Mass Exchange Network Synthesis for Single Component Problems

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Abstract

The paper deals with the synthesis problem of mass exchange networks (MEN's) for waste minimization by adopting a mathematical programming approach based on the stage-wise superstructure representation of the MEN's, analogous to the one introduced by Yee and Grossmann (1990a,1990b) for synthesis of heat exchange networks (HEN's). Not using any heuristics that are based on the concept of pinch points, the proposed superstructure-based representation for MEN's is formulated as a mixed-integer nonlinear programming (MINLP) optimization model, and therefore the operating cost for the external lean mass separating agents and the annualized equipment cost for exchange units can be minimized simultaneously. One numerical example is illustrated the applicability of proposed approach for synthesis of MEN's.

Introduction

Absorption, stripping, extraction, leaching, adsorption, and ion exchange—all are the indispensable mass exchange operations used in chemical industries A common nature of these mass exchange operations is the transfer of single or multiple components from a rich stream, usually process effluents containing valuable materials or undesirable contaminants, into a relatively lean mass separating agents (MSAs) to conduct the separations. The mass transfer of these components between different streams is usually executed by contacting the rich streams and MSAs in counter-currently direct-contacted mass exchange units.

Until recently, compared to the burgeoning investigations on the synthesis of heat exchange networks (HEN's), there are not many research reports have been presented to address the integration of mass exchange networks (MEN's), though the design of individual mass exchange unit is a well-established topic.

In the early development, a systematic two-staged procedure for the synthesis of cost-effective MEN's is firstly proposed by El-Halwagi and Manousiouthakis (1989). Therein the pinch points, the thermodynamic obstacles that limit the extent of mass transfer between the rich streams and MSAs, are identified and a preliminary network is generated to feature maximum mass exchange in the first stage of synthesis. The preliminary network is then improved in the second stage to develop a final cost effective configuration to satisfy the assigned exchange obligations. Thereafter, a linear transshipment model is also established for automatic synthesis of MEN's with single-component targets (El-Halwagi and Manousiouthakis, 1990a). This work is further extended in a later report to include networks for regeneration of recyclable lean streams (El-Halwagi and Manousiouthakis, 1990b). Therein a mixed-integer nonlinear program is formulated to obtain the minimum cost of mass-separating and regenerating agents. Then a mixed-integer linear program is provided to solve the configuration with minimum number of mass exchange units. The synthesis problem of reactive MEN's is also discussed by El-Halwagi and Srinivas (1992), where chemical as well as physical MSAs can be used to separate a certain species from a set of rich streams.

Recently, Hallale and Fraser (2000a-d) present a method in a series of papers for targeting the capital cost as well as the operating cost of a mass exchange network, and these costs are further combined to give total annual cost target. The design of MEN's to meet the targets is also discussed in detail. These papers demonstrate that, contrary to previous belief, using the minimum number of units does not necessarily lead to a minimum cost design (Hallale and Fraser, 2000a). The primary limitation of these sequential approaches, as pointed by Papalexandri, Pistikopoulos and Floudas (1994), is due to the inappropriate consideration of all cost factors and trade-offs.

In contrast to previous works that simplify the problem by decomposition based on the concept of pinch points, a hyperstructure-based representation of MEN's is proposed by Papalexandri et al. (1994). Therein the MEN synthesis problem is formulated as a mixed-integer nonlinear programming optimization problem with both network operating and investment costs being optimized simultaneously. The impact of simultaneously minimizing operating and investment costs to waste minimization problems are demonstrated via several examples. The formulation is somewhat intricate for many designers, however.

In this paper, a simple but general mathematical programming approach based on a stage-wise superstructure, a structure analogous to that for HEN syntheses problems introduced by Yee and Grossmann (1990), is presented for the synthesis of MEN's. The MEN synthesis problems can be formulated as a mixed-integer nonlinear programming (MINLP) model, where the operating cost from the use of external MSAs and the annualized equipment cost for exchange units are minimized simultaneously. One typical example from literature will be illustrated to demonstrate the efficacy of the proposed MEN synthesis method.

