

Separation Network Design with Mass and Energy Separating Agents

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Abstract

The mathematical model developed in this paper deals with simultaneous synthesis of the integrated separation network, where both mass separating agents (MSAs) and energy separating agents (ESAs) are taken into account. The proposed model formulation is believed to be superior to the available ones. Traditionally, the tasks of optimizing ESA-based and MSA-based processes were either performed individually or studied on a heuristic basis. In this work, both kinds of processes are incorporated into a single comprehensive flowsheet and a novel state-space superstructure with multi-stream mixings is adopted to capture all possible network configurations. By properly addressing the issue of interactions between the MSA and ESA subsystems, lower total annualized cost (TAC) can be obtained by solving the corresponding mixed-integer nonlinear programming (MINLP) model. A benchmark problem already published in the literature has been investigated to demonstrate how better conceptual designs can be generated by our proposed approach.

Keywords: simultaneous synthesis, process integration, separation network design, state-space superstructure, MINLP model

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Introduction

Separation operations, which transform chemical mixtures into new mixtures and/or essentially pure components, are of central importance in process industries. Separations involve different modes and one way of classifying these separation processes is based on the nature of separating agents, which take the form of MSAs and ESAs. Typical ESA and MSA processes, especially distillation sequences and the mass exchange networks (MEN), have been the subject of extensive investigations due to the significant capital and operating costs associated with such processes.

In the past decades, a number of approaches have been proposed for the systematic synthesis of distillation sequences, including heuristic methods (Seader and Westerberg, 1977), evolutionary techniques (Stephanopoulos and Westerberg, 1976), hierarchical decomposition (Douglas, 1998), explicit and implicit enumerations (Chavez et al., 1986; Fraga and McKinnon, 1995), stochastic methods (Fraga and Matias, 1996; Wang et al., 2008; An and Yuan, 2009), matrix based methods (Ivakkpou and Kasiri, 2009; Shah and Agrawal, 2009), temperature collocation approaches (Zhang and Linninger, 2004; Ruiz et al., 2010) and superstructure based optimization (Andrecovich and Westerberg, 1985; Floudas and Paules, 1988; Bagajewicz and Manousiouthakis, 1992; Yeomans and Grossmann, 2000a; Yeomans and Grossmann, 2000b; Caballero and Grossmann, 2001; Caballero and Grossmann, 2004; Proios and Pistikopoulos, 2005). As observed by Yeomans and Grossmann (1999), while there are relative merits and shortcomings of these different approaches, superstructure optimization can provide a systematic framework with which the various subsystems can be simultaneously optimized and interconnected in a natural way. For instance, a superstructure optimization model for distillation can be readily incorporated as part of the optimization of a process flowsheet. As we will demonstrate later in this paper, our research fully takes such advantage and is

specifically aimed at addressing the formulation and interactions of distillation system with MEN design.

Compared to the extensive investigations on distillation sequences, it was not until late 1980s that pollution preventions and economic considerations had drawn attention to a more specialized separation problem, MEN synthesis. In the early development, a systematic sequential procedure that can synthesize MEN was first proposed by El-Halwagi and Manousiouthakis (1989). In this work, preliminary network featuring maximum mass exchange was generated and then improved to obtain a final cost effective configuration which satisfies the assigned exchange duty. Later, El-Halwagi and Manousiouthakis (1990a) introduced the linear transshipment model (Papoulias and Grossmann, 1983) to the synthesis of MEN with single-component targets. Their work was further developed to incorporate the associate mass-exchange regeneration networks which deal with lean stream recycling (El-Halwagi and Manousiouthakis, 1990b). In recent studies, Hallale and Fraser (2000a, b, c, d) presented a series of papers for targeting the capital and operating cost estimates with simple approximations when calculating annualized costs. Apart from the aforementioned sequential procedures based on pinch technique, mathematical optimization techniques for MEN have been used to handle more complex trade-offs of all cost factors. Papalexandri et al. (1994) first developed an MINLP model based on a hyperstructure representation. The two-way balance between operating cost and investment cost was explored and this model was also extended to include regeneration networks. However, Papalexandri and his co-workers failed to capture the optimal solution due to the limited capability of their solution strategy. In later studies, the stage-wise superstructure proposed by Yee and Grossmann (1990a,b) has been widely used in MEN design. Chen and Hung, (2005a) and Szitkai et al. (2006) respectively presented a mathematical programming approach based on the

