered the problem of transferring was certain species from a set of rich process streams to a set of lean streams. A two-stage procedure was defined to find cost of MENs. In the first stage, a thermodynamic procedure was used to identify the thermodynamic bottle neck (pinch points), which limits the extent of mass exchanged between rich and lean process streams. A minimum allowable composition difference (ε) between rich and lean streams is introduced. Their work resulted in targets for the minimum amount of MSA, also referred to as utilities. The second stage was a development of final configuration of the MENs that satisfied the assigned exchange duty at minimum venture cost. However, their methods can be only applied to a single key component, and the final network was not derived through a systematic procedure.

In their later work, El-Halwagi and Manousiouthakis (1990) presented a two stage automated procedure for synthesizing the MENs. Linear programming (LP) is used in the first stage to determine the pinch point as well as the minimum cost of MSAs. A mixed integer linear programming (MILP) transshipment was then used to minimize the number of mass exchangers in the second stage. The completed networks were then cost and the one featuring the lowest cost was selected. This was carried out iteratively for range of ε values in an attempt to minimize the annualized total cost of network. The main weak point of this procedure was its sequential approach. Capital and operating costs are not considered simultaneously. This means that the correct trade off between these costs is unlikely to be found. Another limitation of this procedure is that it apparently considers only networks featuring the minimum number of units. These networks do not necessarily achieve the minimum capital cost. Also, many network designs must be completed and evaluated. The procedure is computationally intensive

Papalexandri *et al.* (1994) later developed a procedure based on multi-period mixed integer nonlinear programming (MINLP) to overcome the limitation of the sequential procedure in the linear programming. In this MINLP approach, they generated a network hyperstructure containing many network alternatives to present the MENs. Optimization was done based on this hyperstructure in order to get a minimum total annual cost (TAC). However, besides the great amount of efforts required to set up and optimize the network hyperstructure, this method does not alway guarantee the generation of an optimum network. This is due to the fact that this hyperstructure does not take into account the thermodynamic bottleneck of the networks.

Next, Wang and Smith (1994) addressed the case of MENs with single lean stream (water) for the minimization of wastewater in the process industries. To design the optimum MENs that achieve minimum wastewater flowrate, two design methods are presented. The first design procedure maximizes driving forces in individual operations whilst the second minimizes the number of water sources for each operation. Both single and multiple contaminants are addressed in this paper. However, capital cost targets did not exist for all MENs cases. Later, they addressed wastewater minimization with flow rate constraints. Local recycling or splitting of operations is used as alternative ways to meet those flow rate constraints.

Krirkkraikijporn and Srinophakun (2002) introduced the concept of simultaneous heat and Mass exchanger networks technique for multi-components to eliminate unsatisfied substances out of process streams through pinch analysis. The temperature of rich streams is exchanged with MSA in order to reach the equilibrium temperature where maximum mass exchange occurs. The optimization program called GAMs was used to solve the MINLP optimization problem based on the minimum operating cost. A user interface program was developed to link with GAMs. When the equilibrium temperature was considered, the mass between rich and lean stream more exchanged. Additionally, the more use the equilibrium equations between rich and lean streams, the more reliable the result because the heat and mass balance are guaranteed

In the previously mentioned works on the synthesis of optimal MEN, they always emphasize only in the continuous process. Foo *et al.* (2004), hence, introduce the synthesis of MEN for batch process systems. Two utility-targeting approaches are presented for this synthesis i.e. vertical and horizontal cascading. In the vertical cascading technique, they intend to minimize utility targets by maximizing the reuse of the process MSA in each time interval. In contrast, the horizontal cascading method try to maximize the use of the available driving force from the process MSA before the use of any external MSA. Three cases studied are performed i.e. the single batch process with and without mass storage, and repeated batch processes with storage system. Single batch process without storage consumes the highest external MSA while the repeated batch processes with storage maximizes the use of the process MSA. In addition, the utility consumption for a repeated batch processes with storage is the same as that for the continuous process.

The following work of Foo *et al.* (2005), in the MEN for a batch process, describes the methodology for setting the minimum number of mass exchange units and a procedure for designing a maximum mass recovery network correspond to the minimize utility targets. Two feasibility criteria for stream matching are to be follows. First is stream population, the number of rich streams should be less than the number of lean streams above the pinch. Below the pinch, the number of rich streams should be more than the number of lean streams. Secondly, operating versus equilibrium line – a match above the pinch must have a minimum driving force of ε at the pinch side whereas the opposite for below the pinch. In the section of reducing the number of mass exchange units, the network can be simplified using the loop and part network “relaxation” technique. Consequently, the decreasing of the number of exchangers in the network impact the increasing of the external MSA used in the process. However, the flexibility of the designed network depends on the tradeoff between the operating cost and the capital cost of the exchanger.

Recently, Henwatthana (2006) proposed the development of MENs module for ASPEN PLUS. This module was developed as an interface to extract the required data from the simulation on ASPEN PLUS and communicate with the GA optimizer, Matlab. In this work, the freshwater minimization is considered as an example through reuse and regeneration recycle system with only single contaminant. The problem is formulated as a mixed integer nonlinear programming (MINLP) which variables are classified into two groups; independent and dependent variables. The mentioned MENs module is tested with two chemical processes namely, a tricresyl phosphate process and an ethyl chloride process. The results indicate that the module report the reliable solution for freshwater minimization. Additionally, the results also agree with the theory that the reuse and the regeneration recycle can reduce the freshwater consumption including wastewater generated but reuse cannot decrease the mass load of the specified contaminant while regeneration recycle can. The advantage of the constructed module is that it is flexible for any chemical processes and can decrease time consuming for calculating the input for freshwater minimization more than manually calculation.

1.1 Mass Exchanger Networks (MENs)

In the existing process industries, several mass exchanger units are available. In order to minimize the cost of MSAs, it is necessary to maximize the use of process MSAs before considering the application of external MSAs. Primarily, it signifies to realize on the thermodynamic limitations of the mass exchange equilibrium in the exchanger. Towards this end, one may use a method called “pinch diagram”. The mass exchange pinch diagram is a graphical method that is presented to identify the target of minimum external MSAs used in the process. The initial step in constructing the pinch diagram is creating a global representation for all rich streams. By plotting mass exchanged versus composition, each rich stream is represented as an arrow whose its tail corresponds to the supply composition and head to its target composition. We can define the mass that is removed from the i^{th} rich stream by:

$$MR_i = G_i(y_i^s - y_i^t) \qquad \text{where } i = 1, 2, ..., N_R \tag{1}$$

A convenient way to place each arrow is to stack the rich stream on the top of one another, starting with the stream which has the lowest target composition. The sample of two rich streams on the pinch diagram is given in Figure 3 (a). After construct each individual rich stream on the diagram, a rich composite stream has to be created. The rich composite stream is the cumulative mass lost by all rich streams. Using "diagonal rule" for superposition to add up mass in the overlapped regions of streams, the rich composite streams then can be formed as depicted in Figure 3 (b).

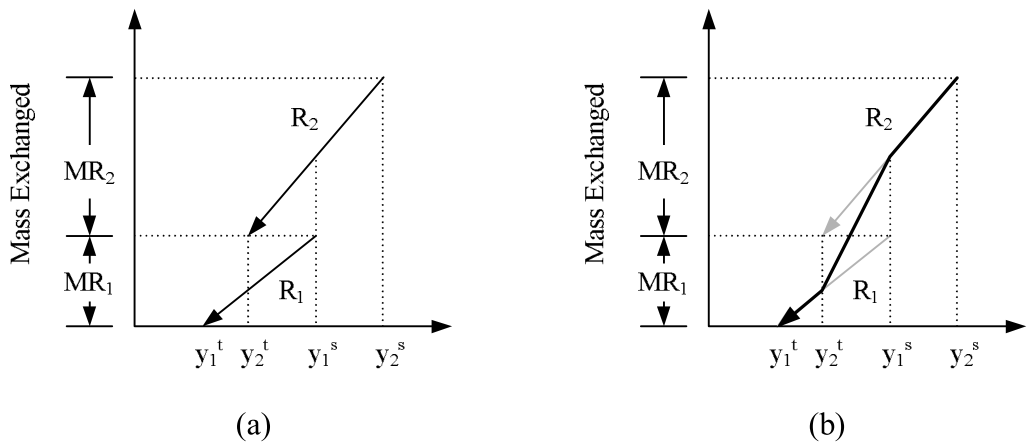


Figure 3 a) Representation of mass exchange by two rich streams
b) Constructing a rich composite stream using superposition

Source: Mann and Liu (1999)

Next, the global representation of all lean streams is developed as a lean composite stream. In contrast to the rich stream, the mass that can be gained by each process MSA is plotted versus the composition of that MSA using the relation of:

$$MS_j = L_j(x_j^t - x_j^s) \qquad \text{where} \qquad j = 1, 2, ..., N_{SP} \tag{2}$$

A convenient way to place the arrow is to stack the process MSA on the top of one another starting with the MSA having the lowest supply composition, Figure 4 (a). Hence, the lean composition stream can be obtained by cumulate the mass that can be gained by all process MSA using "diagonal rule". The complete lean composite stream is illustrated in Figure 4 (b).

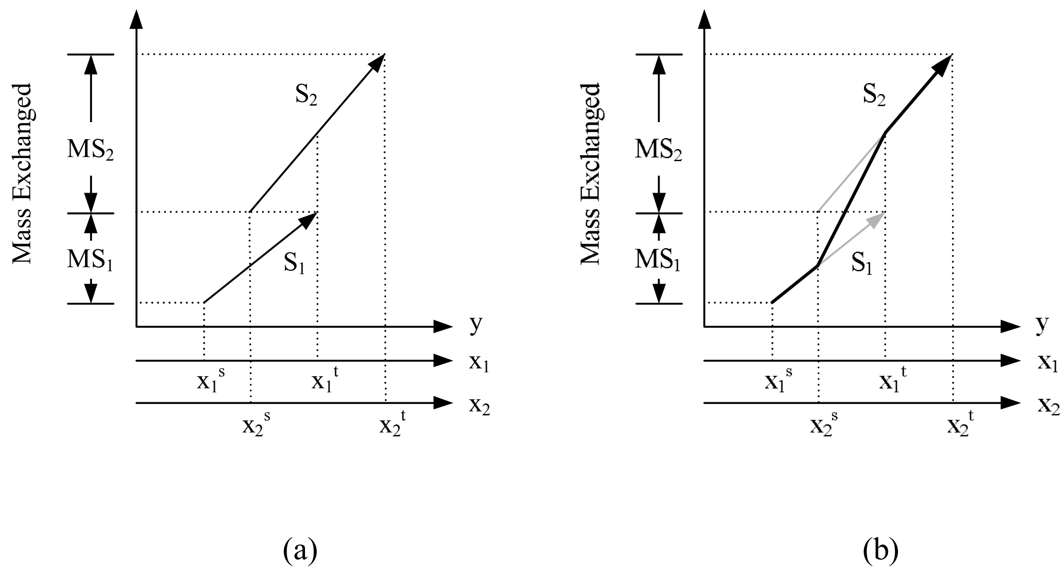


Figure 4 a) Representation of mass exchange by lean rich streams
b) Constructing a lean composite stream using superposition

Source: Mann and Liu (1999)

where $$x_j = \frac{y_i - b_j}{m_j} - \varepsilon_j$$

Next, both composite streams are plotted in the same diagram. On this diagram, thermodynamic feasibility of mass exchange is guaranteed when the lean composite stream always lies above the rich composite stream (Figure 5). The lean composite stream can be moved down until it touches the rich composite stream. The point where two composite streams touch is called “mass exchange pinch point”.

On the pinch diagram, the vertical overlap between two composite streams represents the maximum amount of the contaminant that can be transfer from the rich (waste) stream to the lean (MSA) stream. The mentioned zone is referred to as the “Integrated Mass Exchange”. The vertical distance above the integrated mass exchange zone is referred to as the “Excess Process MSAs”. It corresponds to the capacity of the process MSAs that unable to use because of the thermodynamic infeasibility. Finally, the vertical distance which lies below the integrated mass exchange zone is called “External MSAs Required”. This zone represents the amount of contaminants in the rich stream that have to be removed by external MSAs. Obviously, the increasing of the minimum concentration approach, ε, influence the mass exchange in the process – lower integrated mass exchange and more external MSAs are required.

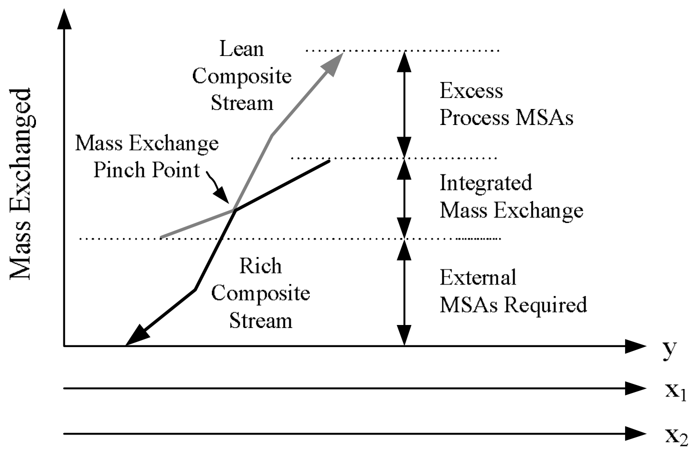


Figure 5 The mass-exchange pinch diagram

Source: Mann and Liu (1999)

As can be seen from Figure. 5, the pinch decomposes the synthesis problem into two regions: a rich end and a lean end. The rich end comprises all streams or parts of streams richer than the pinch composition. Similarly, the lean end includes all the streams or parts of streams leaner than the pinch composition. Above the pinch, exchange between the rich and the lean stream takes place. External MSAs are not required. Using an external MSA above the pinch will incur a penalty of eliminating an equivalent amount of process and the external lean streams should be used. Furthermore, Figure 5 indicates that if any mass is transferred across the pinch, the composite lean stream will move upward and, consequently, external MSAs in excess of the minimum requirement will be used. Therefore, to minimize the cost of external MSAs, mass should not be transferred across the pinch.

As mentioned earlier, the concept of MENs synthesis has been implemented in many process industries. Especially, it has been used in wastewater process. For wastewater process, wastewater is generated from many sources in the process. If the amount of wastewater is large, the cost for treatments and manufacturing will high. Generally, the most generated wastewater depends upon the amount of freshwater used in the process. Therefore, the freshwater consumption should be minimized. Thus, there are three general approaches to minimize the freshwater.

1. Reuse. Wastewater can be reused directly in other water-using operations when the level of previous contamination is in the acceptable range of the operations. This approach reduces both freshwater and wastewater volume but the mass load contaminant essentially unchanged. The schematic of this approach can be illustrated as shown in Figure 6.

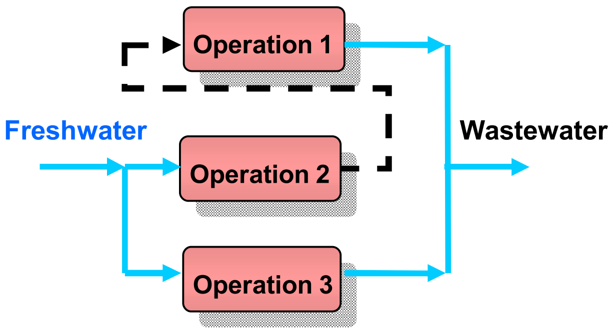


Figure 6 Water minimization through reuse

Source: Mann and Liu (1999)

2. Regeneration reuse. Wastewater can be regenerated by partial or total treatment to remove the contaminants. After regeneration, the regenerated water is not allowed to reenter the water-using operation in which the water stream has already been used. Therefore, the regenerated water will be reused in other water-using operation. This approach reduces both freshwater and wastewater volume and also decreases the mass load of contaminant. The schematic of regeneration reuse can be illustrated as shown in Figure 7.

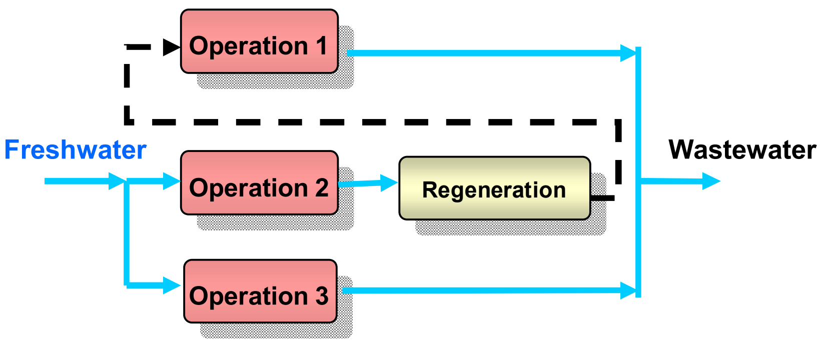


Figure 7 Water minimization through regeneration reuse

Source: Mann and Liu (1999)

3. Regeneration recycles. This approach is similar to regeneration reuse but the regenerated water may enter the water-using operation in which the water stream has already been used. The schematic of regeneration reuse can be illustrated as shown in Figure 8.

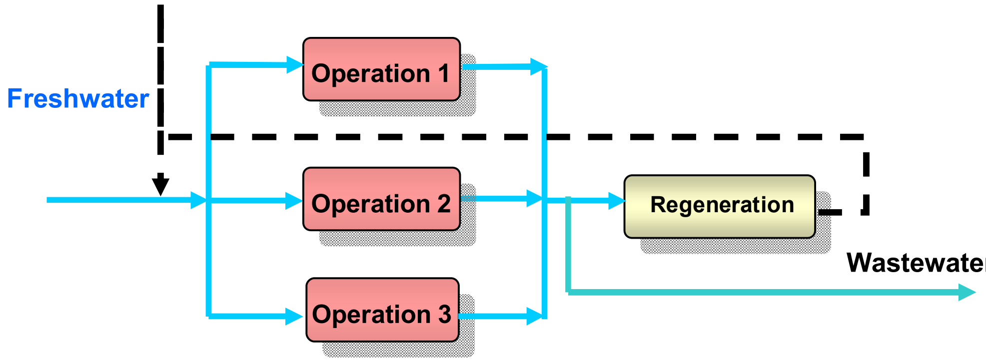


Figure 8 Water minimization through regeneration recycle

Source: Mann and Liu (1999)

2. Flexible Mass Exchanger Network design

While good design methodologies exist for the design of MENs with fixed parameters, changes in operating parameters might cause deviation from optimality or even cause control and safety problems. Therefore, a good MEN should be optimal for a set of nominal conditions as well as provide the flexibility to handle a range of operating conditions. Changes in operating conditions might originate from uncertainty in parameters during operation or from different operating conditions, which then refers to multi-period operation.