The Stage-wise Superstucture

The analogue of the pinch analyses for syntheses of mass and heat exchange networks has been emphasized by several authors (El-Halwagi and Manousiouthakis, 1989, 1990a, 1990b; Hallale and Fraser, 2000a-d). For modeling the synthesis problem of mass exchange networks via simultaneous optimization, we thus directly adapt the stage-wise superstructure of Yee and Grossmann (1990a,1990b) developed for heat exchange network synthesis in this study.

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 number of stages in series. Therein the outlets of exchange units from splits of a common stream are 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 1 shows an example of a stage-wise superstructure involving two rich and two lean streams. 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 a typical MEN's. 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 and Manousiouthakis, 1990b).

Model Formulation

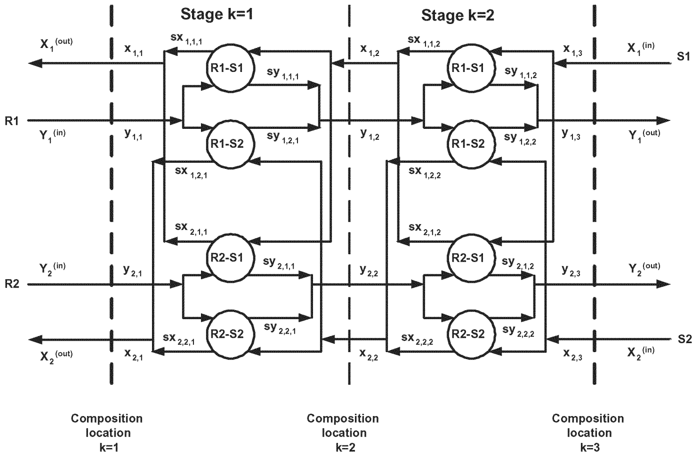

Figure 1: *Two-stage superstructure for mass exchange network synthesis*

In order to formulate the nonlinear model for synthesis of mass exchange networks, and for estimating the required mass separation area for packed towers or stage numbers for perforated-plate columns in the stage-wise superstructure, the following definitions are provided:

1. Indices:

i = rich process stream
j = lean stream or mass separation agent (MSA)
k = index for stage, $1, \ldots, N_S$, or composition location, $1, \ldots, N_S + 1$

2. Sets:

$$\begin{aligned}
\text{RP} &= \{i | i \text{ is a rich process stream}, i = 1, \ldots, N_R\} \\
\text{LP} &= \{j | j \text{ is a lean stream or MSA}, j = 1, \ldots, N_L\} \\
\text{LP}^{(\text{t})} &= \{j | j \text{ a lean stream or MSA using tray column}\} \\
\text{LP}^{(\text{h})} &= \{j | j \text{ a lean stream or MSA using packed col}\} \\
\text{ST} &= \{k | k \text{ is a stage}, k = 1, \ldots, N_S\}
\end{aligned}$$

3. Parameters:

AC_j = annual operating cost of lean stream j
$\text{AC}_{ij}^{(\text{t})}$ = per stage annual cost of tray column for i rich and j lean match
$\text{AC}_{ij}^{(\text{h})}$ = 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
$K_y a$ = overall mass transfer coefficient
$L_j^{(\text{up})}$ = 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, Γ = large positive values

$X_j^{(\text{in})}$ = inlet composition of lean stream j

$X_j^{(\text{out})}$ = outlet composition of lean stream j

$X_j^{(\text{up})}$ = upper bound composition of lean stream j

$Y_i^{(\text{in})}$ = inlet composition of rich stream i

$Y_i^{(\text{out})}$ = outlet composition of rich stream i

$Y_i^{(\text{up})}$ = upper bound composition of rich stream i

ε_{ij} = minimum composition difference between rich stream i and lean stream j

4. Variables:

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

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

ℓ_{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

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

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

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

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

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

With these definitions, the stage-wise superstructure for mass exchange networks can now be presented.