stage-wise representation of the MEN. Then, Isafiade and Fraser (2008) proposed the interval based MINLP superstructure (IBMS) on mass and regeneration network design. Recently, this work was extended to handle split streams going through two or more exchangers in series (Isafiade and Fraser, 2010). Although better and cost-effective designs can almost always be obtained, by assuming mixing within stages these methods may preclude a class of good solutions, where the optimal solution may actually lie.

In the aforementioned studies, the tasks of synthesizing distillation sequences and MEN were examined individually. However, as separation tasks are performed with the aid of either separating agent, or combinations thereof, there is a need for considering simultaneously both MSAs and ESAs for the conceptual design. The earliest attempts to synthesize separation systems which involve the use of both MSAs and ESAs date back to the early 1970s. Thompson and King (1972) proposed the product separability matrix and used heuristic and algorithmic programming to determine key components, type and order of separation, as well as the MSAs to be used. Based on the results of the previous synthesis, the entire synthesis is repeated several times in order to obtain better cost estimates. Later, Nath and Motard (1981) devised a systematic way to choose the product set, the key component, the type of separator as well as MSAs with the help of a heuristic evaluation function. Despite considerable contributions accomplished by these synthesis methods, all these heuristic procedures have a common and serious limitation: they have not addressed the problem of minimizing the TAC which is subjected to the thermodynamic constraints. By overlooking such phase equilibrium relations, the proposed methods may generate separation networks which are thermodynamically infeasible. In addition, the economic optimality of these resulting networks cannot be guaranteed because of the inability of these procedures to consider the optimal design of

each separator (such as the purity, number of equilibrium stages and/or reflux ratio) and the balances of all cost items. These limitations can be mitigated by our methods introduced below.

In this work, a general mathematical programming model based on state-space superstructure, a framework that takes all stream mixing possibilities into consideration, is presented for the design of separation network with both MSAs and ESAs. Considering the thermodynamic constraints as well as relevant shortcut models for each type of separator, the overall synthesis problem can be formulated as an MINLP model, where the operating costs (including costs of process and external MSAs, regenerating agents, cold and hot utilities) and equipment cost (including costs of mass exchange units and distillation columns) are minimized simultaneously. Since (1) the state-space representation does not contain any simplifying assumptions of the network topologies, and (2) the trade-offs between capital and operating costs, between ESA and MSA costs can be properly carried out, it is reasonable to expect that the TAC of the overall separation network can be reduced. To describe the design method developed in this work and its applications, the rest of this paper is organized as follows. The separation network design problem is formally defined in the next section. All issues pertaining to the modified state-space representation and the corresponding MINLP model are described in Section 3. Four examples are then presented in Section 4 to demonstrate the effectiveness of the proposed method and the conclusions of this research are provided in the last section.

Problem Statement

To facilitate the concise formulation of the mathematical model, several important assumptions are made to simplify this problem: the mass flow rates of all streams remain unchanged throughout the network; the equilibrium relation governing the transferable component is linear and independent

of other components; the mass exchange units are of the counter-current type; ESAs are only used for solvent regeneration; the constant molar overflow is adopted in the column design; the feed to the distillation column is assumed to remain at its bubble point and heat integration between streams is not considered.