Cheng and Ping (2007) proposed a simple strategy for the synthesis of flexible heat exchange networks (HEN's) or mass exchange networks (MEN's) that involves expected disturbance range in the flow rates and temperatures (for HEN's) or compositions (for MEN's) of the inlet process streams. Optimization is done based on the stage-wise superstructure in order to get a minimum total annual cost (TAC).

3. Parametric programming

In an optimization framework, where the objective is to minimize or maximize a performance criterion subject to a given set of constraints and where some of the parameters in the optimization problem vary between specified lower and upper bounds, parametric programming is a technique for obtaining (i) the objective function and the optimization variables as a function of these parameters and (ii) the regions in the space of the parameters where these functions are valid (Fiacco, 1983; Gal, 1995; Acevedo and Pistikopoulos, 1996, 1997; Pertsinidis *et al.*, 1998; Papalexandri, 1998; Acevedo, 1999; Dua and Pistikopoulos, 1999). Some recent applications of this technique are,

1. hybrid parametric/stochastic programming (Acevedo and Pistikopoulos, 1997a; Hene´, Dua and Pistikopoulos,2001);
2. process planning under uncertainty (Pistikopoulos and Dua, 1998);
3. material design under uncertainty (Pistikopoulos and Dua,1998);
4. multi-objective optimization (Pistikopoulos and Grossmann,1988; Pertsinidis, 1992; Papalexandri and Dimkou, 1998);
5. flexibility analysis (Bansal *et al.,* 2000a);
6. computation of singular multi-variate normal probabilities (Bansal *et al.,* 2000b).

The main advantage of using the parametric programming techniques to address such problems is that for problems pertaining to plant operations, such as for process planning Pistikopoulos *et al.* (1998) and scheduling, one can obtain a complete map of all the optimal solutions. Moreover, as the operating conditions fluctuate, one does not have to re-optimize for the new set of conditions since the optimal solution as a function of parameters (or the new set of conditions) is already available. Pistikopoulos *et al.* (2002) propose the mathematical model for multi-parametric mixed-integer nonlinear programming problems of the following form:

$$
\begin{aligned}
& z(\theta) = \min d^T y + f(x) \\
& \text{s.t. } Ey + g(x) \le b + F\theta \\
& \theta_{\min} \le \theta \le \theta_{\max} \\
& x \in X \subseteq \Re^n \\
& y \in Y = \{0,1\}^m \\
& \theta \in \Theta \subseteq \Re^s,
\end{aligned} \tag{3}
$$

where y is a vector of 0 –1 binary variables, x is a vector of continuous variables, f is a scalar, continuously differentiable and convex function of x, g is a vector of continuously differentiable and convex functions of x, b and d are constant vectors, E and F are constant matrices, θ is a vector of parameters, θ_{min} and θ_{max} are the vectors of lower and upper bounds on θ, and X and Θ are compact and convex polyhedral sets of dimensions n and s, respectively. While the detailed theory and algorithms for solving Eq. (3) was proposed by Dua *et al.,* (1999, 2000), the engineering significance of solving Eq. (3) by using parametric programming.

4. Optimal operation of mass exchanger networks

In order to obtain the optimal operation, two conditions need to be considered. The first condition is to be sufficient of degree of freedom (DOFs) or manipulated variables for control which is used to check the possibility to control each variable independently by using the definition of number of degrees of freedom (N_{DOF}) proposed by Marselle *et al.* (1982) as shown in the following equation.

$$N_{DOF} = N_{units} - N_t \tag{4}$$

where

N_{units} is the number of exchanger units or manipulated variables (degrees of freedom)

N_t is the number of target temperatures.

This equation can calculate the number of degree of freedom by rank of heat exchanger unit. The determination of rank of heat exchanger unit is shown as example in figure 9.

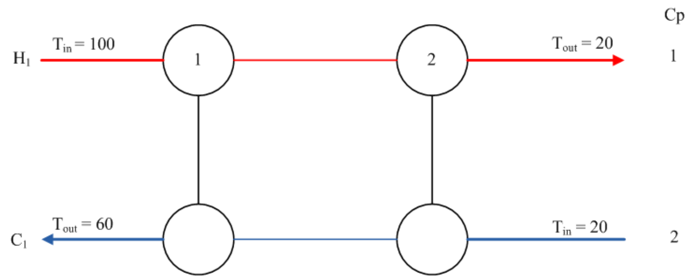


Figure 9 Heat exchanger network

From overall heat balance equation

$$(TIN_{i,p} - TOUT_{i,p})F_{i,p} = \sum_{st}\sum_{j} q_{i,j,st,p}$$

$$(TOUT_{j,p} - TIN_{j,p})F_{j,p} = \sum_{st}\sum_{i} q_{i,j,st,p}$$

$$\begin{aligned} 1(100-20) &= q_1+q_2 \\ 2(60-20) &= q_1+q_2 \end{aligned} \qquad \begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix} \qquad \text{Rank} = 1$$

Moreover, number of degree of freedom of mass exchanger networks can be calculated by rank of mass exchanger unit in this equation too. The example is shown in figure 10.

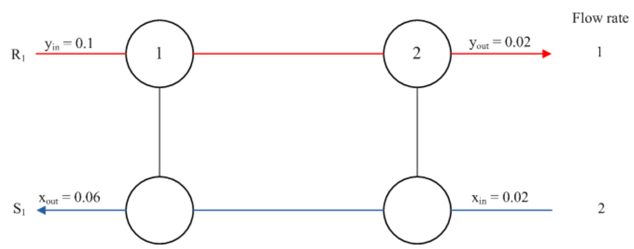


Figure 10 Mass exchanger network

From overall mass balance equation

$$(y_{i1} - y_{i,Ns+1})G_i = \sum_{k \in ST} \sum_{j \in LP} M_{ijk}$$
$$(x_{j1} - x_{j,Ns+1})L_j = \sum_{k \in ST} \sum_{i \in RP} M_{ijk}$$

$$\begin{aligned} 1(0.1-0.02) &= M_1+M_2 \\ 2(0.06-0.02) &= M_1+M_2 \end{aligned} \quad \Rightarrow \quad \begin{bmatrix} 1 & 1 \\ 1 & 1 \end{bmatrix} \quad \text{Rank} = 1$$

The condition $N_{DOF} > 0$ is necessary for the operation to be feasible and utility cost optimizable. Moreover, this condition is used to check the control design for setpoint satisfaction in case $N_{DOF} > 0$. Excluding two conditions, it has to using the more precise definition of the number of degrees of freedom with respect to utility cost optimization ($N_{DOF,U}$) proposed by Glemmestad (1997) as shown in the following:

$$N_{DOF,U} = DS + N_U - N_t \qquad (5)$$

where DS is the dimensional space spanned by the manipulated variables in the inner HEN to the outer HEN and N_U is the number of utility types.

So that can be concluded that if the first condition $N_{DOF,U} \geq 0$, the operation will be feasible and if the second condition $N_{DOF,U} > 0$, there will be extra degrees of freedom for utility cost optimization. This conclusion is shown in Table 1.

Table 1 The implication of $N_{DOF,U}$

Condition	(1)	(2)
$N_{DOF,U}$<0	No	No
$N_{DOF,U}$=0	Yes	No
$N_{DOF,U}$>0	Yes	Yes

Therefore, it is shown when $N_{DOF,U} > 0$, the operation of HENs will be more challenging. Aside from the setpoint satisfaction, the utility cost should also be minimized. This means that a common heuristic rule for the control design such as manipulating the last heat exchanger on a stream for a direct effect (Marselle *et al.*, 1982; Calandranis and Stephanopoulos, 1988; Mathisen, 1994) may not be preferred from an energy point of view.

MATERIALS AND METHODS

Materials

1. Personal computer
   a) CPU (Intel core2Duo CPU 1.66 GHz)
   b) 1.00 GB of RAM
   c) 70 GB of hard disk
2. Operating System: Microsoft Window Vista Home Premium service pack 1
3. Softwares
   a) GAMS versions 22.8 Beta
   b) MATLAB version 2007b
   c) ASPEN PLUS version 2006.5
   d) ASPEN DYNAMICS version 2006.5
   e) ASPEN ICARUS version 2006.5

Methods

1. The Mathematical Model for MENs Synthesis

Similar to heat exchanger networks, the synthesis of mass exchanger networks using the formulation of the mathematical model is proposed into two main techniques. First is the sequential synthesis whereas the minimum MSAs cost (operating cost) is considered follow by the minimization of the number of unit – investment cost. Second is the simultaneous synthesis in which the trade off between the operating cost and the investment cost are taken into account to minimize the total annual cost. Recently research showed that the optimal result from the simultaneous synthesis is more efficient than the sequential synthesis in the way of lower total annual cost of the network. This thesis, hence, will formulate the mathematical model to synthesis the mass exchanger networks using the simultaneous synthesis technique.

The simultaneous synthesis of mass exchanger networks is described based on the stage-wise superstructure which is directly adapted from that Yee and Grossmann presenting for the mass exchanger networks. In the stage-wise superstructure, potential exchanges between any pair of rich and lean streams can occur within each stage, and different sequences for matching streams are allowable by appending several numbers of stages in series. Therein the outlet of exchange units from splits of a common stream is mixed and then defines the streams for the next stage. Such as stated by Yee and Grossmann (1990a, 1990b) for synthesis of HEN's and due to the fact that an optimal design usually does not require a large number of exchange units, the number of stages required to model the mass integration is seldom be greater than either the number of rich streams *N*R or the number of lean streams *N*L. The number of stages is typically fixed at *N*S = max{*N*R, *N*L}. However, one additional stage is sometimes recommended to search for potential better networks. Figure 11 shows an example of a stage-wise superstructure involving two rich and two lean streams.

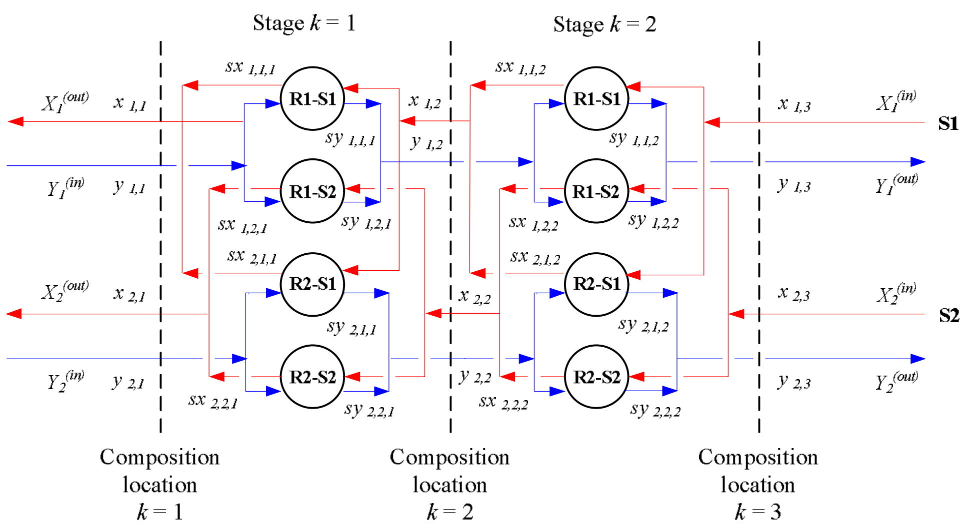


Figure 11 Two-stage superstructure for mass exchanger networks

Source: Chen and Ping (2005)

The two stages are represented by eight exchangers, with four possible matches in each stage and variable compositions between each stage. Instead of assuming iso-composition mixing of the split streams, the split streams in the same stage can possess different compositions. This is due to the fact that all compositions and flow rates for lean streams are variables for the typical MENs. The composition material balances around each stage for lean streams will still result in nonlinear constraints, even if we adopt the iso-composition assumption. Thus, one cannot guarantee a convex feasible space defined by a set of linear constraints with the simplified assumption. Notice that the derivation of the stage-wise superstructure does not require the identification of pinch point(s) or the partitioning into subnetworks. Furthermore, the model does not rely on any composition interval definition nor any transshipment type constraints (El-Halwagi & Manousiouthakis, 1990b).

For the model formulation of the simultaneous synthesis of mass exchanger networks, it can be formulated as a mixed integer nonlinear programming (MINLP) model. The objective function of this task is the minimum total annual cost which is subjected to eight constraints below.

Constraint 1*: Overall mass balance over the whole network*

An overall mass balance is needed to ensure sufficient exchange of any transferred component for all rich streams. The constraints specify that the overall mass transferable requirement of each rich stream must be equal to the sum of mass which is exchanged with other lean process streams or MSAs at each stage. Similar constraints also apply for all lean streams, as stated in the following.

$$(Y_i^{(\mathrm{in})} - Y_i^{(\mathrm{out})})G_i = \sum_{k \in ST} \sum_{j \in LP} M_{ijk} \qquad i \in RP \tag{6}$$

$$(X_j^{(\mathrm{out})} - X_j^{(\mathrm{in})})L_j = \sum_{k \in ST} \sum_{i \in RP} M_{ijk} \qquad j \in LP \tag{7}$$

***Constraint 2**: Mass balance in each stage*

These constraints are also needed to determine the composition of the transferable component as well as partition of flow rates for all parallel units. The composition location k = 1 involves the highest composition. The component and total mass balance for each stream in each stage are as follows:

$$(y_{ik} - y_{i,k+1})G_i = \sum_{j \in LP} M_{ijk} \qquad i \in RP \quad k \in ST \tag{8}$$

$$(x_{jk} - x_{j,k+1})L_j = \sum_{i \in RP} M_{ijk} \qquad j \in LP \quad k \in ST \tag{9}$$

$$G_i = \sum_{j \in LP} gg_{ijk} \qquad i \in RP \quad k \in ST \tag{10}$$

$$L_j = \sum_{i \in RP} ll_{ijk} \qquad j \in LP \quad k \in ST \tag{11}$$

***Constraint 3**: Mass balance in each exchange unit*

For the exchange unit between rich stream i and lean stream j in stage k, the compositions before mixers, sy_{ijk} and sx_{ijk}, are defined. A component mass balance is needed for each local exchange unit, where gg_{ijk} and ll_{ijk} are split mass flow rates.

$$gg_{ijk}(y_{ik} - sy_{ijk}) = M_{ijk} \qquad i \in RP \quad j \in LP \quad k \in ST \tag{12}$$

$$ll_{ijk}(sx_{ijk} - x_{j,k+1}) = M_{ijk} \qquad i \in RP \quad j \in LP \quad k \in ST \tag{13}$$

***Constraint 4**: Assignment of superstructure inlet/outlet compositions*

The given inlet/outlet compositions of the rich and lean streams are assigned as the inlet/outlet compositions to the superstructure. For rich streams, the superstructure inlet corresponds to composition location k = 1. While for lean streams, the inlet corresponds to location k = NS + 1.

$$Y_i^{(\text{in})} = y_{i,1} \qquad Y_i^{(\text{out})} = y_{i,Ns+1} \qquad i \in RP \tag{14}$$

$$X_j^{(\text{in})} = x_{j,Ns+1} \qquad X_j^{(\text{out})} = x_{j,1} \qquad j \in LP \tag{15}$$

Constraint 5*: Feasibility of the transferable component*

Constraints are also needed to guarantee monotonic decrease of all compositions at successive stages.

$$y_{ik} \geq y_{i,k+1} \qquad i \in RP \quad k \in ST \tag{16}$$

$$x_{jk} \geq x_{j,k+1} \qquad j \in LP \quad k \in ST \tag{17}$$

Constraint 6*: Logical constraints*

Logical constraints and binary variables, z_{ijk}, are needed to determine the existence of process match between streams i,j in stage k. An integer value of 1 for binary variable z_{ijk} designates that the match between rich stream i and lean stream j in stage k is presented in the optimal network. z_{ijk} is zero if the match (i, j) in stage k is absent, and thus the mass load M_{ijk} also becomes zero.

$$M_{ijk} - Uz_{ijk} \leq 0 \qquad i \in RP \quad j \in LP \quad k \in ST \tag{18}$$

Constraint 7*: Feasibility constraints of the equilibrium relationships*

Let $dyxi_{ijk}^{p}$ and $dyxo_{ijk}^{p}$, respectively, denote the composition approaches in the lean and rich ends of the mass exchanger between rich stream i and lean stream j in stage k. The following equilibrium relationships guarantee that the required minimum driving force, $m_{ij}\varepsilon_{ij}$, around the mass exchanger should this unit exist, (i.e. $z_{ijk} = 1$) will be overcome. These constraints are automatically relaxed if $z_{ijk} = 0$.

$$dyxi_{ijk}^{p}, dyxo_{ijk}^{p} \geq m_{ij}\varepsilon_{ij} \qquad i \in RP \quad j \in LP \quad k \in ST \tag{19}$$

$$dyxi_{ijk}^{p} \leq y_{ik}^{p} - m_{ij}sx_{ijk}^{p} - b_{ij} + \Gamma_{ij}^{p}\left(1 - z_{ijk}\right) \qquad i \in RP \quad j \in LP \quad k \in ST \tag{20}$$

$$dyxo_{ijk}^{p} \leq sy_{ijk}^{p} - m_{ij}sx_{jk+1}^{p} - b_{ij} + \Gamma_{ij}^{p}\left(1 - z_{ijk}\right) \qquad i \in RP \quad j \in LP \quad k \in ST \tag{21}$$

Constraint 8: *Sizing equation for mass transfer units*

Mass exchanger can be classified into two main categories: stage-wise exchangers and continuous-contact exchangers. The common types of stage-wise exchangers are tray or plate columns, and for continuous-contact exchangers are packed towers. When mass exchange takes place in a tray column, the number of required stages must be determined from the Kremser equation. Thus, the log-mean Kremser equation for stage numbers can be formulated as follows:

$$N_{ijk} \cong z_{ijk} \left[\frac{\left(\begin{array}{l} \max(0, y_{ik} - sy_{ijk}) \times \max(0, sy_{ijk}^{*} - y_{ij,k+1}^{*}) \\ \qquad \times \left[\max(0, y_{ik} - sy_{ijk}) + \max(0, sy_{ijk}^{*} - y_{ij,k+1}^{*}) \right] \end{array} \right) + \delta}{\left(\begin{array}{l} \max(0, y_{ik} - sy_{ijk}^{*}) \times \max(0, sy_{ijk} - y_{ij,k+1}^{*}) \\ \qquad \times \left[\max(0, y_{ik} - sy_{ijk}^{*}) + \max(0, sy_{ijk} - y_{ij,k+1}^{*}) \right] \end{array} \right) + \delta} \right]^{1/3} \tag{22}$$