Overall mass balance for transferable component over the whole network:
An overall mass balance is needed to ensure sufficient exchange of the transferred component for all rich streams. The constraints specify that the overall mass transferable requirement of each rich stream for the transferred component must be equal to the sum of the component which exchanged with other lean process streams or MSAs at each stage.

$$
\begin{aligned}
(y_{i1} - y_{i,N_S+1})G_i &= \sum_{\forall k \in \text{ST}} \sum_{\forall j \in \text{LP}} M_{ijk} \quad \forall\, i \in \text{RP} \\
(x_{j1} - x_{j,N_S+1})L_j &= \sum_{\forall k \in \text{ST}} \sum_{\forall i \in \text{RP}} M_{ijk} \quad \forall\, j \in \text{LP}
\end{aligned} \tag{1}
$$

Mass balance in each stage:
A mass balance is also needed in each stage for each stream to determine the composition of each transferable component as well as partition of flow rates for all parallel units. For a superstructure with N_S stages, $N_S + 1$ levels of composition are involved. Note that the index k is used to represent the stage and the composition location in the superstructure. For both cases, stage or composition location $k = 1$ involves the highest compositions. The component and total mass balance for each stream in each stage are as follows.

$$
\begin{aligned}
(y_{ik} - y_{i,k+1})\, G_i &= \sum_{\forall j \in \text{LP}} M_{ijk} \quad \forall\, i \in \text{RP}, k \in \text{ST} \\
(x_{jk} - x_{j,k+1})\, L_j &= \sum_{\forall i \in \text{RP}} M_{ijk} \quad \forall\, j \in \text{LP}, k \in \text{ST} \\
G_i &= \sum_{\forall j \in \text{LP}} g_{ijk} \quad \forall\, i \in \text{RP}, k \in \text{ST} \\
L_j &= \sum_{\forall i \in \text{RP}} \ell_{ijk} \quad \forall\, j \in \text{LP}, k \in \text{ST}
\end{aligned} \tag{2}
$$

Mass balance in each exchanger:
A component mass balance is needed for each local exchange unit. The new variables include split mass flow rates, g_{ijk} and ℓ_{ijk}, and the transferred composition, sy_{ijk} and sx_{ijk}, before mixers, as illustrated in Figure 1. There is no need of extra composition variables for rich as well as lean streams in the splitters, such as stated in the following.

$$
\begin{aligned}
g_{ijk}\,(y_{ik} - sy_{ijk}) &= M_{ijk} \quad \forall\, i \in \text{RP}, j \in \text{LP}, k \in \text{ST} \\
\ell_{ijk}\,(sx_{ijk} - x_{j,k+1}) &= M_{ijk} \quad \forall\, i \in \text{RP}, j \in \text{LP}, k \in \text{ST}
\end{aligned} \tag{3}
$$

Notice that the balance equations around all mixers from split streams are not necessary since these equations are redundant to Eqs. (2)∼(1) (Bjork and Westerlund, 2002).

Assignment of superstructure inlet/outlet compositions:
The given inlet/outlet compositions of rich and lean process 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 $k = N_S + 1$.

$$
\begin{aligned}
Y_i^{(\text{in})} &= y_{i1} \qquad Y_i^{(\text{out})} = y_{i,N_S+1} \quad \forall\, i \in \text{RP} \\
X_j^{(\text{in})} &= x_{j,N_S+1} \qquad X_j^{(\text{out})} = x_{j1} \quad \forall\, j \in \text{LP}
\end{aligned} \tag{4}
$$

Feasibility of the transferable component:
Constraints are also needed to guarantee monotonic decrease of the transferred composition at successive stages.

$$
\begin{aligned}
y_{ik} &\geq y_{i,k+1} \quad \forall\, i \in \text{RP}, k \in \text{ST} \\
x_{jk} &\geq x_{j,k+1} \quad \forall\, j \in \text{LP}, k \in \text{ST}
\end{aligned} \tag{5}
$$

Logical constraints:
Logical constraints and binary variables, z_{ijk}, are needed to determine the existence or absence of process matches (i, j) in stage k. An integer value of one for binary variable z_{ijk} designates that match between rich stream i and lean stream j in stage k is present in the optimal network. The constraints are as follows where U is a upper bound for mass exchanged

$$
\begin{aligned}
M_{ijk} - U z_{ijk} &\leq 0 \quad \forall\, i \in \text{RP}, j \in \text{LP}, k \in \text{ST} \\
z_{ijk} &\in \{0, 1\}
\end{aligned} \tag{6}
$$