The separation network design problem addressed in this work can be stated as follows: Given a set of rich process streams, a set of lean streams (process and external MSAs), a set of mass regenerating agents and ESAs for solvent recycling, it is desired to synthesize a cost-optimal separation network that can fulfill the separation requirements of all streams and also satisfy the energy and composition requirements imposed at various locations in the network. More specifically, the given model parameters of this optimization problem include: (1) the process data of every rich process stream (i.e., its flow rate, components and the inlet and outlet compositions), (2) the process data of every lean stream (i.e., its flow rate, cost, components and the inlet and/or outlet compositions), (3) the process data of the regenerating agents (i.e., its flow rate, cost, components and the inlet and/or outlet compositions), (4) the costs of ESAs (i.e., the unit cost of hot and cold utilities), (5) the capital costs of counter-current mass exchange unit and distillation column, (6) the phase equilibrium relations for mass transfer between relevant components and the minimum composition difference, (7) the overall mass transfer coefficient and the relative volatility. The resulting separation network design should include: (1) the number and throughput of every mass exchange unit and distillation column, (2) the consumption rates of MSAs, ESAs and regenerating agents, and (3) the complete network configuration with the flow rate and composition of each branch stream.

Mathematical Model

The state-space superstructure was proposed by Bagajewicz and Manousiouthakis (1992) and

Bagajewicz et al. (1998) as an alternative representation of the mass and heat exchange network. Recently, this representation has been modified in a series of work for water-allocation network, heat exchange work and integrated water-allocation and heat exchange network design (Dong et al., 2008a,b; Zhou et al., 2009; Li et al., 2010). In our work, this original structure has been improved to incorporate additional design options, e.g., ESA and MSA process operators. More specifically, the overall separation network is viewed as a system of two interconnected blocks (see Figure 1). One is referred to as the distribution network (DN), in which all mixers, splitters and the connections between them are embedded. The other is the so-called process operator (PO), which can be further divided into two sub-blocks, i.e., PO-MSA and PO-ESA. All primary and regeneration mass exchange processes are performed in the former sub-block, while all distillation units are placed in the latter. Their inner stream connections and the corresponding mathematical models are described in the sequel.

Distribution network

In previous modeling approaches, system flows are specified by the identities of streams. Although such notations are quite straightforward, yet ambiguities may arise when it comes to stream mixing. To circumvent such problem, we propose a novel method which characterizes all flows with the splitting and/or mixing nodes (i.e., the splitters and/or mixers in DN) at both ends of the streams. Specifically, all rich and lean streams enter into or exit from the system via external nodes attached on DN, while all other nodes connected with PO block are considered as the internal nodes. Every input to DN is split into several branches at the splitting node and each of them is connected to a mixing node at the exit leading to one of the PO sub-blocks or to the environment. These splitting and mixing nodes are divided into several groups depending upon the original identities of streams or their

connections with the separation units in the PO block. For the sake of simplicity, the set SP is introduced to represent all splitting nodes on DN, while MX is used to denote all mixing nodes on DN. Notice that, at every splitter and mixer, the flow rate and mass balances must all be satisfied, i.e.,

$$f_{sp}^{\text{in}} = \sum_{mx \in MX} f_{sp,mx} \quad \forall sp \in SP \quad (1)$$

$$f_{mx}^{\text{out}} = \sum_{sp \in SP} f_{sp,mx} \quad \forall mx \in MX \quad (2)$$

$$f_{mx}^{\text{out}} \cdot c_{mx}^{\text{out}} = \sum_{sp \in SP} f_{sp,mx} \cdot c_{sp}^{\text{in}} \quad \forall mx \in MX \quad (3)$$

where f_{sp}^{in} denotes the total inlet flow rate to splitting node sp ; f_{mx}^{out} stands for the total outlet flow rate from mixing node mx ; $f_{sp,mx}$ denotes the flow rate from nodes sp to mx ; c_{sp}^{in} and c_{mx}^{out} represent respectively the compositions of key component at nodes sp and mx . Note that equation (3) is bilinear and can further be replaced with linear inequalities (Quesada and Grossmann, 1995). Furthermore, since not all streams are allowed to mix at certain mixing points, the following constraints should be enforced:

$$f_{sp,n_{sp}} = 0 \quad \forall sp \in SP, n_{sp} \in N_{SP} \quad (4)$$

$$nfs(sp, n_{sp}) = 0 \quad \forall sp \in SP, n_{sp} \in N_{SP} \quad (5)$$

where set N_{SP} denotes all forbidden mixing nodes of stream from node SP ; $nfs(sp, n_{sp})$ are binary variables that stand for the existence/nonexistence of the flow between nodes sp and n_{sp} . Finally, a negligible amount of flow is not allowed in the optimal operating policy and such uneconomically amount can be eliminated by the addition of the following constraint:

$$F_s^{\text{min}} \cdot nfs(sp, mx) + \varepsilon \leq +\varepsilon f_{sp,mx} \leq F_s^{\text{max}} \cdot nfs(sp, mx) + \varepsilon \quad \forall sp \in SP, mx \in MX \quad (6)$$

where F_s^{max} and F_s^{min} specify the upper and lower bounds of the flow rates in DN, ε is a sufficiently small positive value.

PO-MSA

Notice that, for a given design problem, the number of external nodes attached on DN can be obviously fixed so as to ensure a one-to-one correspondence between stream and node. However, the numbers of interior nodes associated with PO-MSA block are adjustable parameters and they should be chosen by designers. Generally, it is imperative to place enough interior nodes to provide opportunities to match the rich and lean streams. For illustration convenience, the number of rich streams, lean streams and the total number of internal nodes for rich and lean streams in PO-MSA are denoted as N_R , N_L , $size(N_R)$ and $size(N_L)$. It has been found the number of internal nodes connected with PO-MSA can be chosen within the following ranges:

$$N_R \cdot N_L \leq size(N_R) \leq 2N_R \cdot N_L \quad (7)$$

$$N_R \cdot N_L \leq size(N_L) \leq 2N_R \cdot N_L \quad (8)$$

The lower and upper bounds here can be obtained by providing respectively one and two potential match opportunities for each pair of rich and lean streams in the whole range of the composition scale. Here, it should be noted that (1) compared with the stage-wise superstructure, fewer possible combinations between rich and lean streams are needed to be given a priori, (2) the upper bounds of the junctions can guarantee the inclusion of the global optimal network. In this work, the appropriate number of interior nodes is initially fixed at the lower bounds and then their values are increased one-at-a-time from the lower limits. Correspondingly, the optimal separation network is identified by fixing the number of interior nodes and then solving the resulting MINLP model. This trial-and-error procedure is performed until the objective function of the problem concerned stops to improve. All mathematical constraints for PO-MSA are summarized as follows.

a. Flow Rate and Mass Balances for Each Mass Exchange Unit

The flow rate and composition balances around each mass exchange unit can be written as the

following linear equations:

$$fr_{me} = f_{rin_{me}}^{out} = f_{rout_{me}}^{in} \quad \forall me \in ME, rin_{me} \in RIN_{ME}, rout_{me} \in ROU_{ME} \quad (9)$$

$$cr_{me}^{in} = c_{rin_{me}}^{out} \quad \forall me \in ME, rin_{me} \in RIN_{ME} \quad (10)$$

$$cr_{me}^{out} = c_{rout_{me}}^{in} \quad \forall me \in ME, rout_{me} \in ROU_{ME} \quad (11)$$

$$fl_{me} = f_{lin_{me}}^{out} = f_{lout_{me}}^{in} \quad \forall me \in ME, lin_{me} \in LIN_{ME}, lout_{me} \in LOU_{ME} \quad (12)$$

$$cl_{me}^{in} = c_{lin_{me}}^{out} \quad \forall me \in ME, lin_{me} \in LIN_{ME} \quad (13)$$

$$cl_{me}^{out} = c_{lout_{me}}^{in} \quad \forall me \in ME, lout_{me} \in LOU_{ME} \quad (14)$$

where RIN_{ME} and LIN_{ME} , the subsets of MX , denote respectively the mixing nodes of rich and lean streams entering to unit ME ; ROU_{ME} and LOU_{ME} , the subsets of SP , represent respectively the splitting nodes of rich and lean streams released from unit ME ; fr_{me} and fl_{me} are the flow rates of rich and lean streams passing through unit me ; cr_{me}^{in} and cr_{me}^{out} stand for the compositions of rich stream at the inlet and outlet of unit me ; cl_{me}^{in} and cl_{me}^{out} stand for the compositions of lean stream at the inlet and outlet of unit me . Constraints 9-14 are obvious since all flow rates and compositions are identical at the outlet/inlet of DN and the inlet/outlet of PO-MSA.