When absorption or stripping takes place, a continuous-contact packed towers is suggested for mass exchange. The required packed height for *i,j* match in stage *k* for separation is characterized by a number of imaginary transfer units, NTU_{ijk}, and the overall height of a transfer unit, HTU_{ijk}. The overall packed height is given by the following equation, where the log-mean calculation is also applied from Chen's approximation and the smooth approximation method.

$$\mathrm{HTU}_{ijk} = \frac{g_{ijk}}{K_y aS} \qquad\qquad \mathrm{NTU}_{ijk} = \frac{y_{ik} - sy_{ijk}}{\text{log mean}\left[y_{ik} - sy^{*}_{ijk} \;,\; sy_{ijk} - y^{*}_{ij,k+1} \right]}$$

$$H_{ijk} = \mathrm{HTU}_{ijk} \times \mathrm{NTU}_{ijk} = \frac{M_{ijk}}{K_y aS} \times \frac{1}{\text{log mean}\left[y_{ik} - sy^{*}_{ijk} \;,\; sy_{ijk} - y^{*}_{ij,k+1} \right]}$$

$$= \frac{M_{ijk}}{K_y aS}\left[(y_{ik} - sy^{*}_{ijk})(sy_{ijk} - y^{*}_{ij,k+1}) \left(\frac{y_{ik} - sy^{*}_{ijk} + sy_{ijk} - y^{*}_{ij,k+1}}{2} \right) + \delta \right]^{-1/3}$$

$$= \frac{M_{ijk}}{K_y aS}\left[\left(\begin{array}{l} \max(0, y_{ik} - sy^{*}_{ijk}) \times \max(0, sy_{ijk} - y^{*}_{ij,k+1}) \\ \qquad \times \frac{1}{2}\left[\max(0, y_{ik} - sy^{*}_{ijk}) + \max(0, sy_{ijk} - y^{*}_{ij,k+1}) \right] \end{array} \right) + \delta \right]^{-1/3}$$

(23)

where $y^{*}_{ij,k+1} = m_{ij} x_{j,k+1} + b_{ij}$ and $sy^{*}_{ijk} = m_{ij} sx_{ijk} + b_{ij}$ are equilibrium composition

***Objective Function**: Minimum Total Annual Cost*

The objective function can be defined as the total annual cost for the network. The annual cost involved the combination of the operating cost of MSAs, and the tray and/or height costs for each exchanger.

Minimize $$\sum_{j \in LP} (AC_j)(L_j) + ACt \left(\sum_{i \in RP} \sum_{j \in LP} \sum_{k \in ST} \left(N_{ijk} \right) \right) + ACh \left(\sum_{i \in RP} \sum_{j \in LP} \sum_{k \in ST} \left(H_{ijk} \right) \right) \quad (24)$$

where N_{ijk} represented as a log mean Kremser equation for stage numbers using in the tray column mass exchanger

H_{ijk} represented as a log mean Hallale and Fraser equation for packed height using in the packed column mass exchanger

2. Feasibility test

After the multi-period simultaneous MINLP model has provided the optimal Mass Exchange Network structure for certain periods, one may ask if the configuration between the defined conditions is feasible. Chen and Ping (2007) was proposed how to ensure that the network is feasible in operating not only over these specified periods, but also over the whole range of the specified parameters, is discussed. This is referred to the task of keeping the outlet mass fraction in the network, defined by the MINLP model, at their target values during a short and long time horizon. Note that there is an assumption of perfect control, i.e., control can be adjusted depending on the realization of uncertain parameters and no delays in the measurements, or adjustments in the control are considered.

The NLP formulation has been developed by Chen and Ping (2007) to analyze the structural and final flexibility of the MEN. To identify the feasibility of each testing point, slack variables $sdyxi^{o}{}_{ijk}$ and $sdyxo^{o}{}_{ijk}$ are used to measure the violation of feasible driving forces for existing mass exchange units. The fixed binary parameters $z_{ijk}^{(ex)}$ are used to define whether the constraint of specific composition difference is involved or not.

$$z_{ijk}^{(ex)}\left(dyxi_{ijk}^{p} - \left(y_{ik}^{0} - m_{ij}sx_{ijk}^{0} - b_{ij} + sdyxi_{ijk}^{0}\right)\right) = 0 \tag{25}$$

$$z_{ijk}^{(ex)}\left(dyxo_{ijk}^{p} - \left(sy_{ijk}^{0} - m_{ij}x_{jk+1}^{0} - b_{ij} + sdyxo_{ijk}^{0}\right)\right) = 0 \tag{26}$$

$$\forall i \in RP, j \in LP, k \in ST, o \in \mathrm{O}$$

The problem of measuring the overall violation of each testing point $o \in O$ can be formulated as the following nonlinear programming (NLP).

$$\underset{x_{m'} \in \Omega_{M'}}{\text{Minimize}}\ J_{M'}^{o} = \sum_{\forall i \in RP} \sum_{\forall j \in LP} \sum_{\forall k \in ST} z_{ijk}^{(ex)} (sdyxi_{ijk}^{o} + sdyxo_{ijk}^{o}) \quad \forall o \in \mathrm{O} \tag{27}$$

where x_m and $\Omega_{M'}$ are design variables and feasible space, respectively, which are almost the same as x_m and $\Omega_{M'}$ except that all binary variables have fixed values, all $p \in$ MP are replaced by $o \in O$ and Eq. (25) is included. A positive $J^{o}_{M',\min}$ value implies the infeasible MEN structure for the oth testing point. The most violated testing point(s) can be appended as new period(s) for the synthesis of new MEN structure.

The synthesis problem of MENs in this work is not guarantee the global optimization.

3. Optimal operation of mass exchanger networks

This part describes the optimal operation of HENs that brings the method to optimal operation of MENs in this work.

3.1 LP formulation for optimal operation of MENs

Consider mass exchanger networks where the objective is to maintain optimal operation in spite of the variations in the inlet mass fraction. E.M. Sparrow *et.al*, (1998) showed that the corresponding steady-state optimal operation of MENs can be formulated as a linear programming (LP) problem:

Subject to:

$$\begin{aligned} &\min\ c^T x \\ &Ax \le b \\ &A_{eq}x = b_{eq} \\ &x_{\min} \le x \le x_{\max} \end{aligned} \tag{28}$$

The vector x consists of the inlet and outlet mass fraction on the rich side (y_i^{in} and y_i^{out}) and lean side (x_i^{in} and x_i^{out}) of all exchangers, as well as the duties of all exchangers (M_i-process exchanger and M_j-lean utility exchanger). The equality constraints include the process models, the internal connections, and given supply mass fraction y_i^s and target mass fraction y_i^t. The inequality constraints include the lower and upper bounds on the duties of all mass exchangers. The objective function allows for many problem formulations including maximum mass fraction problem. In this research, the objective is to minimize the utility cost in which all elements of the cost vector *c* are zero except the elements related to the duties of utility exchangers. The LP problem formulation for optimal operation of MENs is given in equations 29-41.

Objective function: $$\min \quad (\sum_i c_i M_i + \sum_j c_j M_j) \tag{29}$$

subject to

Constraint 1: Process models (mass balances)

for process exchanger *i*:

$$M_i - G_i \left(y_i^{in} - y_i^{out}\right) = 0 \tag{30}$$

$$M_i - L_i \left(x_i^{out} - x_i^{in}\right) = 0 \tag{31}$$

for regenerate *i*:

$$M_i - L_i \left(x_i^{out} - x_i^{in}\right) = 0 \tag{32}$$

Constraint 2: Connecting equations

supply connection:

$$y_i^{in} = y_i^s \tag{33}$$

$$x_i^{in} = x_i^s \tag{34}$$

internal connection:

$$y_i^{out} - y_j^{in} = 0 \tag{35}$$

$$x_i^{out} - x_j^{in} = 0 \tag{36}$$

target connection:

$$y_i^{out} = y_i^t \tag{37}$$

$$x_i^{out} = x_i^t \tag{38}$$

Constraint 3: Lower and upper bounds of mass exchanged

lower bound:

$$-M_i \leq 0 \tag{39}$$

upper bound: assuming constant mass transfer efficiency (ε) and mass flowrate

$$M_i \leq \varepsilon G_i \, (y_i^{in} - x_i^{in}) \tag{40}$$

ε : mass transfer efficiency of exchanger *i*,

$$\varepsilon = \frac{NTU_{r,i}(1 - e^{(NTU_{s,i} - NTU_{r,i})})}{NTU_{r,i} - NTU_{s,i} e^{(NTU_{s,i} - NTU_{r,i})}} \tag{41}$$

$$NTU_{r,i} = \frac{1}{R_m (m_a^{rich})_{\min}} = \frac{M_i}{(m_a^{rich})}, \; NTU_{s,i} = \frac{1}{R_m (m_a^{lean})_{\min}} = \frac{M_i}{(m_a^{lean})_{\min}}$$

3.2 Switching between active constraints

This section describes possible methods to implement the optimal policy by tracking the changing set of active constraints. To extend the application of split-range control to implement optimal operation of chemical processes, let us show a motivation example on a scrubber as shown in Figure 12. There are two types of MSA (MSA A and MSA B) for using in the scrubber. Assume that MSA A is cheaper than

MSA B. Hence, to operate the scrubber in an optimal manner (minimizing MSA cost), MSA A should be used in the nominal condition while MSA B should be a supplementary (e.g. the flow of MSA A reaches the upper limit). When MSA A is in use, the flow of MSA B presents the active constraint (saturated) at the lower bound. Likewise, when MSA B is in use (i.e. the flow of MSA A reaches the maximum limit), the flow of MSA A present at the active constraint (saturated) at the upper bound. This optimal operation can be implemented by the split-range control as shown in Figure 12.

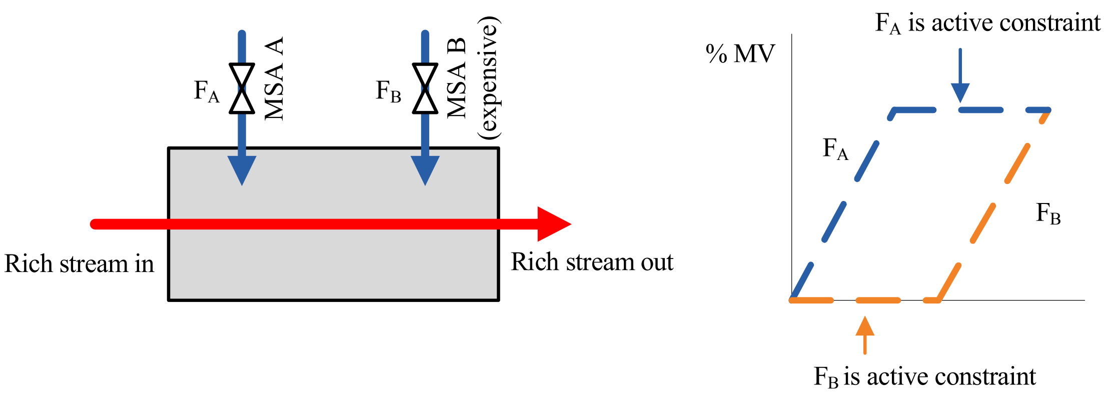


Figure 12 A scrubber system

For this scrubber system supposing that optimal operation always lies at the constraints of F_A or F_B depending on the variation of the desired outlet temperature. Hence, the active constraint regions can be divided to two regions as summarized in Table 2. In the two regions, F_A and F_B switch alternately as active constraint. For this example, the set of active constraints can be found obviously by inspection. However, for more complicated systems, a systematic method is required. In general, parametric programming (Kvasnica *et al.*, 2004) can be served for this task.

Table 2 Set of active constraints for furnace system

Region	F_A	F_B
1	U	S
2	S	U

U - Unsaturated manipulated variable (inactive constraint) to be used for control a system
S - Saturated manipulated variable (active constraint)

The information of set of active constraints will be used to design optimal split-range control structure as discussed in the next section.

3.3 Determination of optimal split-range control structure

From Table 2, to operate the furnace in an optimal manner, F_A and F_B should be switched alternately between being manipulated variables for and active constraints. Clearly, this can be implemented using a split-range controller with the combination of F_A and F_B. However, for more complicated system such as more number of manipulated variables and more number of active constraint regions, an integer linear programming (ILP) formulation (Lersbamrungsuk *et al.*, 2008) should be used in the design of optimal split-range control structure. The problem formulation can be shown as Case P1.

Constraint 1: Set of controlled and manipulated variables

CV: set of controlled variables, $CV = \{ CV_1 , CV_2 , \ldots, CV_{N_{CV}-1}, CV_{N_{CV}} \}$

MV: set of manipulated variables, $MV = \{ MV_1, MV_2, \ldots, MV_{Nm-1}, MV_{Nm} \}$

MVAAT: subset of *MV* with manipulated variables which are always active constraints (saturated at upper or lower bounds)

MVINAT: subset of *MV* with manipulated variables which are always inactive constraints (never saturated)

MVAT: subset of *MV* with manipulated variables which change between being active and inactive constraints

Constraint 2: Primary and secondary manipulated variables

Primary manipulated variable is a manipulated variable that is used for controlling an output (target), except when it is saturated. Secondary manipulated variable is a manipulated variable that is used to take over the task of a saturated primary manipulated variable.

Constraint 3: Relationship between primary and secondary manipulated variables

Let $x_{i,j}$ (where $i, j \in MV$) be a binary variable which represents the relationship between manipulated variable MV_i and manipulated variable MV_j

for $i=j$, $x_{i,j} = 1$ implies MV_i is a primary manipulated variable and $x_{i,j} = 0$ implies MV_i is a secondary manipulated variable or unused

for $i \neq j$, $x_{i,j} = 1$ implies MV_j is a secondary manipulated variable for MV_i and $x_{i,j} = 0$ implies MV_j is not a secondary manipulated variable for MV_i

Constraint 4: Relative order between manipulated variables and controlled variables

Let $r_{k,j}$ be relative order between controlled variable CV_k and manipulated variable MV_j. Relative order is a structural measure of how direct an effect an input has on an output (Daoutidis and Kravaris, 1992). However, we here assume $r_{k,j}$ as the number of exchangers between controlled variable CV_k and manipulated variable MV_j

Constraint 5: Relationship between controlled variables and manipulated variables

Let $z_{k,j}$ (where $k \in CV, j \in MV$) be a binary variable that represents the relationship between controlled variable CV_k and manipulated variable MV_j

$z_{k,j} = 1$ implies controlled variable CV_k is paired with manipulated variable MV_j and $z_{k,j} = 0$ implies controlled variable CV_k is not paired with manipulated variable MV_j

Case P1

Objective Function:

J_I : Minimizing the number of ''inter-connection'' or ''complexity'' of control structure (unnecessary relationships between primary and secondary manipulated variables).

J_{II} : Minimizing the sum of relative order of the control pairs.

$$J = \min(wJ_I + J_{II})$$

$$J_I = \sum_{i \in MV} \sum_{j \in MV, j \neq i} x_{i,j} \qquad\qquad J_{II} = \sum_{k \in CV} \sum_{j \in MV} r_{k,j} z_{k,j}$$

Subject to

Constraint 1: Assign one primary manipulated variables to each control objective.

$$\sum_{i \in MV} x_{i,i} = N_{CV} \tag{42}$$

Constraint 2: Manipulated variables MV_i that always is an active constraint should not be used for other purposes

$$x_{i,i} = 0 \qquad i \in MVAAT \tag{43}$$

$$\sum_{j \in MV, j \neq i} x_{i,j} = 0 \qquad i \in MVAAT \tag{44}$$

$$\sum_{j \in MV, j \neq i} x_{j,i} = 0 \qquad i \in MVAAT \tag{45}$$

Constraint 3: Manipulated variables MV_i that is never an active constraint is used as a primary manipulated variable with no need of a secondary manipulated variable.

$$x_{i,i} = 1 \qquad i \in MVINAT \tag{46}$$

$$\sum_{j \in MV, j \neq i} x_{i,j} = 0 \qquad i \in MVINAT \tag{47}$$

$$\sum_{j \in MV, j \neq i} x_{j,i} = 0 \qquad i \in MVINAT \tag{48}$$

Constraint 4: Manipulated variables MV_i that changes between being an active and inactive constraint may be a primary or secondary manipulated variables.

$$-x_{i,i} + \sum_{j \in MVAT, j \neq i} x_{i,j} = 0 \qquad i \in MVAT \tag{49}$$

$$x_{j,j} + \sum_{i \in MVAT, i \neq j} x_{i,j} \geq 1 \qquad j \in MVAT \tag{50}$$

$$M(x_{j,j} - 1) + \sum_{i \in MVAT, i \neq j} x_{i,j} \leq 0 \qquad j \in MVAT \tag{51}$$

Constraint 5: Possible and impossible split-range combination of manipulated variables (these constraints are obtained from the information of active constraint regions).

Constraint 5A: Impossible split-range combination of manipulated variables

$$\sum_{i \in MVAT^{A,R}} \sum_{j \in MVAT^{A,R}, j \neq i} x_{i,j} = 0 \qquad R \in RS \qquad (52)$$

Constraint 5B: Possible split-range combination of manipulated variables

$$x_{j,j} + \sum_{i \in MVAT^{I,R}} x_{i,j} \geq 1 \qquad j \in MVAT^{A,R}, R \in RS \qquad (53)$$

$$x_{i,i} + \sum_{j \in MVAT^{A,R}} x_{j,i} \geq 1 \qquad i \in MVAT^{I,R}, R \in RS \qquad (54)$$

Constraint 6: Assign one manipulated variables to each control objective

$$\sum_{j \in MV} z_{k,j} = 1 \qquad k \in CV \qquad (55)$$

Constraint 7: Only primary manipulated variables are paired with controlled variables.

$$-x_{j,j} + \sum_{k \in CV} z_{k,j} = 0 \qquad j \in MV \qquad (56)$$

It is possible for the ILP to have no feasible solution, that is, no optimal split-range control structure can be found. This may happen when there are conflicts among the equations in constraint 5. In this case, an online optimization may be suggested for implementing optimal operation.