Feasibility constraints of the equilibrium relationships:
The feasibility constraints of the equilibrium relationships ensure positive driving forces for the potential process exchange units. Binary variables are used for these constraints to ensure that only the non-negative driving forces exist for existing matches where the associated binary variables all equal one. If a match does not occur, the associated binary variable equals zero and the large positive upper bound Γ can deem the equation redundant. In these equations, a streams-and-component dependent minimum composition approach ε_{ij} is also chosen so that feasible mass transfer in a finite number of equilibrium stages or finite area can be achieved in each transfer unit.

$$\begin{aligned} dyx_{ijk} &\le y_{ik} - m_{ij}sx_{ijk} - b_{ij} + \Gamma(1 - z_{ijk}) \\ &\qquad \forall\, i \in \text{RP}, j \in \text{LP}, k \in \text{ST} \\ dyx_{ij,k+1} &\le sy_{ijk} - m_{ij}x_{j,k+1} - b_{ij} + \Gamma(1 - z_{ijk}) \\ &\qquad \forall\, i \in \text{RP}, j \in \text{LP}, k \in \text{ST} \\ dyx_{ijk} &\ge m_{ij}\varepsilon_{ij} \\ &\qquad \forall\, i \in \text{RP}, j \in \text{LP}, k \in \text{ST} \cup \{N_S + 1\} \end{aligned} \tag{7}$$

Bounds on variables:
Bounds are set on the mass flow rate of lean streams, their final compositions and on the final compositions of rich streams, when they are not fixed.

$$\begin{array}{lll} L_j \le L_j^{(\text{up})} & x_{j1} \le X_j^{(\text{up})} & \forall\, j \in \text{LP} \\ & y_{i,N_S+1} \le Y_i^{(\text{up})} & \forall\, i \in \text{RP} \end{array} \tag{8}$$

Optional constraints:
Some additional constraints such as no stream splits, forbidden matches, and required and restricted matches can be easily included in this model. For example, the stream splitting can be prevented by constraining the number of matches for split streams in each stage, such as,

$$\begin{array}{lll} \sum\limits_{\forall i \in \text{RP}} z_{ijk} \le 1 & \forall\, j \in \text{LP}, k \in \text{ST} \\ \sum\limits_{\forall j \in \text{LP}} z_{ijk} \le 1 & \forall\, i \in \text{RP}, k \in \text{ST} \end{array} \tag{9}$$

The maximum total number of mass exchange units can be limited by adding an upper bound for selected exchangers, MEU, in the following constraint.

$$\sum_{\forall i \in \text{RP}} \sum_{\forall j \in \text{LP}} \sum_{\forall k \in \text{ST}} z_{ijk} \le \text{MEU} \tag{10}$$

Other restrictions can also be considered by assigning suitable values for specific integer variables. For example, should the match in between rich stream $i = 1$ and lean stream $j = 2$ is not allowable, then one can assign $z_{12k} = 0\, \forall k \in \text{ST}$.

Sizing equations for mass transfer units:
Mass exchangers can be classified into two main categories: stagewise exchangers and continuous-contact exchangers. The most common types of stagewise exchangers are tray or plate columns. When mass exchange takes place in a tray column, the number of required stages can be determined from the Kremser equation. The traditional form for the Kremser equation can be expressed as follows should the operating and equilibrium lines are both straight (Treybal, 1981; Szitkai et al., 2002; Shenoy and Fraser, 2003):

$$N_{ijk} = \begin{cases} \dfrac{\ln\left[\left(\dfrac{y_{ik} - y^*_{ij,k+1}}{sy_{ijk} - y^*_{ij,k+1}}\right)\left(1 - \dfrac{m_{ij}g_{ijk}}{\ell_{ijk}}\right) + \dfrac{m_{ij}g_{ijk}}{\ell_{ijk}}\right]}{\ln\left[\dfrac{\ell_{ijk}}{m_{ij}g_{ijk}}\right]} & \text{for } \dfrac{\ell_{ijk}}{m_{ij}g_{ijk}} \ne 1 \\ \dfrac{y_{ik} - sy_{ijk}}{sy_{ijk} - y^*_{ij,k+1}} & \text{for } \dfrac{\ell_{ijk}}{m_{ij}g_{ijk}} = 1 \end{cases} \tag{11}$$