b. Mass Load in Each Mass Exchanger

To determine the existence of each mass exchange unit, the following equations are needed:

$$m_{me} = fr_{me} \cdot (cr_{me}^{in} - cr_{me}^{out}) \quad \forall me \in ME \quad (15)$$

$$m_{me} = fl_{me} \cdot (cl_{me}^{out} - cl_{me}^{in}) \quad \forall me \in ME \quad (16)$$

$$M_{me}^{\min} \cdot w(me) + \varepsilon \leq m_{me} \leq M_{me}^{\max} \cdot w(me) + \varepsilon \quad \forall me \in ME \quad (17)$$

Here, the binary variable $w(me)$ is activated when there is a mass load m_{me} between M_{me}^{\min} and M_{me}^{\max} , which corresponds to the lower and upper limit of mass transfer. Bilinear constraints 15-16 are introduced to enforce that an equivalent amount of species is removed from the rich stream and then

transferred to lean stream.

c. Composition Differences

The composition differences in PO-MSA are calculated as follows:

$$\Delta c_{me}^1 = cr_{me}^{\text{in}} - (h_{me} \cdot cl_{me}^{\text{out}} + b_{me}) \quad \forall me \in ME \quad (18)$$

$$\Delta c_{me}^2 = cr_{me}^{\text{out}} - (h_{me} \cdot cl_{me}^{\text{in}} + b_{me}) \quad \forall me \in ME \quad (19)$$

$$\Delta c_{me} = \left[\frac{(cr_{me}^{\text{in}} - cr_{me}^{\text{out}})^{0.3275} + (h_{me} \cdot cl_{me}^{\text{out}} - h_{me} \cdot cl_{me}^{\text{in}})^{0.3275}}{2} \right]^{\frac{1}{0.3275}} \quad \forall me \in ME \quad (20)$$

where Δc_{me}^1 and Δc_{me}^2 denote the composition driving forces at both ends of the mass exchanger me ; h_{me} and b_{me} are equilibrium coefficients in exchanger me ; Δc_{me} stands for the logarithmic mean composition difference for unit me . In previous works, various forms of approximations for the logarithmic mean have been proposed to avoid numerical difficulties. In particular, Chen's modification of Underwood approximation is adopted (Chen, 1987), as shown in constraint 20. Here, it should be mentioned that in many cases, compared with Chen's first approximation (Chen, 1987), which is widely used in heat exchanger network designs, constraint 20 is more computationally tractable while barely bears any numerical differences. Finally, the compositions of rich and lean stream around every mass exchanger should satisfy the following thermodynamic constraints:

$$\frac{cr_{me}^{\text{in}} - b_{me}}{h_{me}} - cl_{me}^{\text{out}} \geq \Delta C_{me}^{\text{min}} \cdot w(me) \quad \forall me \in ME \quad (21)$$

$$\frac{cr_{me}^{\text{out}} - b_{me}}{h_{me}} - cl_{me}^{\text{in}} \geq \Delta C_{me}^{\text{min}} \cdot w(me) \quad \forall me \in ME \quad (22)$$

$$cr_{me}^{\text{in}} - cr_{me}^{\text{out}} \geq 0 \quad \forall me \in ME \quad (23)$$

$$cl_{me}^{\text{out}} - cl_{me}^{\text{in}} \geq 0 \quad \forall me \in ME \quad (24)$$

By specifying the minimum composition difference $\Delta C_{me}^{\text{min}}$, constraints 21-22 also ensure that

exchangers with infinite sizes do not occur in the network.