4. Procedure for design of optimal split-range control structure

Additionally, the procedure for design of optimal split-range control structure of HENs can be developed to find optimal control structure of MENs.

The procedure for design of optimal split-range control structure consists of two steps:

Step 1: Check degrees of freedom (DOFs). Only the system with some degrees of freedom for optimization will be considered.

1. For systems in which no degree of freedom is available for optimization, the optimal operation can be reduced as control problems. (e.g. control pairing problems that can be handled using direct effect or RGA input saturation problem or input constraint problems (Giovanini *et al.,*2003))

2. For systems in which some degrees of freedom are available for optimization, go further to step 2.

Step 2: Find active constrain regions using parametric programming. (e.g. with MPT toolbox (Kvasnica *et al.,*2004))

3. For a number of regions: an optimal control structure can be found using Problem P1.

Example 1: A trivial MEN

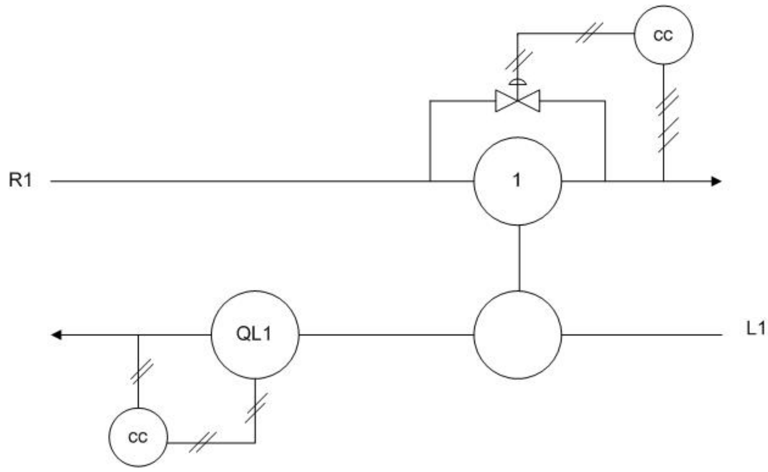


Figure 13 A trivial MEN

The MEN in Figure 13 contains one process exchanger and one utility types (N_U=1). Two outlet stream temperatures have targets (N_t =2). The dimensional space spanned by the manipulated variables in the inner MEN to the outer MEN (*DS*) is equal to 1 (see the calculation in Glemmestad, 1997). Using equation (5), we have $N_{DOF,U}=1+1-2=0$. Hence there is no need of an optimal operation strategy. If direct effect rules (Daoutidis *et al.*, 1992 and Soroush, 1996) are applied, then the resulting control structure can be shown in Figure 13.

Proposed strategy of an optimal operation for mass exchanger network is shown in Figure 14. First, the optimal mass exchanger network is formulated as a mixed integer nonlinear programming (MINLP). The mathematical programming formulate to minimize the total consumption of mass separating agents, the total number of stream matches and the total size of mass exchange units. Then, non linear programming (NLP) is used to verify whether the current network is operable for a large number of uncertain parameters that are generated randomly within the expected operating ranges; appending test point(s) that mostly violates the constraints of the network when the network configuration is proved infeasible through simulations, then returning to step (1) again to synthesize a new structure. After that the parametric programming is used to find active constraint regions. Then, ILP is used to formulate an optimal split-range control structure. Finally, check an optimal control structure using dynamics test.

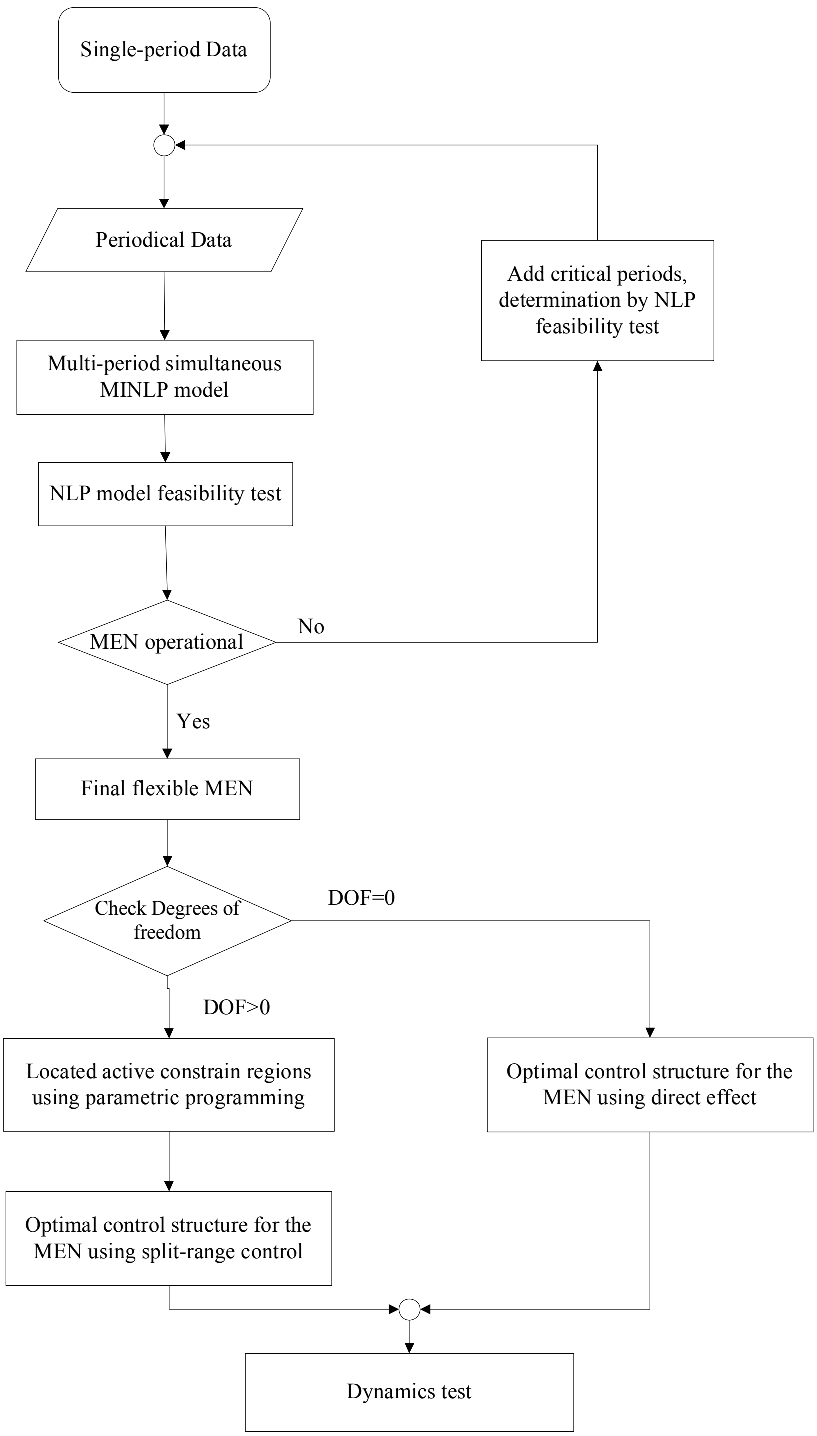


Figure 14 Proposed strategy of an optimal operation for MENs

RESULTS AND DISCUSSION

The results have been divided into three parts. The first part shows MINLP model and NLP feasibility in order to get a superstructure of Mass Exchange Networks (MENs) with flexibility. The second part is set of active constraints using multi parametric toolbox to find optimal control structure. The third part is the dynamical test of superstructure to ensure that the target composition can be controlled. In addition, this work presents three case studies.

Case study 1

The MENs introduced by El-Halwagi and Manousiouthakis (1989) involves the removal of H_2S from coke-oven gas (COG) a mixture H_2, CH_4, CO_2, N_2, NH_3, CO and H_2S, R_1 and Claus unit tail gases, R_2. Two external MSAs are used for removal of H_2S: aqueous ammonia, S_1 and chilled methanol, S_2. Stream data are given in Table 3.

Table 3 Streams data (nominal condition) for the case study 1

Rich streams	Description		G_i (kg/s)	Mass fraction	
				$Y^{(in)}$	$Y^{(up)}$
R_1	COG		0.9	0.07	0.0003
R_2	tail gases		0.1	0.051	0.0001
MSAs	Description	$L_j^{(up)}$ (kg/s)	Mass fraction		Annual cost ($/kg year)
			$X^{(in)}$	$X^{(up)}$	
S_1	Aq. NH3	2.3	0.0006	0.031	117360
S_2	Methanol	∞	0.0002	0.0035	176040

The equilibrium solubility data for H_2S in aqueous ammonia (with subscript 1) and methanol (subscript 2) can be correlated by the follow relations, respectively.

$$Y^{H2S} = 1.45x_1^{H2S}$$

$$Y^{H2S} = 0.26x_2^{H2S}$$

Table 4 Streams data (multi-period) for the case study 1

Period	streams	Flowrate (kg/s)	Mass fraction	
			in	out
Period 1	R_1	0.9	0.07	0.0003
	R_2	0.1	0.057	0.0001
	S_1	2.3	0.0006	0.031
	S_2	∞	0.0002	0.0035
Period 2	R_1	0.9	0.076	0.0003
	R_2	0.12	0.057	0.0001
	S_1	2.3	0.0006	0.031
	S_2	∞	0.0002	0.0035
Period 3	R_1	0.9	0.071	0.0003
	R_2	0.178	0.057	0.0001
	S_1	2.3	0.0006	0.031
	S_2	∞	0.0002	0.0035
Period 4	R_1	0.9	0.078	0.0003
	R_2	0.2	0.057	0.0001
	S_1	2.3	0.0006	0.031
	S_2	∞	0.0002	0.0035

Thus stage-wise superstructure approach is applied to synthesize the problem. To solve the multi-period simultaneous (MINLP) model for the related model, the General Algebraic Modeling System (GAMS) is used as the main solution tool. The solvers used are "DICOPT" for MINLP, "MINOS" for NLP and "CPLEX" for MIP.

Results of MINLP model are illustrated in Table 4. It features a total annual cost about \$936,650 year^{-1}, out of which is the cost of MSAs (\$813,187 year^{-1}). The networks contain five mass exchangers with the capital cost approximately \$123,463 year^{-1}. Moreover, the networks have 2 units for regenerate MSAs.

Table 5 Results of MINLP model about existence of match Rich Streams and lean Streams ($Z_{i,j,k}$) in stage k for the case study 1

Match (i,j)	Stage		Utility
	1	2	
R_1,S_1	1	1	
R_1,S_2		1	
R_2,S_1		1	
R_2,S_2		1	
S_1			1
S_2			1

This Table shows that there is one unit; match R_1-S, in stage 1. And there are four units, match R_1-S_1, match R_1-S_2, match R_2-S_1 and match R_2-S_2, in stage 2. In additional, there are 2 regenerate units for regenerate MSA of lean streams and recycle to inlet streams again. The final network configuration from MINLP is shown Figure 15.

The following step is to check the network with the NLP feasibility test model to ensure that network feasible for the overall operating range. The nominal stream data was given in Table 3, where the ammoniacal solution, R_1, may vary in between

[0.07, 0.078] %w/w for composition and chilled methanol, R_2, may vary between [0.1, 0.2] kg/s for flow rate.

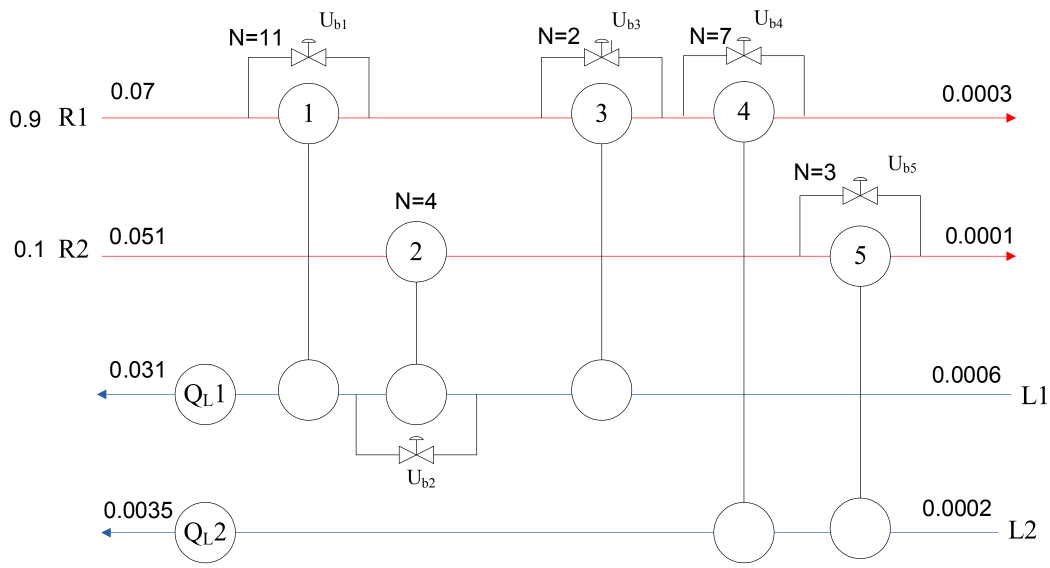


Figure 15 Final network configuration for the case study 1

After that, degrees of freedom are checked according to equation (5). We can be obtained $N_{DOF,U}$ =5+2-4=3. When DS = 5, N_U = 2 and N_t = 4. There are three degrees of freedom for utility cost optimization in case study 1. Therefore a strategy for optimal operation is needed.

And then, the information of set of active constraints will be used to design optimal split-range control structure. The multi-parametric toolbox (Kvasnica *et al.*, 2004) is used to find amount of active constraint regions. It is shown in Table 6.

Table 6 Set of active constraints in the case study 1

Active constraints region	Manipulated variables						
	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}
1	U	U	S_L	S_L	S_L	U	U
2	S_L	U	S_L	U	S_L	U	U

U - Unsaturated manipulated variable (inactive constraint)

S_L - Saturated manipulated variable (active constraint) at the lower bound

There are three active constraints in Table 6. It demonstrates that manipulated variable Q_{L1} and U_{b2} can become active constraints and they are combined as a split-range pair. Since manipulated variables U_{b1} and U_{b3} are saturated in both regions, they will not be active constraints. Furthermore, the manipulated variable Q_{L2}, U_{b4} and U_{b5} have never saturated, there is no need of split-range combinations.

Relative orders show relationship between manipulate variables and controller outputs. In Table 7, relative order = 1 represents that controller output is changed extremely when manipulate variable is adjusted. If relative order is increasing, controller output will be slightly changed when manipulate variable is adjusted. Moreover relative order = ∞ represents that controller output is hardly changed when manipulate variable is adjusted. It implied that manipulate variable adjust is not effect on controller output change as relative order = ∞.

Table 7 Relative orders of the MEN in the case study 1

MV / CV	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}
R1	∞	∞	4	5	3	1	2
R2	∞	∞	4	2	3	∞	1
L1	1	∞	2	3	4	∞	∞
L2	∞	1	∞	5	3	2	4

Then, the solver "CPLEX" of "GAMs" is used to solve the ILP problem. The results are optimal split-range pairs and controllability. The values of binary variables $x_{i,j}$ and $z_{k,j}$ from solving problem are shown in Tables 8 and 9, respectively. In Table 8, the primary manipulated variables are Q_{L2}, U_{b1}, $U_{b2,}$ $U_{b3,}$ U_{b4} and U_{b5} while the secondary manipulated variable is Q_{L1} for U_{b2}. If the primary manipulated variable, U_{b2}, is saturated, the secondary manipulated variable, Q_{L1}, is used to control output. Table 9 shows the control pairing, the direct effect, which are R1-U_{b4}, R2-U_{b5}, L1-Q_{L1} and L2-Q_{L2}. The resulting control structure is shown in Figure 16.