Where, $y^*_{ij,k+1} = m_{ij}x_{j,k+1} + b_{ij}$ and $sy^*_{ijk} = m_{ij}sx_{ijk} + b_{ij}$ are equilibrium compositions. The linear equilibrium relation and the material balance equation can be further substituted into Eq.(11) to give the following alternative from for the Kremser equation (Shenoy and Fraser, 2003):

$$N_{ijk} = \begin{cases} \dfrac{\text{L}_{\text{og}}\text{M}_{\text{ean}}\left[y_{ik} - sy_{ijk}, sy^*_{ijk} - y^*_{ij,k+1}\right]}{\text{L}_{\text{og}}\text{M}_{\text{ean}}\left[y_{ik} - sy^*_{ijk}, sy_{ijk} - y^*_{ij,k+1}\right]} & \text{for } \dfrac{\ell_{ijk}}{m_{ij}g_{ijk}} \ne 1 \\ \dfrac{y_{ik} - sy_{ijk}}{y_{ik} - sy^*_{ijk}} = \dfrac{y_{ik} - sy_{ijk}}{sy_{ijk} - y^*_{ij,k+1}} & \text{for } \dfrac{\ell_{ijk}}{m_{ij}g_{ijk}} = 1 \end{cases} \tag{12}$$

Notably the Log-mean Kremser representation for case of $\dfrac{\ell_{ijk}}{m_{ij}g_{ijk}} \ne 1$ will be equivalent to the second one for $\frac{\ell_{ijk}}{m_{ij}g_{ijk}} = 1$ case. However, Eq.(12) will still lead to numerical difficulties for some zero values in the log-mean. Thus an approximation for the composition difference term is required to avoid numerical problem when the approach compositions of both sides of the mass exchange unit are equal (Yee and Grossmann, 1990a,1990b; Shenoy and Fraser, 2003). Here, the Chen's approximation is used (Chen, 1987), as shown in the following:

$$\begin{aligned} &\text{L}_{\text{og}}\text{M}_{\text{ean}}\left[y_{ik} - sy_{ijk}, sy^*_{ijk} - y^*_{ij,k+1}\right] \\ &= \frac{(y_{ik} - sy_{ijk}) - (sy^*_{ijk} - y^*_{ij,k+1})}{\ln\left[\dfrac{y_{ik} - sy_{ijk}}{sy^*_{ijk} - y^*_{ij,k+1}}\right]} \\ &\cong \left[(y_{ik} - sy_{ijk})(sy^*_{ijk} - y^*_{ij,k+1}) \frac{(y_{ik} - sy_{ijk} + sy^*_{ijk} - y^*_{ij,k+1})}{2}\right]^{1/3} \end{aligned} \tag{13}$$

Thus the log-mean Kremser equation for stage numbers, Eq.(12), can be re-formulated as follows:

$$N_{ijk} \cong \left[\frac{(y_{ik} - sy_{ijk})\left(sy^*_{ijk} - y^*_{ij,k+1}\right)}{\left(y_{ik} - sy^*_{ijk}\right)\left(sy_{ijk} - y^*_{ij,k+1}\right)} \frac{\left(y_{ik} - sy_{ijk} + sy^*_{ijk} - y^*_{ij,k+1}\right)}{\left(y_{ik} - sy^*_{ijk} + sy_{ijk} - y^*_{ij,k+1}\right)} \right]^{1/3} \quad \forall \ \frac{\ell_{ijk}}{m_{ij} g_{ijk}} \tag{14}$$

When absorption or stripping takes place, a continuous-contact packed tower is suggested for mass exchange. The required packed height for (i, j) match in stage k, H_{ijk}, is characterized by a number of imaginary transfer units, NTU_{ijk}, and the overall height of a transfer unit, HTU_{ijk} (Treybal, 1981; Hallale and Fraser, 2000b). Therein calculations are based on the conditions in the rich stream. The overall packed height is given by the following equation, where the log-mean calculation is also applying Chen's approximation.