Table 8 The value of $x_{i,j}$ after solving problem for the case study 1

Primary MV \ Secondary MV	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}
Q_{L1}	1			1			
Q_{L2}		1					
U_{b1}			1				
U_{b3}					1		
U_{b4}						1	
U_{b5}							1

(the remaining entries are zero)

Table 9 The value of $z_{k,j}$ after solving problem for the case study 1

CV \ MV	Q_{L1}	Q_{L2}	U_{b4}	U_{b5}
R1			1	
R2				1
L1	1			
L2		1		

(the remaining entries are zero)

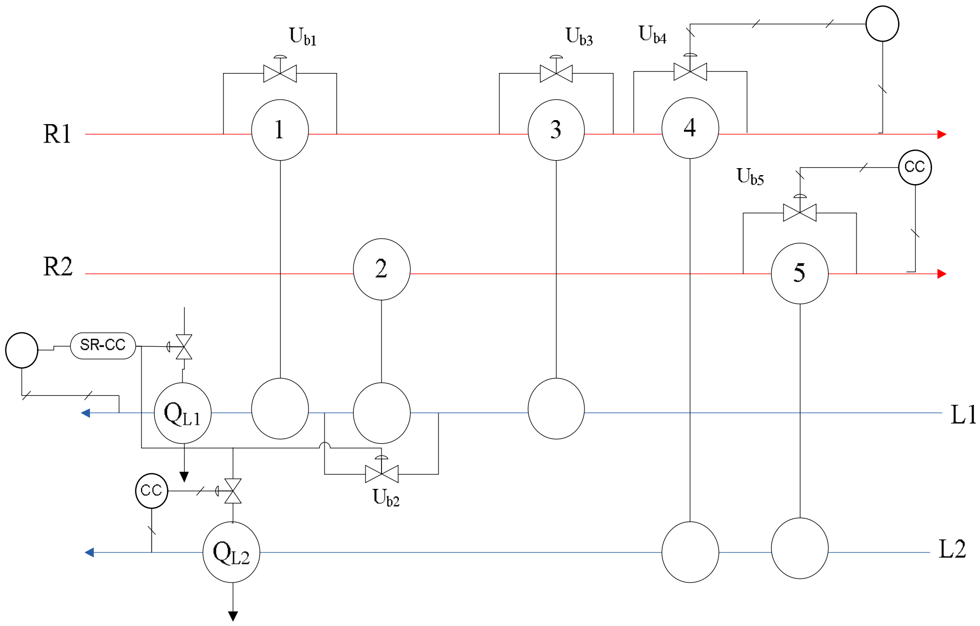


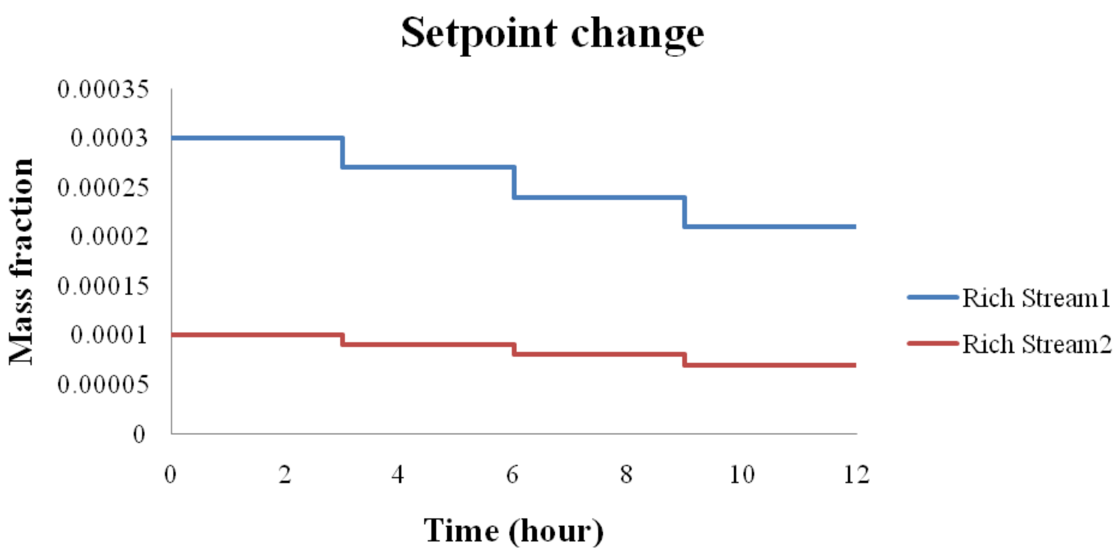
Figure 16 The control structure for the case study 1

Split-range signal from controllers can be obtained by referring to the information of active constraint regions in Table 6. The split-range signal of the pair of $Q_{L1}$and U_{b2} can switch alternatively to their lower constraints (SR-CC is split-range composition control).

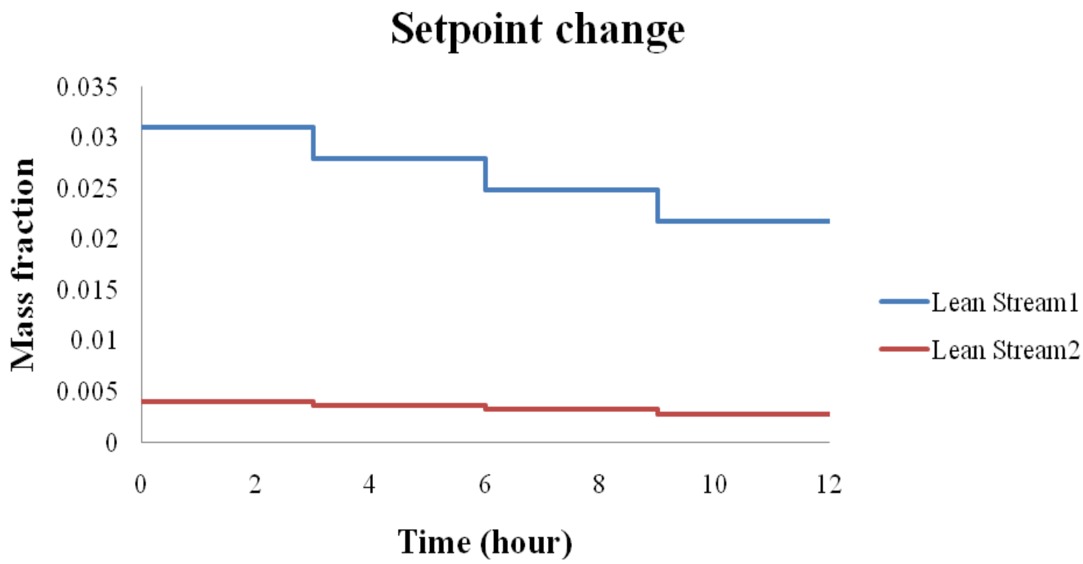
The result of control structures in MEN case study 1 is tested by performing the dynamic simulation on Aspen Dynamics v2006.5. The information of disturbances and active constraints of the system in case study 1 are shown in Table 10. The dynamic results show that the control structures can provide to optimality. Figure 17 illustrates the dynamic result of MEN with the control structures and shows the control efficiency to mass fraction under disturbance. And Figure 17c shows the response of split-range control that Q_{L1} is primary manipulate variable and U_{b2} is secondary manipulate variable to keep mass fraction of lean stream 2.

Table 10 Disturbances and active constraints in the case study 1

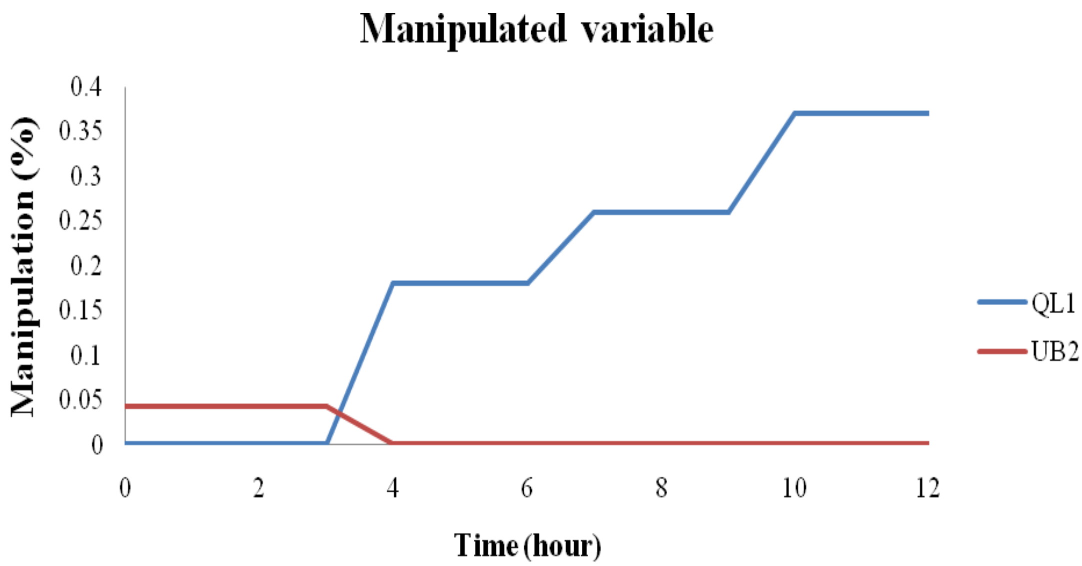
Time (hour)	Disturbance of mass fraction (set point change)				Active constraint	
	ΔR_1	ΔR_2	ΔL_1	ΔL_2	Q_{L1}	U_{b2}
<3	0	0	0	0	S_L	
>3	-0.00003	-0.00001	-0.0031	-0.00035		S_L
>6	-0.00006	-0.00002	-0.0062	-0.00070		S_L
>9	-0.00009	-0.00003	-0.0093	-0.00105		S_L



a) setpoint change of R1 and R2



b) setpoint change of S1and S2



c) Manipulated variable (Q_{L1} and U_{b2})

Figure 17 Dynamic simulation of the MEN in case study 1

These Figures 17a and 17b illustrated the results of dynamic test while system of MENs disturbed within reducing 10, 20 and 30% of mass fraction of rich streams and lean streams, respectively. And Figure 17c shows the results of manipulated variable while MENs disturbed by reduce 10, 20 and 30% of mass fraction. At more than 3 hours, U_{b2} is primary manipulate variable switch to Q_{L1} is secondary manipulated variable to adjust the rich stream 1 in order to obtain new setpoint. Then manipulated variable Q_{L1} increase to 0.27 and 0.35 for adjusted lean stream 1 in order to obtain new setpoint while MENs disturbed by reducing 20 and 30% of mass fraction. It can be concluded that manipulated variable Q_{L1} is direct proportion with reduced mass fraction.

And then, the determination basic equipment design data and construction cost for synthesized MEN using Aspen Icarus is approximately 4,200,000 US dollar. The equipment design data of each exchanger containing shell material, liquid volume, vessel diameter, vessel tangent to tangent height, design temperature, design gauge pressure, vacuum design gauge pressure, base material thickness and total weight are shown in Table 11.

Table 11 Equipment design data for the case study 1

Exchanger / Data	SEP 1	SEP 2	SEP 3	SEP 4	SEP 5	REGEN 1	REGEN 2
Shell material	A515	A515	A515	A515	A515	A515	A515
Liquid volume (GALLONS)	634.56	634.56	1,128.10	1,665.72	1,546.74	7,601.51	9,024.86
Vessel diameter (FEET)	3	3	4	4.5	4.5	7.5	8
Vessel tangent to tangent height (FEET)	12	12	12	14	13	23	24
Design temperature (DEG F)	250	250	250	250	250	250	250
Design gauge pressure (PSIG)	14.1	14.1	14.1	14.1	14.1	14.1	14.1
Application	CONT	CONT	CONT	CONT	CONT	CONT	CONT
Vacuum design gauge pressure (PSIG)	-14.696	-14.696	-14.696	-14.696	-14.696	-14.696	-14.696
Base material thickness (INCHES)	0.3125	0.3125	0.3125	0.3125	0.3125	0.375	0.375
Total weight (LBS)	2,600	2,600	3,600	4,400	4,300	13800	15200

Case study 2

The MENs from El-Halwagi and Manousiouthakis (1990a) involves the removal of a single component, the copper, from an ammoniacal etching solution and a rinsewater stream. Two external MSAs are available for removal of copper: LIX63 (an aliphatic α-hydroxyoxime, S1) and P1 (an aromatic β- hydroxyoxime, S2). Stream data are given in Table12.

Table 12 Stream data (nominal condition) for the case study 2

Rich stream	Description		G_i(kg/s)		Mass fraction $Y_i^{(in)}$	Mass fraction $Y_i^{(out)}$
R_1	Ammonium solution		0.25		0.13	0.10
R_2	Rinsewater		0.10		0.06	0.02
MSAs	Description	$L_j^{(up)}$	Mass fraction $X_j^{(in)}$	Mass fraction $X_j^{(out)}$	Annual operating cost ($/kg)	Annual operating cost ($/kg year)
S_1	LIX63	∞	0.03	0.07	0.01	58680
S_2	P1	∞	0.001	0.02	0.12	704160

Suppose mass transfers of copper are governed by the following linear equilibrium relations, where y and x denotes weight percent of copper in the rich and the lean streams respectively.

$$(R_1,S_1):\ y_1 = 0.734x_1 + 0.001$$
$$(R_2,S_1):\ y_2 = 0.734x_1 + 0.001$$
$$(R_1,S_2):\ y_1 = 0.111x_1 + 0.008$$
$$(R_2,S_2):\ y_2 = 0.148x_1 + 0.013$$

Table 13 Streams data (multi-period) for the case study 2

Period	streams	Flowrate (kg/s)	Mass fraction	
			in	out
Period 1	R_1	0.25	0.13	0.10
	R_2	0.10	0.06	0.02
	S_1	∞	0.03	0.07
	S_2	∞	0.001	0.02
Period 2	R_1	0.194	0.121	0.10
	R_2	0.10	0.06	0.02
	S_1	∞	0.03	0.07
	S_2	∞	0.001	0.02
Period 3	R_1	0.306	0.135	0.10
	R_2	0.10	0.06	0.02
	S_1	∞	0.03	0.07
	S_2	∞	0.001	0.02
Period 4	R_1	0.218	0.14	0.10
	R_2	0.10	0.06	0.02
	S_1	∞	0.03	0.07
	S_2	∞	0.001	0.02

Thus stage-wise superstructure approach is applied to synthesize the problem. To solve the multi-period simultaneous (MINLP) model for the related model, the General Algebraic Modeling System (GAMS) is used as the main solution tool. The solvers used are “DICOPT” for MINLP, “MINOS” for NLP and “CPLEX” for MIP.

Results of MINLP model are illustrated in Table 14. It features a total annual cost about \$614,299 year^{-1}, out of which is the cost of MSAs (\$542,205 year^{-1}). The networks contain three mass exchangers with the capital cost approximately \$72,094 year^{-1}. Moreover, the networks have 2 units for regenerate MSAs.

Table 14 Results of MINLP model about existence of match Rich Streams and lean Streams ($Z_{i,j,k}$) in stage k for the case study 2

Match (i,j)	Stage		Utility
	1	2	
R_1,S_1	1		
R_2,S_1	1		
R_2,S_2		1	
S_1			1
S_2			1

This Table shows that there are two units, match R_1-S_1 and match R_2-S, in stage 1. And there is one unit, match R_2-S_2, in stage 2. In additional, there are 2 regenerate units for regenerate MSA of lean streams and recycle to inlet streams again. The final network configuration from MINLP is shown Figure 18.

The following step is to check the network with the NLP feasibility test model to ensure that network feasible for the overall operating range. The nominal stream data was given in Table 12, where the ammoniacal solution, *R*1, may vary in between [0.194, 0.306] kg/s for flow rate and [0.12, 0.14] %w/w for composition.

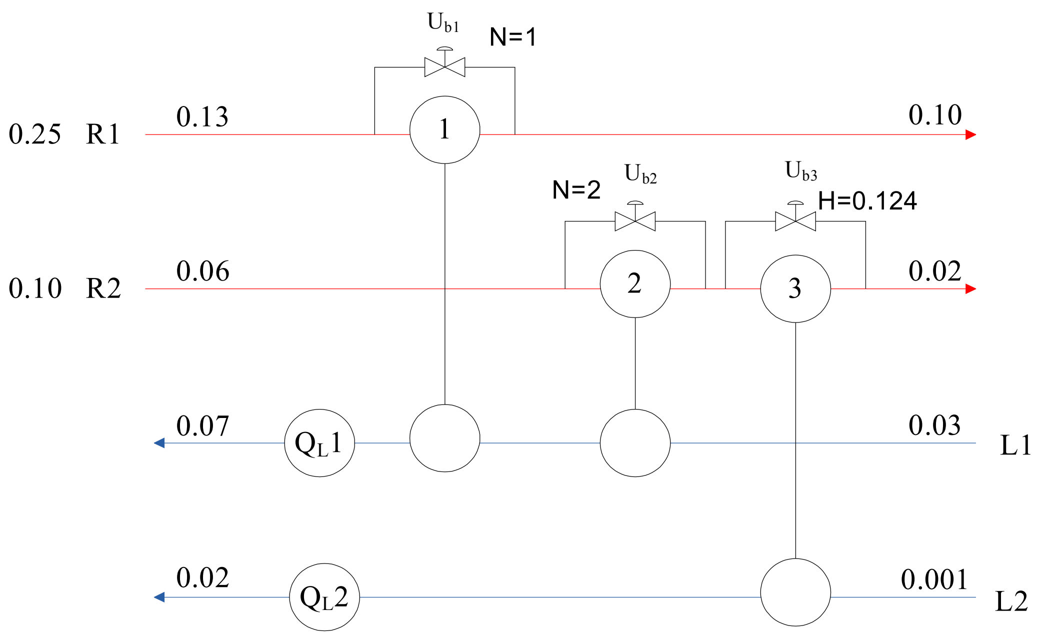


Figure 18 Final network configuration for the case study 2

After that, degrees of freedom are checked. We can be obtained N_{DOF} =1. There is one degree of freedom for utility cost optimization in case study 2. Therefore a strategy for optimal operation is needed.

And then, the information of set of active constraints will be used to design optimal split-range control structure. The multi-parametric toolbox (Kvasnica *et al.*, 2004) is used to find amount of active constraint regions. It is shown in Table 15.

Table 15 Set of active constraints in the case study 2

Active constraints	Manipulated variables				
region	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}
1	U	U	U	S_L	U
2	U	U	U	U	S_L

U - Unsaturated manipulated variable (inactive constraint)

S_L – Saturated manipulated variable (active constraint) at the lower bound

There are 2 active constraints in Table15. It demonstrates that manipulated variable U_{b2} and U_{b3} can become active constraints and they are combined as a split-range pair. Furthermore, the manipulated variables Q_{L2}, Q_{L1} and U_{b1} have never saturated, there is no need of split-range combinations.

The addition information of relative orders is shown in Table 16.

Table 16 Relative orders of the MEN in the case study 2

MV / CV	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}
R1	∞	∞	1	2	∞
R2	∞	∞	∞	2	1
L1	1	∞	2	3	∞
L2	∞	1	∞	3	2

Then, the solver "CPLEX" of "GAMs" is used to solve the ILP problem. The results are optimal split-range pairs and controllability. The values of binary variables $x_{i,j}$ and $z_{k,j}$ from solving problem are shown in Tables 17 and 18, respectively. In Table 17, the primary manipulated variables are Q_{L1}, Q_{L2}, $U_{b1}$and U_{b2} while the secondary manipulated variable is U_{b3} for U_{b2}. If the primary manipulated variable, U_{b3}, is saturated, the secondary manipulated variable, U_{b2}, is used to control output. Table 18 shows the control pairing which are R1-U_{b1}, R2-U_{b3} , L1-Q_{L1} and L2-Q_{L2}. The resulting control structure is shown in Figure 19.

Table 17 The value of $x_{i,j}$ after solving problem for the case study 2

Secondary MV / Primary MV	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}
Q_{L1}	1				
Q_{L2}		1			
U_{b1}			1		
U_{b2}				1	1

(the remaining entries are zero)

Table 18 The value of $z_{k,j}$ after solving problem for the case study 2

MV / CV	Q_{L1}	Q_{L2}	U_{b1}	U_{b3}
R1			1	
R2				1
L1	1			
L2		1		

(the remaining entries are zero)

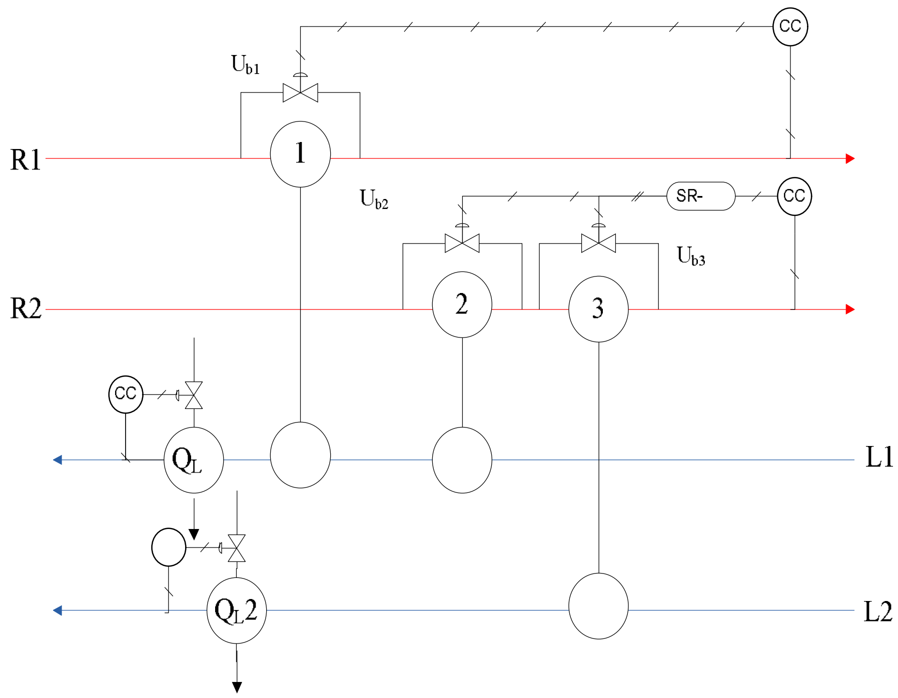


Figure 19 The control structure for the case study 2

Split-range signal from controllers can be obtained by referring to the information of active constraint regions in Table 15. The split-range signal of the pair of U_{b2} and U_{b3} can switch alternatively to their lower constraints (SR-CC is split-range composition control).

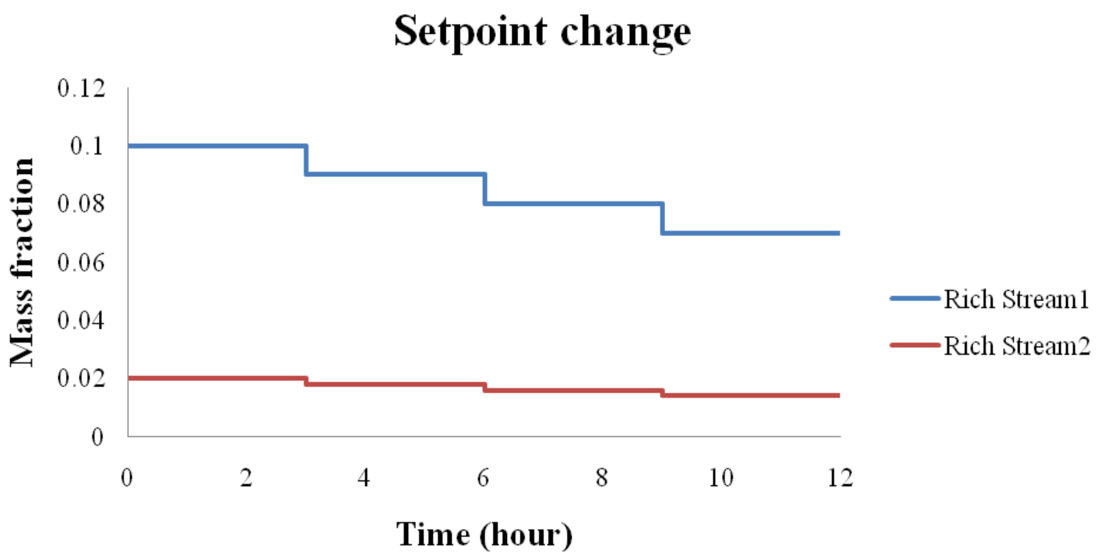
The result of control structures in MEN case study 2 is tested by performing the dynamic simulation on Aspen Dynamics v2006.5. The information of disturbances and active constraints of the system in case study 2 are shown in Table 19. The dynamic results show that the control structures can provide the optimality. Figure 20 illustrates the dynamic result of MEN with the control structures and shows the control efficiency to mass fraction under disturbance. And Figure 20b shows the response of split-range control that U_{b2} is primary manipulate variable and U_{b3} is secondary manipulate variable to keep mass fraction of lean stream 1.

Table 19 Disturbances and active constraints in the case study 2

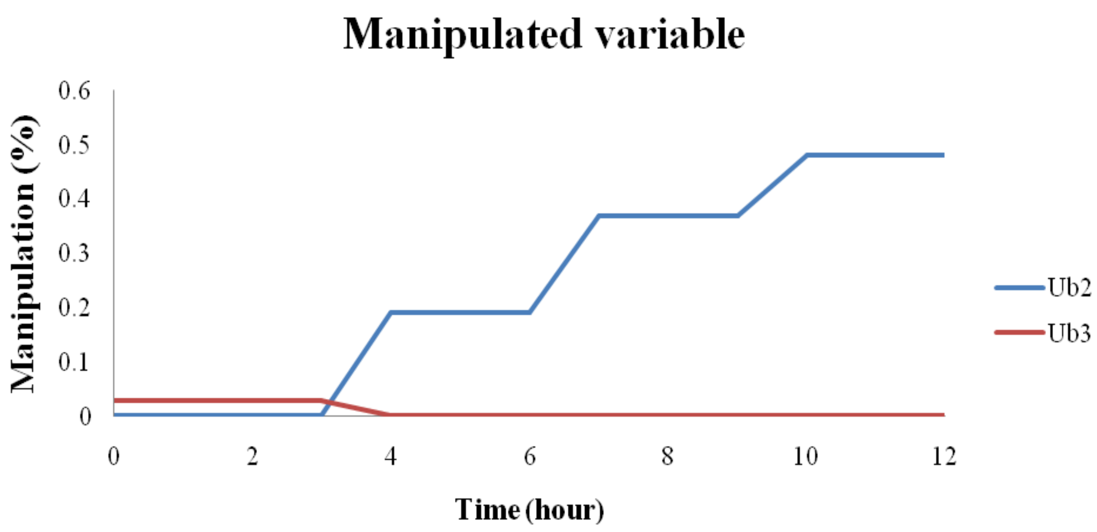
Time (hour)	Disturbance of mass fraction (set point change)				Active constraint	
	ΔR_1	ΔR_2	ΔL_1	ΔL_2	U_{b2}	U_{b3}
<3	0	0	0	0	S_L	
>3	-0.001	-0.002	-0.007	-0.002		S_L
>6	-0.002	-0.004	-0.014	-0.004		S_L
>9	-0.003	-0.006	-0.021	-0.006		S_L

These Figure 20a illustrated the results of dynamic test while system of MEN disturbed within reduce 10, 20 and 30% of mass fraction of rich streams and lean streams, respectively. And Figure 20b shows the results of manipulated variable while MENs disturbed by reduce 10, 20 and 30% of mass fraction. At more than 3 hours, U_{b3} is primary manipulate variable switch to U_{b2} is secondary manipulated variable to adjust the rich stream 1 in order to obtain new setpoint. Then manipulated variable U_{b2} increase to 0.36 and 0.48 for adjusted rich stream 1 in order to obtain new setpoint while MENs disturbed by reducing 20 and 30% of mass fraction. It can be concluded that manipulated variable U_{b2} is direct proportion with reduced mass fraction.

And then, determination basic equipment design data and construction cost for synthesized MEN using Aspen Icarus is construction cost approximately 3,360,000 US dollar. The equipment design data of each exchanger containing shell material, liquid volume, vessel diameter, vessel tangent to tangent height, design temperature, design gauge pressure, vacuum design gauge pressure, base material thickness and total weight are shown in Table 20.



a) setpoint change of R1 and R2



b) Manipulated variable (U_{b2} and U_{b3})

Figure 20 Dynamic simulation of the MEN in case study 2

Table 20 Equipment design data for the case study 2

Exchanger / Data	SEP 1	SEP 2	SEP 3	REGEN 1	REGEN 2
Shell material	A515	A515	A515	A515	A515
Liquid volume (GALLONS)	1,546.742	2,129.892	1,546.742	1,128.108	1,546.742
Vessel diameter (FEET)	4.5	5	4.5	4	4.5
Vessel tangent to tangent height (FEET)	13	14.5	13	12	13
Design temperature (DEG F)	250	250	250	250	250
Design gauge pressure (PSIG)	14.1	35.304	14.1	14.1	14.1
Application	CONT	CONT	CONT	CONT	CONT
Vacuum design gauge pressure (PSIG)	-14.696	-14.696	-14.696	-14.696	-14.696
Base material thickness (INCHES)	0.3125	0.3125	0.3125	0.3125	0.3125
Total weight (LBS)	4,300	5,800	4,300	3,600	4,300

Case study 3

The MENs introduced by El-Halwagi and Manousiouthakis (1990b) involves the removal of phenols from four aqueous waste streams (R1, R2, R3 and R4). Two MSAs are available: light oil (*S*1) and activated carbon (*S*2). Data are given in Table 21.

Table 21 Stream data (nominal condition) for the case study 3

Rich streams	Description	G_i (kg/s)	Mass fraction	
			$Y^{(in)}$	$Y^{(up)}$
R1	Wastewater	3.3	0.05	0.0015
R2	Condensate	0.6	0.07	0.003
R3	Waste stream	1.4	0.02	0.003
R4	Wastewater	0.2	0.03	0.002

MSAs	Description	$L_j^{(up)}$	Mass fraction		Annual operating cost	
			$X_j^{(in)}$	$X_j^{(out)}$	(\$/kg)	(\$/kg year)
S1	Light oil	10	0.0013	0.025	0.01	58680
S2	Caustic soda	10	0.001	0.015	0.07	417060

The equilibrium correlations for mass transfer between the rich streams and MSAs are:

$$S_1 : y = 0.71x_1 + 0.001$$
$$S_2 : y = 0.13x_2 + 0.001$$

Table 22 Streams data (multi-period) for the case study 3

Period	streams	Flowrate (kg/s)	Mass fraction	
			in	out
Period 1	R_1	3.3	0.05	0.0015
	R_2	0.6	0.07	0.003
	R_3	1.4	0.02	0.003
	R_4	0.2	0.03	0.002
	S_1	10	0.0013	0.025
	S_2	10	0.001	0.015
Period 2	R_1	3.3	0.051	0.0015
	R_2	0.61	0.07	0.003
	R_3	1.4	0.02	0.003
	R_4	0.2	0.03	0.002
	S_1	10	0.0013	0.025
	S_2	10	0.001	0.015
Period 3	R_1	3.3	0.055	0.0015
	R_2	0.63	0.07	0.003
	R_3	1.4	0.027	0.003
	R_4	0.2	0.03	0.002
	S_1	10	0.0013	0.025
	S_2	10	0.001	0.015

Table 22 (Continued)

Period	streams	Flowrate (kg/s)	Mass fraction	
			in	out
Period 4	R_1	3.3	0.053	0.0015
	R_2	0.614	0.07	0.003
	R_3	1.4	0.025	0.003
	R_4	0.2	0.03	0.002
	S_1	10	0.0013	0.025
	S_2	10	0.001	0.015

Thus stage-wise superstructure approach is applied to synthesize the problem. To solve the multi-period simultaneous (MINLP) model for the related model, the General Algebraic Modeling System (GAMS) is used as the main solution tool. The solvers used are “DICOPT” for MINLP, “MINOS” for NLP and “CPLEX” for MIP.

Results of MINLP model are illustrated in Table 23. It features a total annual cost about \$703,490 year^{-1}, out of which is the cost of MSAs (\$553,746 year^{-1}). The networks contain six mass exchangers with the capital cost approximately \$149,744 year^{-1}. Moreover, the networks have 2 units for regenerate MSAs.

Table 23 Results of MINLP model about existence of match Rich Streams and lean Streams ($Z_{i,j,k}$) in stage k for the case study 3

Match (i,j)	Stage			Utility
	1	2	3	
R_1,S_1	1	1		
R_1,S_2			1	
R_2,S_1			1	
R_3,S_1			1	
R_4,S_2			1	
S_1				1
S_2				1

This Table shows that there is one unit; match R_1-S_1, in stage 1. And there is one unit, match R_1-S_1, in stage 2. And there are four units, match R_1-S_1, match R_2-S_1, match R_3-S_1 and match R_4-S_2, in stage 3. In additional, there are 2 regenerate units for regenerate MSA of lean streams and recycle to inlet streams again. The final network configuration from MINLP is shown Figure 21.

The following step is to check the network with the NLP feasibility test model to ensure that network feasible for the overall operating range. The nominal stream data was given in Table 21, where the phenols solution, R_1, may vary in between [0.05, 0.055] %w/w for composition and R_2, may vary between [0.6, 0.63] kg/s for flow rate and R_3, vary between [0.02, 0.027] %w/w for composition.

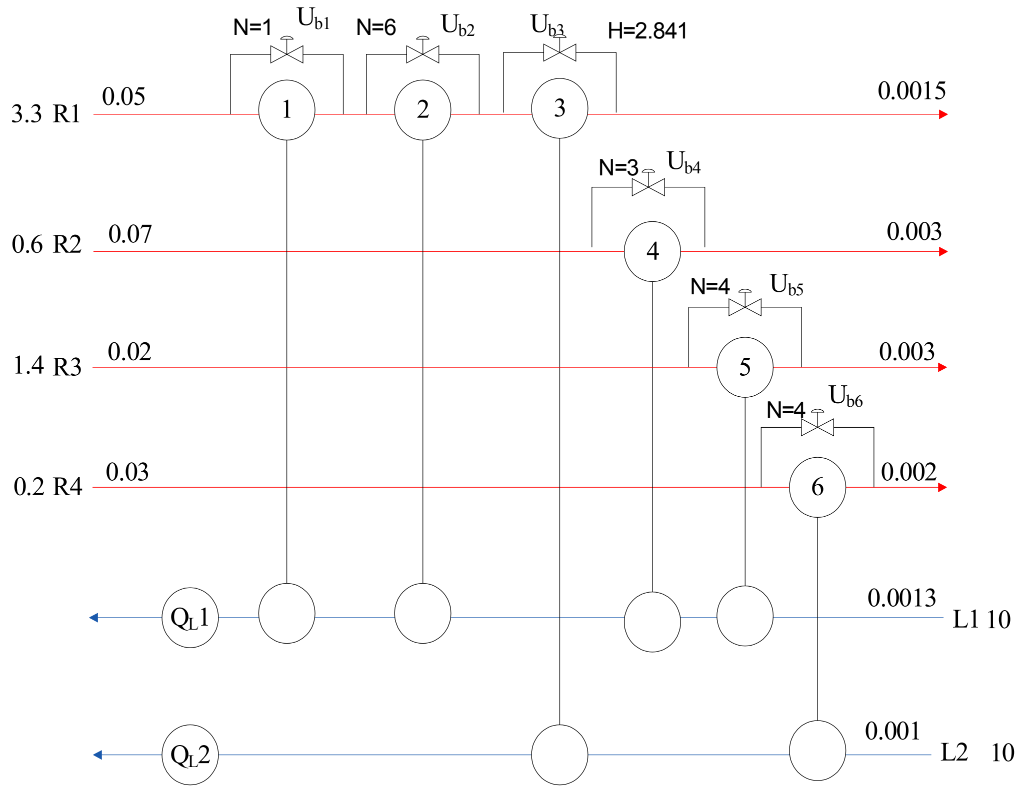


Figure 21 Final network configuration for the case study 3

After that, degrees of freedom are checked. We can be obtained $N_{DOF,U}$ =2. There are two degrees of freedom for utility cost optimization in case study 3. Therefore a strategy for optimal operation is needed.

And then, the information of set of active constraints will be used to design optimal split-range control structure. The multi-parametric toolbox (Kvasnica *et al*., 2004) is used to find amount of active constraint regions. It is shown in Table 24.

Table 24 Set of active constraints in the case study 3

Active constraints region	Manipulated variables							
	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}	U_{b6}
1	U	U	U	U	S_L	U	U	U
2	U	U	S_L	U	U	U	U	U

U - Unsaturated manipulated variable (inactive constraint)
S_L – Saturated manipulated variable (active constraint) at the lower bound

There are three active constraints in Table 24. It demonstrates that manipulated variable U_{b1} and U_{b3} can become active constraints and they are combined as a split-range pair. Since manipulated variables Q_{L1}, Q_{L2}, $U_{b2,}$ U_{b4}, U_{b5} and U_{b6} have never saturated, there is no need of split-range combinations.

The addition information of relative orders is shown in Table 25.

Table 25 Relative orders of the MEN in the case study 3

MV / CV	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}	U_{b6}
R1	∞	∞	6	3	1	4	5	2
R2	∞	∞	∞	∞	∞	1	2	∞
R3	∞	∞	∞	∞	∞	∞	1	∞
R4	∞	∞	∞	∞	∞	∞	∞	1
L1	1	∞	2	3	9	4	5	∞
L2	∞	1	∞	3	2	4	5	6

Then, the solver "CPLEX" of "GAMs" is used to solve the ILP problem. The results are optimal split-range pairs and controllability. The values of binary variables $x_{i,j}$ and $z_{k,j}$ from solving problem are shown in Tables 26 and 27, respectively. In Table 26, the primary manipulated variables are Q_{L1}, Q_{L2} , U_{b1}, Ub_2, Ub_4, Ub_5 and Ub_6 while the secondary manipulated variables is U_{b3} for U_{b1}. If the primary manipulated variable, U_{b1}, is saturated, the secondary manipulated variable, U_{b3}, is used to control output. Table 27 shows the control pairing which are R1-U_{b3}, R2-U_{b4}, R1-U_{b5}, R2-U_{b6}, L1-Q_{L1} and L2-Q_{L2}. The resulting control structure is shown in Figure 22.