$$\begin{aligned} \text{HTU}_{ijk} &= \frac{g_{ijk}}{K_y a S} \\ \text{NTU}_{ijk} &= \frac{y_{ik} - sy_{ijk}}{\text{L}_{\text{og}}\text{M}_{\text{ean}}\left[y_{ik} - sy^*_{ijk}, sy_{ijk} - y^*_{ij,k+1}\right]} \\ H_{ijk} &= \text{HTU}_{ijk} \times \text{NTU}_{ijk} \\ &= \frac{M_{ijk}}{K_y a S} \times \frac{1}{\text{L}_{\text{og}}\text{M}_{\text{ean}}\left[y_{ik} - sy^*_{ijk}, sy_{ijk} - y^*_{ij,k+1}\right]} \\ &= \frac{M_{ijk}}{K_y a S} \times \left[\left(y_{ik} - sy^*_{ijk}\right)\left(sy_{ijk} - y^*_{ij,k+1}\right) \frac{\left(y_{ik} - sy^*_{ijk} + sy_{ijk} - y^*_{ij,k+1}\right)}{2} \right]^{-1/3} \end{aligned} \tag{15}$$

Objective function:
The objective function of the proposed model simultaneously includes utilities, mass-exchange unit reassignment, new mass-exchange unit installation, and the additional tray/height cost, such as stated in the following, where $\boldsymbol{x}$ and $\boldsymbol{\Omega}$ denote the design variables and the feasible space defined by all constraints, Eqs. (1)∼(8) and Eqs. (14)∼(15), respectively.

$$\begin{aligned} \min_{\boldsymbol{x} \in \boldsymbol{\Omega}} \ \text{TAC} &= \sum_{\forall j \in \text{LP}} \text{AC}_j L_j \\ &+ \sum_{\forall i \in \text{RP}} \sum_{\forall j \in \text{LP}^{(\text{t})}} \sum_{\forall k \in \text{ST}} \text{AC}^{(\text{t})}_{ij} N_{ijk} \\ &+ \sum_{\forall i \in \text{RP}} \sum_{\forall j \in \text{LP}^{(\text{h})}} \sum_{\forall k \in \text{ST}} \text{AC}^{(\text{h})}_{ij} H_{ijk} \end{aligned} \tag{16}$$

Numerical Example

The example, studied by El-Halwagi and Manousiouthakis (1990a), involves the removal of a single component, the copper, from an ammoniacal etching solution and a rinsewater stream. The cost data used in Papalexandri et al. (1994), a modification from the original source of El-Halwagi and his co-workers, are applied for fair comparison, as shown in Table 1. Notably, a 1 meter column diameter for all tray columns and packed towers has been assumed in this work. Thus the investment cost is proportional to the tray number or tower height only.

Table 1: Capital cost data for example

Plate-column	$4552\,N$ \$/yr
Packed column	$4245\,H$ \$/yr

Table 2: Stream data for Example 1

Rich stream	Description	G_i (kg/s)	$Y_i^{(\text{in})}$ (mass	$Y_i^{(\text{out})}$ fraction)	
R_1	ammon. soln	0.25	0.13	0.10	
R_2	rinsewater	0.10	0.06	0.02	
MASs	**Description**	$L_j^{(\text{up})}$ (kg/s)	$X_j^{(\text{in})}$ (mass	$X_j^{(\text{out})}$ fraction)	cost (\$s/kg yr)
S_1	LIX63	∞	0.03	0.07	$58,680$
S_2	P_1	∞	.001	0.02	$704,160$

Etching of copper is achieved through ammoniacal solution, where the etching efficiency is higher for copper compositions between 10 and 13 w/w%. The copper contaminant is continuously removed from the ammoniacal solution, R_1, via solvent extraction and the regenerated etchant is returned to the etching line. Meanwhile the etched printed circuit boards are washed out with water, and the effluent rinse water, R_2 is also decontaminated by extraction and then recycled to the rinse vessel. A schematic representation of the etching process can be found in El-Halwagi and Manousiouthakis (1990a).