Table 26 The value of $x_{i,j}$ after solving problem for the case study 3

Primary MV \ Secondary MV	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}	U_{b6}
Q_{L1}	1							
Q_{L2}		1						
U_{b1}			1		1			
U_{b2}				1				
U_{b4}						1		
U_{b5}							1	
U_{b6}								1

(the remaining entries are zero)

Table 27 The value of $z_{k,j}$ after solving problem for the case study 3

MV / CV	Q_{L1}	Q_{L2}	U_{b3}	U_{b4}	U_{b5}	U_{b6}
R1			1			
R2				1		
R3					1	
R4						1
L1	1					
L2		1				

(the remaining entries are zero)

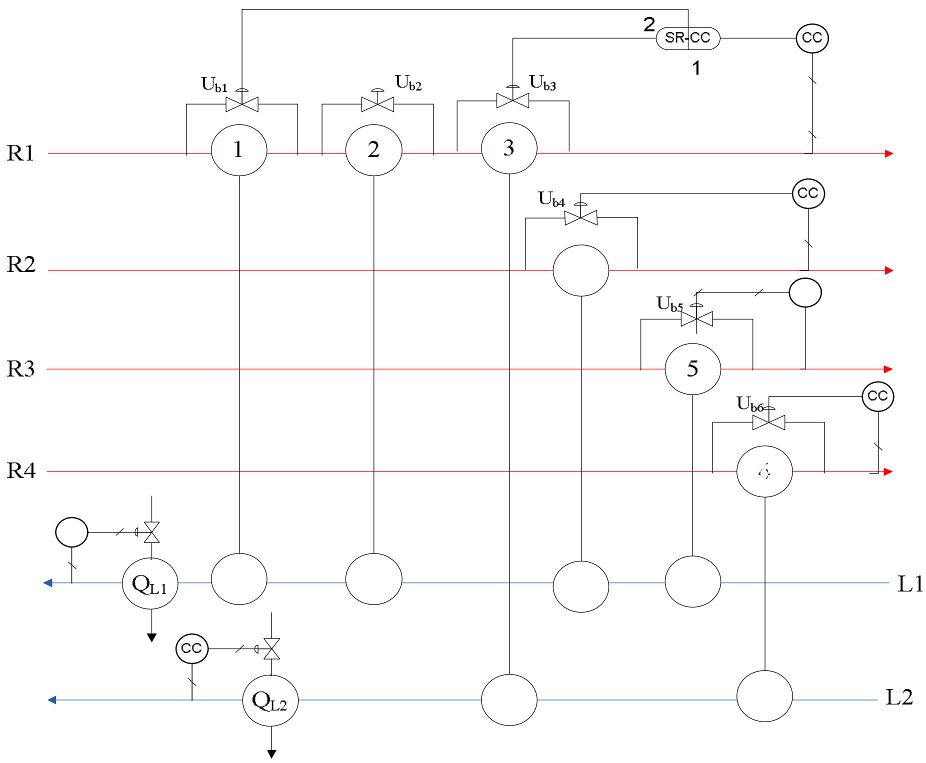


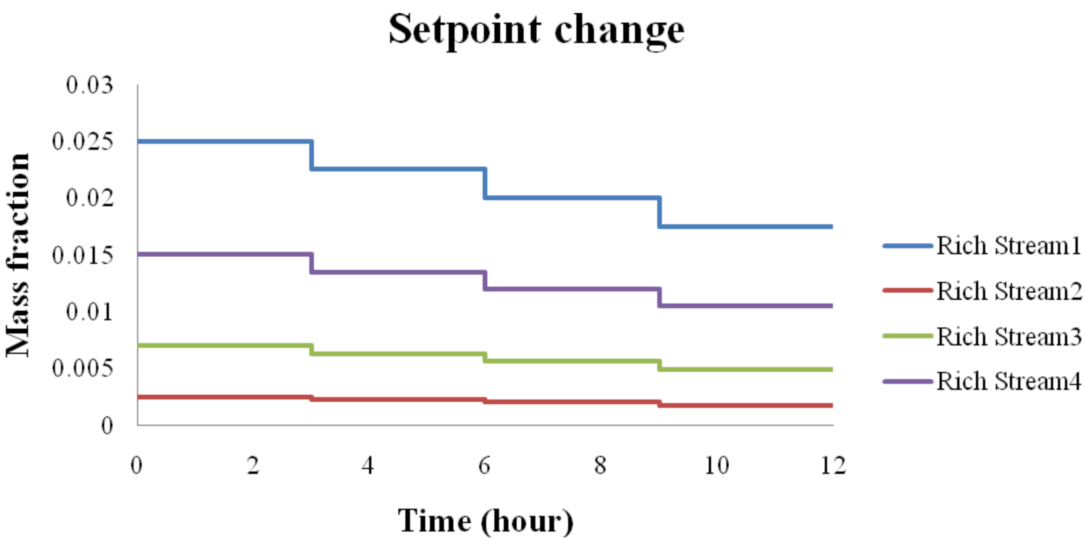
Figure 22 The control structure for the case study 3

Split-range signal from controllers can be obtained by referring to the information of active constraint regions in Table 24. The split-range signal of the pair of U_{b1} and U_{b3} they switch alternatively to their lower constraints (SR-CC is split-range composition control).

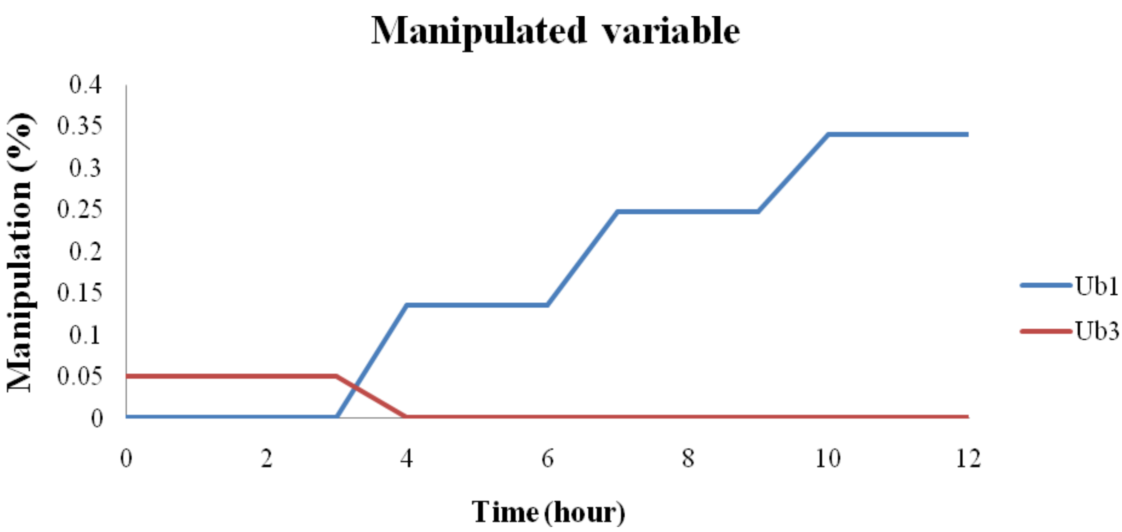
The result of control structures in MEN case study 3 is tested by performing the dynamic simulation on Aspen Dynamics v2006.5. The information of disturbances and active constraints of the system in case study 3 are shown in Table 28. The dynamic results show that the control structures can provide the optimality. Figure 23 illustrates the dynamic result of MEN with the control structures and shows the control efficiency to mass fraction under disturbance. And Figure 23b is show the response of split-range control that U_{b1} is primary manipulate variable and U_{b3} is secondary manipulate variable to keep mass fraction of lean stream 1.

Table 28 Disturbances and active constraints in the case study 3

Time (hour)	Disturbance of mass fraction (set point change)						Active constraint	
	ΔR_1	ΔR_2	ΔR_3	ΔR_4	ΔL_1	ΔL_2	U_{b1}	U_{b3}
<3	0	0	0	0	0	0		S_L
>3	-0.00015	-0.0003	-0.0003	-0.0002	-0.00025	-0.0015	S_L	
>6	-0.00030	-0.0006	-0.0006	-0.0004	-0.00050	-0.0030	S_L	
>9	-0.00045	-0.0009	-0.0009	-0.0006	-0.00075	-0.0045	S_L	



a) setpoint change of R1, R2, R3 and R4



b) Manipulated variable (U_{b1} and U_{b3})

Figure 23 Dynamic simulation of the MEN in case study 3

These Figures 23a illustrated the results of dynamic test while system of MEN disturbed within reduce 10, 20 and 30% of mass fraction of rich streams and lean streams, respectively. And Figure 23b shows the results of manipulated variable while MENs disturbed by reduce 10, 20 and 30% of mass fraction. At more than 3 hours, U_{b3} is primary manipulate variable switch to U_{b1} is secondary manipulated variable to adjust the rich stream 1 in order to obtain new setpoint. Then manipulated variable U_{b1} increase to 0.24 and 0.35 for adjusted rich stream 1 in order to obtain new setpoint while MENs disturbed by reducing 20 and 30% of mass fraction. It can be concluded that manipulated variable U_{b1} is direct proportion with reduced mass fraction.

And then, determination basic equipment design data and construction cost for synthesized MEN using Aspen Icarus is construction cost approximately 4,140,000 US dollar. The equipment design data of each exchanger containing shell material, liquid volume, vessel diameter, vessel tangent to tangent height, design temperature, design gauge pressure, vacuum design gauge pressure, base material thickness and total weight are shown in Table 29.

Table 29 Equipment design data for the case study 3

Exchanger / Data	SEP 1	SEP 2	SEP 3	SEP 4	SEP 5	SEP 6	REGEN 1	REGEN 2
Shell material	A515	A515	A515	A515	A515	A515	A515	A515
Liquid volume (GALLONS)	1,128.108	1,128.108	1,128.108	634.561	863.708	634.561	634.561	634.561
Vessel diameter (FEET)	4	4	4	3	3.5	3	3	3
Vessel tangent to tangent height (FEET)	12	12	12	12	12	12	12	12
Design temperature (DEG F)	250	250	250	250	250	250	250	250
Design gauge pressure (PSIG)	14.1	14.1	14.1	14.1	14.1	14.1	14.1	14.1
Application	CONT	CONT	CONT	CONT	CONT	CONT	CONT	CONT
Vacuum design gauge pressure (PSIG)	-14.696	-14.696	-14.696	-14.696	-14.696	-14.696	-14.696	-14.696
Base material thickness (INCHES)	0.3125	0.3125	0.3125	0.3125	0.3125	0.3125	0.3125	0.3125
Total weight (LBS)	3,600	3,600	3,600	2,600	3,100	2,600	2,600	2,600

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

Systematic approach is presented in this work to obtain an optimal control of mass exchange networks. First, the optimal mass exchanger network was formulated as a mixed integer nonlinear programming (MINLP). Then, NLP was also employed in flexibility testing. After that parametric programming was used to find active constraint regions. Then ILP was used to formulate an optimal split-range control structure. Next, optimal operation was obtained by putting features though dynamics test. Finally, Aspen Icarus was used to estimate the equipment design data and construction cost.

Additionally, three case studies were presented as examples in this work. Case study 1 from El-Halwagi and Manousiouthakis (1989), sweetening of COG, featured the total annual cost of \$936,650 year^{-1} distributed over 7 units of mass exchanger. Two active constraints were found and used to design optimal split-range control structure after degrees of freedom were determined. There were three degrees of freedom in this case. Case study 2 from El-Halwagi and Manousiouthakis (1990a), copper recovery in an etching plant, featured the total annual cost of \$614,299 year^{-1} distributed over 5 units of mass exchanger. Two active constraints were found and used to design optimal split-range control structure after degrees of freedom were determined. There was one degree of freedom in this case. Case study 3 from El-Halwagi and Manousiouthakis (1990b), removal/recovery of phenols from aqueous waste streams of a coal conversion plant, featured the total annual cost of \$703,490 year^{-1} distributed over 8 units of mass exchanger. Two active constraints were found and used to design optimal split-range control structure after degrees of freedom were determined. There were two degrees of freedom in this case. The determination of basic equipment design data and construction costs for synthesized MEN using Aspen Icarus were approximately 4,200,000 US dollars, 3,360,000 US dollars, 4,140,000 US dollars, respectively. Finally, all cases were checked through dynamics testing and they demonstrated that the obtained control structures could keep all target mass fractions at the desired values even under the saturation of some manipulated variables when system of MEN was disturbed by setpoint change.

Recommendations

1. In this work, we adjusted outlet mass fraction only. In practical we can have several ways to disturb the system of MENs such as flowrate change and inlet mass fraction change.

2. In this work, the MENs synthesis models were generated targeting a single contaminant removal. In reality, more than one contaminant might be exchanged in the same mass exchanger. Multi-component mass transfer, hence, should be considered.

3. The synthesis problem of MENs in this work does not guarantee the global optimization.

ITERATURE CITED

Aaltola, J. 2002. "Simultaneous Synthesis of Flexible Heat Exchanger Network", **Applied Thermal Engineering.** 22: 907–918.

Acevedo, J. and E.N. Pistikopoulos. 1996. A parametric MINLP algorithm for process synthesis problems under uncertainty. **Ind. Eng. Chem. Res.** 35: 147–158.

Acevedo, J. and E.N. Pistikopoulos. 1997. A multi parametric programming approach for linear process engineering problems under uncertainty. **Ind. Eng. Chem. Res.** 36: 717–728.

Acevedo, J. and E.N. Pistikopoulos. 1999. An algorithm for multi parametric mixed integer linear programming problems. **Operation Research Letters.** 24: 139–148.

Chen, C.L. and S.H. Ping. 2005, "Simultaneous synthesis of mass exchange networks for waste minimization", **Computers & Chemical Engineering**. 29: 1561-1576

Chen, C.L. and S.H. Ping. 2007, "Synthesis of flexible heat exchange networks and mass exchange networks", **Computers & Chemical Engineering**. 31: 1619-1632

Dua, V. and E.N. Pistikopoulos. 1999. Algorithms for the solution of multi parametric mixed-integer nonlinear optimization problems. **Ind. Eng. Chem. Res.** 38: 3976–3987.

Dua, V. and E.N. Pistikopoulos. 2000. An algorithm for the solution of multi parametric mixed integer linear programming problems. **Annals of Operations Research.** 99: 123–139.

El-Halwagi, M.M. 1997. **Pollution Prevention through Process Integration**, Alabama, Auburn University

El-Halwagi, M.M. and V. Manousiouthakis. 1989. “Synthesis of Mass Exchange Networks”, **American Institute of Chemical Engineers Journal: Chemical Engineering Research and Development**. 35: p. 1233.

El-Halwagi, M.M. and V. Manousiouthakis. 1990. “Automatic Synthesis of Mass Exchange Networks with Single-Component Targets”, **Chemical Engineering Science**. 45, p. 2813.

Fiacco, A.V. 1976. Sensitivity analysis for nonlinear programming using penalty methods. **Mathematical Programming.** 10: 287–311.

Flouds, C. 1995. **Nonlinear and Mixed Integer Optimization: Fundamentals and Applications**, New York, Oxford University Press, 462 p.

Foo, C.Y., Z.A. Mananb, R.M. Yunusb and R.A. Aziza. 2004. “Synthesis of MENs for Batch Processes – Part I: Utility targeting”, **Chemical Engineering Science.** 59: 1009-1026

Foo, C.Y., Z.A. Mananb, R.M. Yunusb and R.A. Aziza. 2005. “Synthesis of MENs for Batch Processes – Part II: Minimum Unit Target and Batch Network Design”, **Chemical Engineering Science.** 60: 1349-1362

Gal, T. 1995. **Postoptimal analyses, parametric programming, and related topics.** New York: de Gruyter.

Giovanini, L.L. and J.L. Marchetti. 2003. Low-level flexible-structure control applied to heat exchanger networks. **Computers & Chemical Engineering.** 27: 1129- 1142.

Glemmestad, B., K.W. Mathisen and T. Gundersen. 1996. Optimal operation of heat exchanger networks based on structural information. **Computers & Chemical Engineering.** 20: 823-828.

Glemmestad, B. 1997. **Optimal Operation of Integrated Processes: Studies on Heat Recovery Systems**. PhD Thesis. Norwegian University of Science and Technology.

Henwatthana, W. 2006. **Development of a Mass Exchange Networks Module for ASPEN PLUS**, Master of Engineering Thesis, Chemical Engineering Program, King Mongkut's University of Technology Thonburi, 117 p.

Kvasnica, M., P. Grieder, and M. Baotic. 2004. **Multi-parametric toolbox (MPT).** Available Source: http://control.ee.ethz.ch/~mpt, 1 March 2007.

Lersbamrungsuk, V. 2008. **Development of Control Structure Design and Structural Controllability for Heat Exchanger Networks.** PhD Thesis, Kasetsart University.

Lersbamrungsuk, V., S. Skogestad, and T. Srinophakun. 2006. A simple strategy for optimal operation of heat exchanger networks. In: **International Conference on Modeling in Chemical and Biological Engineering Sciences**, Bangkok, Thailand.

Lersbamrungsuk, V., T. Srinophakun, S. Skogestad, and S. Narasimhan. 2008. Control structure design for optimal operation of heat exchanger networks. **AIChE J.** 54: 150-162.

Mann, J and A.Y. Liu. 1999. Industrial Water Reuse and Wastwater Minimization, McGraw-Hill Professional.

Marselle, D.F., M. Morari and D.F. Rudd. 1982. Design of resilient processing plantsII Design and control of energy management systems. **Chem. Eng. Sci**. 37: 259-270

Prakotpol, D. 2001. **Development of MATLAB Toolbox with Genetic Algorithm for Water Pinch Technology**, Master of Engineering Thesis, Chemical Engineering Program, King Mongkut's University of Technology Thonburi, 116 p.

Papalexandri, K.P. and E.N. Pistikopoulos. 1994a. Synthesis and retrofit design of operable heat exchanger networks-I, flexibility and structural controllability aspects. **Ind. Eng. Chem. Res.** 33: 1718-1737.

Papalexandri, K.P. and E.N. Pistikopoulos. 1994a. Synthesis and retrofit design of operable heat exchanger networks-II, dynamics and control structure considerations. **Ind. Eng. Chem. Res.** 33: 1738-1755

Papalexandri, K.P., E.N. Pistikopoulos and Floudas, A. 1994. "Mass Exchange Networks for Waste Minimization: A Simultaneous approach", **Chemical Engineering Research & Design.** 72: Part A, p. 279.