Two external MSAs are available for removal of copper: LIX63 (an aliphatic α-hydroxyoxime, S_1) and P_1 (an aromatic β-hydroxyoxime, S_2). The cost data listed in Table 2 come from El-Halwagi & Manousiouthakis (1990a) and Papalexandri et al. (1994), respectively. Noted that there are two annual operating cost data. Papalexandri et al. (1994) found that when using the El-Halwagi and Manousiouthakis's original operating cost, it will lead the investment cost to a relatively small value, and cause the unrealistic percentage of the overall cost. Therefore, Papalexandri et al. (1994) discussed the MEN synthesis problems with a reduced annual operating cost by dividing the operating cost by a factor of five for depreciation and furthermore by reducing the total operating hours from $8,760$ to $8,150$ hours/year. In order to compare with the hyperstructure based optimization solutions of Papalexandri et al. (1994), we adopt the modified cost data in the following discussion. 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 lean streams, respectively (El-Halwagi and

Manousiouthakis, 1990a):

$$\begin{aligned}(R_1, S_1): &\quad y_1 = 0.734x_1 + 0.001\\ (R_2, S_1): &\quad y_2 = 0.734x_1 + 0.001\\ (R_1, S_2): &\quad y_1 = 0.111x_2 + 0.008\\ (R_2, S_2): &\quad y_2 = 0.148x_2 + 0.013\end{aligned}$$

Plate-columns with 1 meter diameter can be used for S_1 while packed columns with 1 meter diameter are suitable for S_2. In order to allow a comparison with the results of Papalexandri et al. (1994), the mass transfer coefficients, K_ya, for rich streams were calculated using the correlations given by El-Halwagi and Manousiouthakis (1990a). that is due to lack of data in Papalexandri et al. (1994). The cost data were originally given for columns with 2 meter diameters and were corrected for the 1 meter diameter columns. The K_ya values are 0.685 kg copper m^{-3}/s for R_1 and 0.211 kg copper m^{-3}/s for R_2 (Hallale and Fraser, 2000d). A minimum composition different of $\varepsilon = 0.0001$ determines feasible mass exchange at the inlet and outlet of all potential mass exchange units.

The proposed MINLP synthesis model is employed to determine a total annual cost with optimal mass exchange network for copper recovery. Operating and annual investment cost are considered simultaneously. With the minimal possible stage number for the superstructure, $N_S = \max\{N_R, N_L\} = 2$, the proposed superstructure based MINLP formulation can result in the same network structure of El-Halwagi and Manousiouthakis (1990a), as shown in Figure 2. In the figure, numerical value in parenthesis denotes the mass transfer load of the exchange unit, and other values denote compositions and/or flow rates, respectively. The flow rates of S_1 and S_2 are 0.276 kg/s and 0.023 kg/s, respectively. This network features a total annual cost of $\$52,300$/yr, out of which $\$32,700$/yr is the cost of the mass separating agents. However, the total annual cost is increased from $\$49,000$ /yr to $\$52,300$/yr (a 6.7% increase) when compared to the solution of Papalexandri et al. (1994) via the hyperstructure based optimization. It can be found that the two-staged superstructure is impossible to support the structure of Papalexandri et al. (1994) unless we increase the stage numbers. When adopting $N_S = 3$, we obtain the same structure of Papalexandri et al. (1994) with the total annual cost of $\$46,000$ /yr (a 5.6% reduction), as shown in Figure 3.

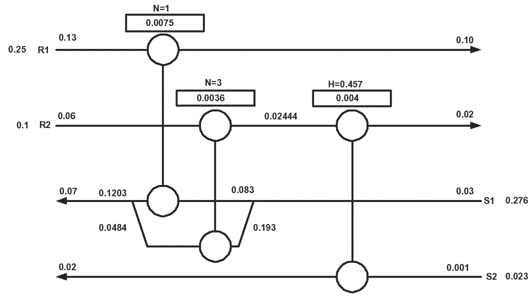

Figure 2: *The resulting MEN structures of example using two stages in the proposed superstructure*

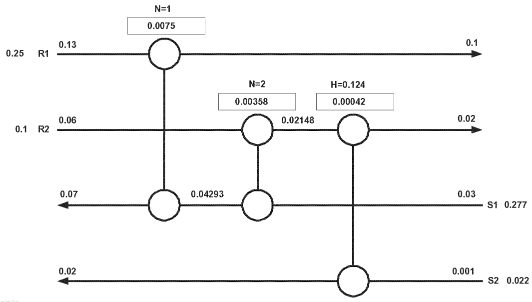

Figure 3: *The resulting MEN structures of example using three stages in the proposed superstructure*

Conclusion

Although the synthesis for mass exchange networks is no less important than that of heat exchange networks, the amount and broadness and depth of research devoting to this problem is far less than that of HEN's. To narrow the gap between these two kinds of synthesis, this article devotes itself to the synthesis problem of MEN's by proposing a mathematical programming approach based on the stage-wise superstructure representation of the MEN's, which can directly handle multiple transferable species and reactive mass separating agents, and can also be easily extended to consider networks for regeneration of recycled lean streams. This representation for MEN's skips any use of heuristics that are based on the concept of pinch points and thus make the MEN synthesis problem a mixed-integer nonlinear programming (MINLP) optimization problem. By taking this approach, the operating cost of consuming the external lean mass separating agents as well as the regenerating agents and the annualized equipment cost of mass exchange units are minimized simultaneously. One typical example from literature will be illustrated to demonstrate the efficacy of the proposed MEN synthesis method.

References

Bjork, K. M., & Westerlund, T. (2002). Global optimization of heat exchanger network synthesis problems with and without the isothermal mixing assumption. *Comp. Chem. Eng.*, **26**, 1581.

El-Halwagi, M. M. & Manousiouthakis, V. (1989). Synthesis of mass exchange networks. *AIChE J.*, **35**, 1233.

El-Halwagi, M. M., & Manousiouthakis, V. (1990a). Automatic synthesis of mass exchange networks with single component targets. *Chem. Eng. Sci.*, **45**, 2813.

El-Halwagi, M. M., & Manousiouthakis, V. (1990b). Simultaneous synthesis of mass-exchange and regeneration networks. *AIChE J.*, **36**, 1209.

El-Halwagi, M. M., & Srinivas, K. (1992). Synthesis of reactive mass-exchange networks. *Chem. Eng. Sci.*, **47**, 2113.

Hallale, N., & Fraser, D. M. (2000a). Capital and total cost targets for mass exchange networks. Part 1: Simple capital cost models. *Comp. Chem. Eng.*, **23**, 1661.

Hallale, N., & Fraser, D. M. (2000b). Capital and total cost targets for mass exchange networks. Part 2: Detailed capital cost models. *Comp. Chem. Eng.*, **23**, 1681.

Hallale, N., & Fraser, D. M. (2000c). Supertargeting for mass exchange networks. Part I: Targeting and design techniques. *Trans. IChemE*, **78**, 202.

Hallale, N., & Fraser, D. M. (2000d). Supertargeting for mass exchange networks. Part II: Applications. *Trans. IChemE*, **78**, 208.

Papalexandri, K. P., Pistikopoulos, E. N., & Floudas, A. (1994). Mass exchange networks for waste minimization: a simultaneous approach. *Trans. IChemE*, **72**, 279.

Szitkai,Z., Lelkes Z., Rev, E. & Fonyo, Z. (2002). "Handling of removable discontinuities in MINLP models for process synthesis problems, formulations of the Kremser equation. *Comp. Chem. Eng.*, **26**, 1501.

Shenoy, U. V., & Fraser, D. M. (2003). A new formulation of the Kremser equation for sizing mass exchangers. *Chem. Eng. Sci.*, **58**, 5121.

Treybal, R. E. (1981). Mass Transfer Operations, 3rd ed., McGraw-Hill, Singapore.

Yee, T. F., & Grossmann, I. E. (1990). Simultaneous optimization models for heat integration-II. Heat exchanger network synthesis. *Comp. Chem. Eng.*, **14**, 1165.