Swaney, R.E. and I.E. Grossmann. 1985a. An index for operational flexibility in chemical process design, past I: formulation and theory. **AICHE J.** 31; 621-630

Swaney, R.E. and I.E. Grossmann. 1985b. An index for operational flexibility in chemical process design, past II: computational algorithms. **AICHE J.** 31; 631-641.

Wang, Y.P. and Smith, R. 1994. "Wastewater Minimization", **Chemical Engineering Science.** 49: p. 981.

Wang, Y.P. and R. Smith. 1995, “Wastewater Minimization with Flowrate Constraints”, **Chemical Engineering Research and Design.** 73: Part A, p. 889.

Thippaloj, K. 2002. **Multi-Component Optimization of Simultaneous Heat and Mass Exchanger Networks Using GAMs**, Master of Engineering Thesis, Chemical Engineering Program, Kasetsart University, 221p.

APPENDIX

Change nominal condition for synthesis mass exchanger networks

Synthesis mass exchanger networks is obtained local optimization, so change nominal condition for synthesis mass exchanger networks effects on gained structure would be changed. But this algorithm can used to control system design by change nominal condition of case study 1 to maximum condition to examine algorithm illustrated following

Example 1

Appendix Table 1 Streams data for the example 1 (nominal condition is a maximum condition)

Period	streams	Flowrate (kg/s)	Mass fraction	
			in	out
Period 1	R_1	0.9	0.078	0.0003
	R_2	0.2	0.057	0.0001
	S_1	2.3	0.0006	0.031
	S_2	∞	0.0002	0.0035
Period 2	R_1	0.9	0.076	0.0003
	R_2	0.12	0.057	0.0001
	S_1	2.3	0.0006	0.031
	S_2	∞	0.0002	0.0035

Appendix Table 1 (Continued)

Period 3	R_1	0.9	0.071	0.0003
	R_2	0.178	0.057	0.0001
	S_1	2.3	0.0006	0.031
	S_2	∞	0.0002	0.0035
Period 4	R_1	0.9	0.07	0.0003
	R_2	0.1	0.057	0.0001
	S_1	2.3	0.0006	0.031
	S_2	∞	0.0002	0.0035

The stage-wise superstructure approach was applied to synthesize the problem. To solve the multi-period simultaneous (MINLP) model for the related model, the General Algebraic Modeling System (GAMS) was used as the main solution tool. The solvers used were "DICOPT" for MINLP, "MINOS" for NLP and "CPLEX" for MIP.

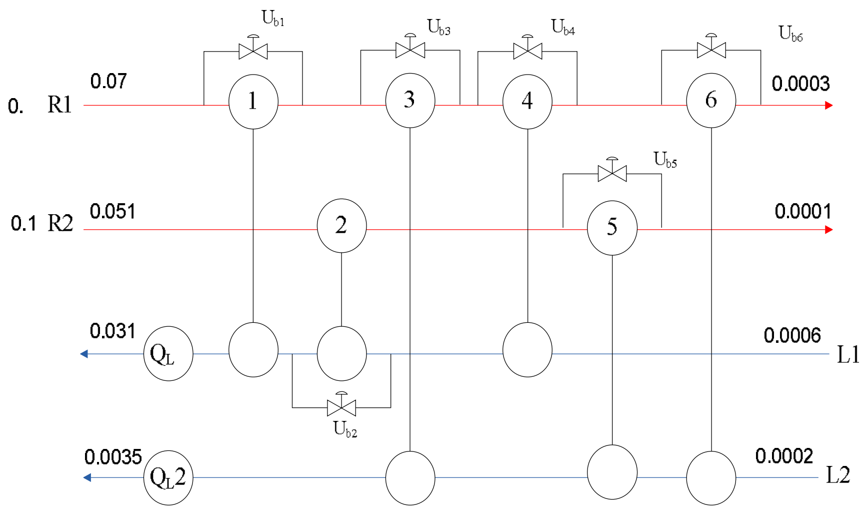
Results of MINLP model are illustrated in Appendix Table 2. It features a total annual cost about \$1,005,601 year^{-1}, out of which \$826,335 year^{-1} is the cost of MSAs. The networks contain five mass exchangers with the capital cost approximately \$179,266 year^{-1}. Moreover, the networks have 2 units for regenerate MSAs.

Appendix Table 2 Results of MINLP model about existence of match ($Z_{i,j,k}$) in stage k for the example 1

Match (i,j)	Stage		Utility
	1	2	
R_1,S_1	1	1	
R_1,S_2	1	1	
R_2,S_1	1	1	
R_2,S_2		1	
S_1			1
S_2			1

The Table shows that there is three units; match R_1-S_1, match R_1-S_2 and match R_2-S_1 in stage 1. And there is four units, match R_1-S_1, match R_1-S_2, match R_2-S_1 and match R_2-S_2, in stage 2. In additional, there are 2 regenerate units.

In the next step, check the network with the NLP feasibility test model to ensure that network feasible for the overall operating range. The final network configuration from MINLP is shown Appendix Figure 1.



Appendix Figure 1 Final network configuration for the example 1

After that, degree of freedom are checked. We can be obtained $N_{DOF,U}$ =4. There are four degrees of freedom for utility cost optimization in example 1. Therefore a strategy for optimal operation is needed.

And then, the information of set of active constraints will be used to design optimal split-range control structure. The multi-parametric toolbox (Kvasnica *et al.*, 2004) is used to find amount of active constraint regions. It is shown in Appendix Table 3.

Appendix Table 3 Set of active constraints in the example 1

Active constraints region	Manipulated variables							
	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}	U_{b6}
1	S_L	S_L	S_L	S_L	$\underline{U}$	U	U	U
2	S_L	S_L	U	S_L	S_L	U	U	U
3	S_L	S_L	U	S_L	S_L	U	U	U

U - Unsaturated manipulated variable (inactive constraint)

S_L – Saturated manipulated variable (active constraint) at the lower bound

There are four active constraints in Appendix Table 3. It demonstrates that manipulated variable U_{b1} and U_{b3} can become active constraints and they are combined as a split-range pair. Since manipulated variables Q_{L1}, QL2 and U_{b2} are saturated in each region, they can't become active constraints. Furthermore, the manipulated variable U_{b4}, U_{b5} and U_{b6} have never saturated. So, there is no need of split-range combinations.

The addition information of relative orders is shown in Appendix Table 4.

Appendix Table 4 Relative orders of the MEN in the example 1

MV / CV	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}	U_{b6}
R1	∞	∞	5	6	3	2	4	1
R2	∞	∞	5	3	∞	4	1	2
L1	1	∞	2	3	5	4	6	∞
L2	∞	1	3	5	2	7	4	6

Then, the solver "CPLEX" of "GAMs" is used to solve the ILP problem. The results from solving the ILP problem are optimal split-range pairs and controllability. The values of binary variables $x_{i,j}$ and $z_{k,j}$ from solving problem are shown in Appendix Tables 5 and 6, respectively. In Appendix Table 5 the primary manipulated variables are Q_{L1}, Q_{L2}, $U_{b1,}$ $U_{b2,}$ $U_{b4,}$ U_{b5} and U_{b6} while the secondary manipulated variable is U_{b3} for U_{b1}. Appendix Table 6 shows the control pairing which are R1-U_{b1}, R2-U_{b5} , L1-Q_{L1} and L2-Q_{L2}.

Appendix Table 5 The value of $x_{i,j}$ after solving problem for the example 1

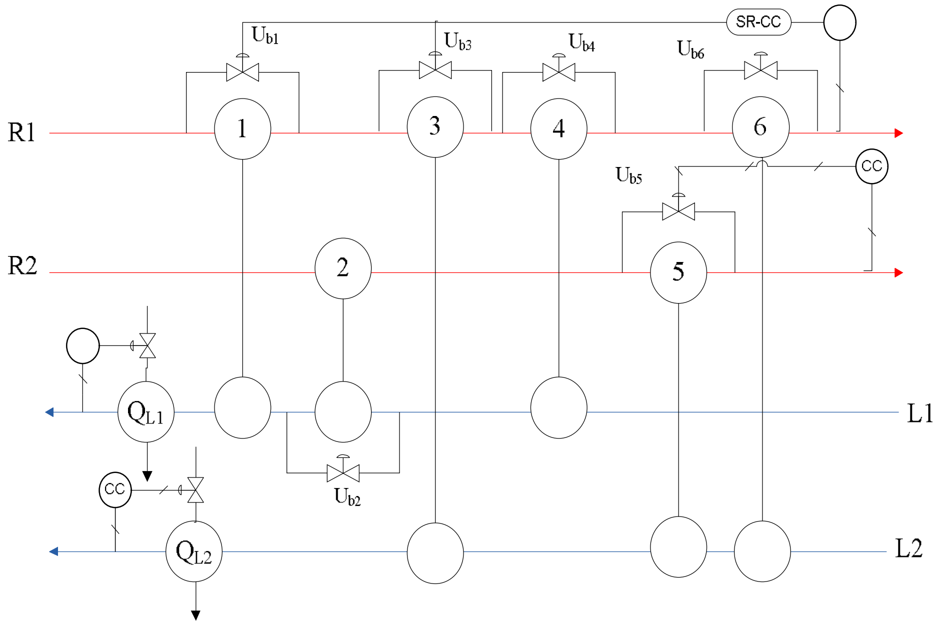
Primary MV \ Secondary MV	Q_{L1}	Q_{L2}	U_{b1}	U_{b2}	U_{b3}	U_{b4}	U_{b5}	U_{b6}
Q_{L1}	1							
Q_{L2}		1						
U_{b1}			1		1			
U_{b2}				1				
U_{b4}						1		
U_{b5}							1	
U_{b6}								1

(the remaining entries are zero)

Appendix Table 6 The value of $z_{k,j}$ after solving problem for the example 1

CV \ MV	Q_{L1}	Q_{L2}	U_{b1}	U_{b5}
R1			1	
R2				1
L1	1			
L2		1		

(the remaining entries are zero)



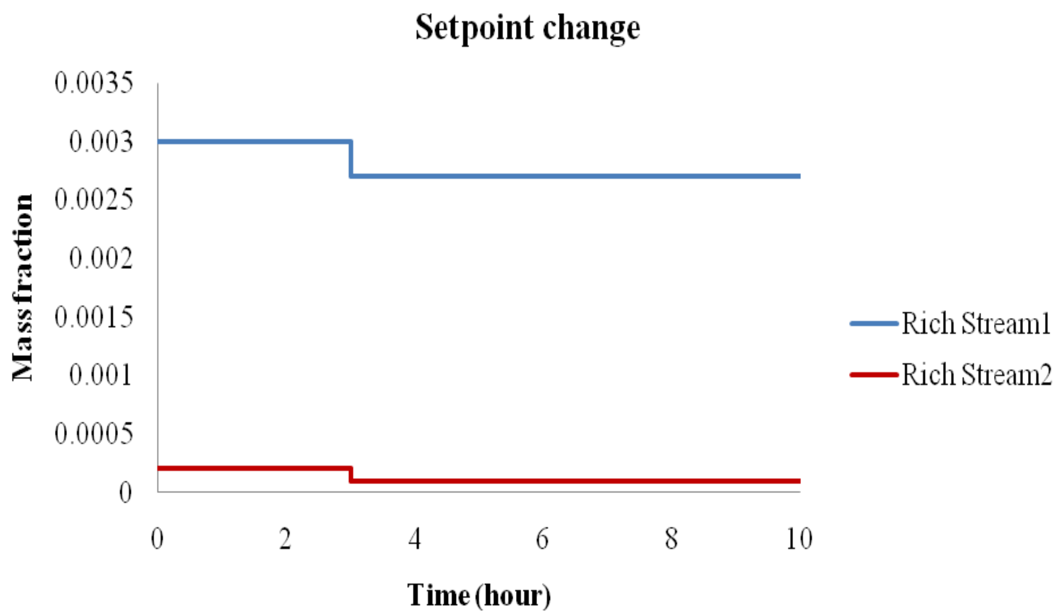
Appendix Figure 2 The control structure for the example 1

Split-range signal from controller can be obtained by referring to the information of active constraint regions in Appendix Table 3. The split-range signal of the pair of U_{b1} and U_{b3} can switch alternately to their lower constraints (SR-CC is split-range composition control).

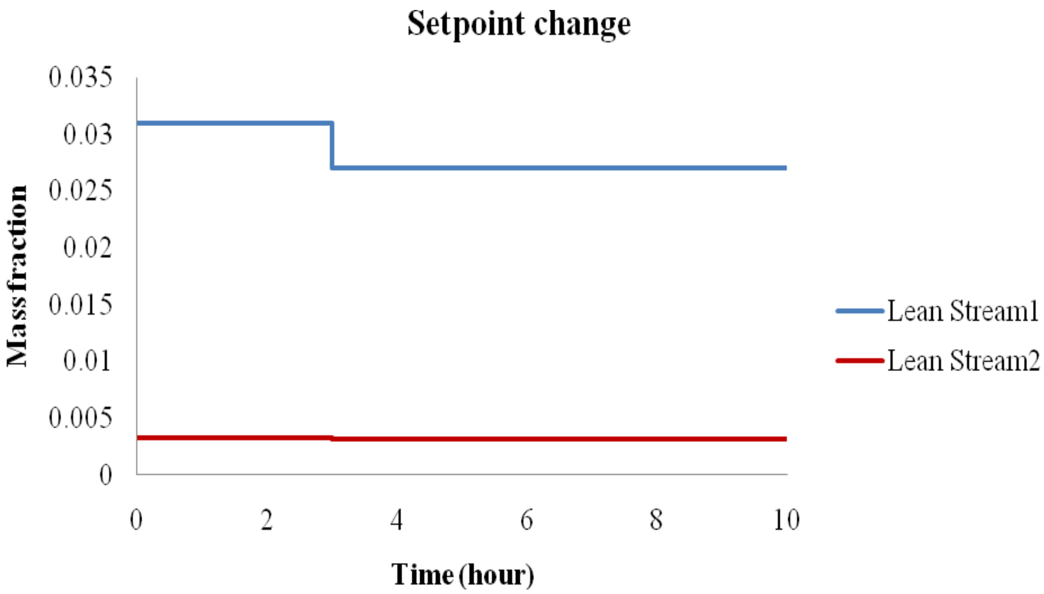
The result of control structures in MEN example 1 is tested by performing the dynamic simulation on Aspen Dynamics v2006.5. The information of disturbances and active constraints of the system in example 1 are shown in Appendix Table 7. The dynamic results show that the control structures can provide optimality. Appendix Figure 3 illustrate the dynamic result of MEN with the control structures and shows the control efficiency to mass fraction under disturbance. And Appendix Figure 3c shows the response of split-range control that U_{b1} is primary manipulate variable and U_{b3} is secondary manipulate variable to keep mass fraction of rich stream 1.

Appendix Table 7 Disturbances and active constraints in the example 1

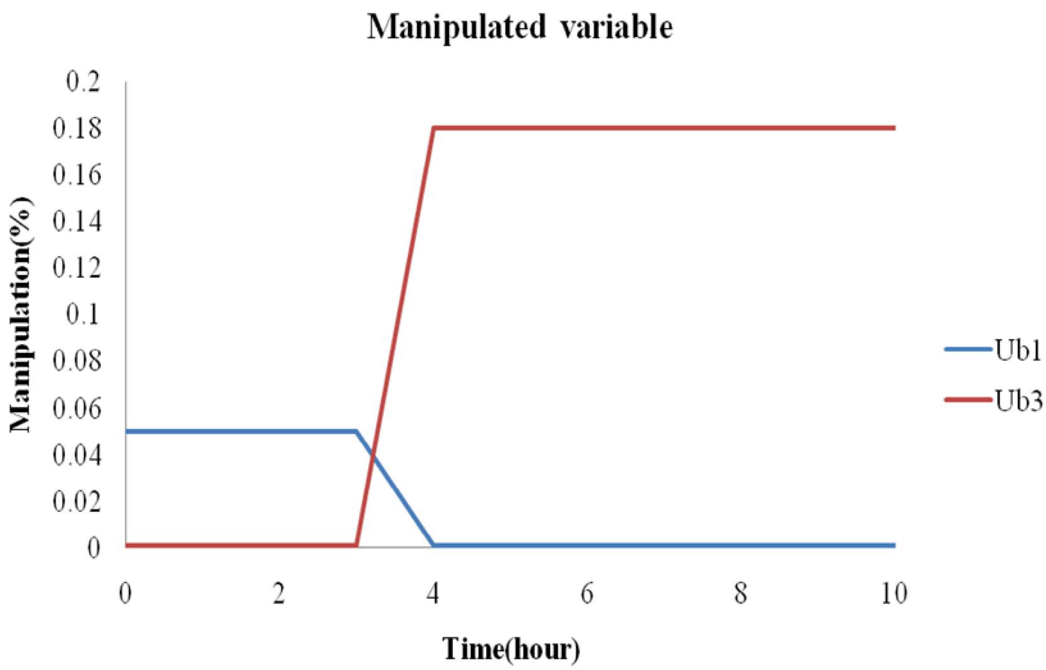
Time (hour)	Disturbance of mass fraction (set point change)				Active constraint	
	ΔR_1	ΔR_2	ΔL_1	ΔL_2	U_{b1}	U_{b3}
>3	0	0	0	0	S_L	
<3	-0.00003	-0.00001	-0.0031	-0.00035		S_L



a) setpoint change of R1 and R2



b) setpoint change of S1and S2



c) Manipulated variable (U_{b1} and U_{b3})

Appendix Figure 3 Dynamic simulation of the MEN in example 1

These Appendix Figures 3a and 3b illustrated the results of dynamic test while system of MEN disturbed within reduce 10% of mass fraction of rich streams and lean streams, respectively. And Appendix Figure 3c show the results of manipulated variable while MENs disturbed by reduce 10% of mass fraction. It can be seen that the manipulated variable; U_{b1} is primary manipulate variable switch to U_{b3} is secondary manipulated variable to adjust the lean stream 1 in order to obtain new setpoint.

CURRICULUM VITAE

NAME	: Mr. Pattarasak Sattarattanakul		
BIRTH DATE	: July 20, 1984		
BIRTH PLACE	: Bangkok, Thailand		
EDUCATION	: **YEAR**	**INSTITUTE**	**DEGREE**
	2007	Kasetsart Univ.	B.Eng (Chem Eng)
SCHOLARSHIP	: Department of Chemical Engineering, Kasetsart University		