d. Sizing equations for mass exchange units

Mass exchange units can be broadly classified into two categories: stage-wise exchangers (denoted by ME^{TRA} in the following notations) and continuous-contact exchangers (represented by ME^{PAC}). When mass exchange takes place in a tray column, the Kremser equation can be adopted for determining the number of stages if both operating and equilibrium lines are linear. However, when using the traditional form of the Kremser equation in a MINLP environment, both the form of this equation and the singularities for certain solvers lead to numerical difficulties. To circumvent such problem, the Kremser equation for stage numbers is written as follows:

$$NT_{me} = \left[\frac{(cr_{me}^{in} - cr_{me}^{out})^{0.3275} + (h_{me} \cdot cl_{me}^{out} - h_{me} \cdot cl_{me}^{in})^{0.3275}}{(\Delta cin_{me})^{0.3275} + (\Delta cout_{me})^{0.3275}} \right]^{\frac{1}{0.3275}} \quad \forall me \in ME^{TRA} \quad (25)$$

where NT_{me} is the number of trays for unit me . It is worth mentioning that this alternative form of the Kremser equation is reformulated on the basis of the aforementioned log-mean composition differences and detailed steps of deriving this formula can be found in Shenoy's work (Shenoy and Fraser, 2003). Furthermore, in practical designs, the number of trays should always be a positive integer and this can be realized by rounding up the continuous variable NT_{me} to its nearest integer, i.e.,

$$NT_{me} \leq N_{me} < NT_{me} + 1 \quad \forall me \in ME^{TRA} \quad (26)$$

where integer variable N_{me} denotes the number of trays needed in the final design of unit me .

On the other hand, a continuous-contact packed tower can also be suggested for mass exchange in absorption or stripping. The required packed height for unit me is determined by a number of imaginary mass transfer units, NTU_{me} , and the overall height of a mass transfer unit, HTU_{me} . Here

calculations are based on the conditions in the rich stream and the overall packed height that are given by the following nonlinear equations:

$$NTU_{me} = \frac{cr_{me}^{in} - cr_{me}^{out}}{\Delta c_{me}} \quad \forall me \in ME^{PAC} \quad (27)$$

$$HTU_{me} = \frac{fr_{me}}{K_{re}a \cdot A_{me}} \quad \forall me \in ME^{PAC} \quad (28)$$

$$H_{me} = NTU_{me} \cdot HTU_{me} \quad \forall me \in ME^{PAC} \quad (29)$$

where H_{me} denotes the height of the packed mass exchange unit me ; $K_{re}a$ is the overall mass transfer coefficient based on the rich stream; and A_{me} denotes the area of unit me . Detailed explanations and derivations of these sizing formulas can be found in Chen's work (Chen and Hung, 2005b).

PO-ESA

Similar to the mass exchange units in PO-MSA, the distillation units used in the state-space model can also be viewed as off-line equipment available for possible installation. Here, it is worth noting that the number of inlet internal nodes attached on PO-ESA is simply the number of solvent which can be recycled by the ESA. It should also be noted that the optimal number of distillation units is subjected to economic considerations and it may not be necessary to use ESA in the final design. The complete mathematical models for PO-ESA are introduced as follows.

a. Flow Rate and Mass Balances For Each Distillation Unit

All constraints involving the flow rate and composition balances at the inlet and outlet of each distillation unit can be mathematically expressed as:

$$q_{dis}^{in} = f_{din_{dis}}^{out} \quad \forall dis \in DIS, din_{dis} \in DIN_{DIS} \quad (30)$$

$$c_{dis}^{in} = c_{din_{dis}}^{out} \quad \forall dis \in DIS, din_{dis} \in DIN_{DIS} \quad (31)$$

$$q_{dis}^{tout} = f_{tout_{dis}}^{in} \quad \forall dis \in DIS, tout_{dis} \in TOUT_{DIS} \quad (32)$$

$$c_{dis}^{tout} = c_{tout_{dis}}^{in} \quad \forall dis \in DIS, tout_{dis} \in TOUT_{DIS} \quad (33)$$

$$q_{dis}^{bout} = f_{bout_{dis}}^{in} \quad \forall dis \in DIS, bout_{dis} \in BOUT_{DIS} \quad (34)$$

$$c_{dis}^{bout} = c_{bout_{dis}}^{in} \quad \forall dis \in DIS, bout_{dis} \in BOUT_{DIS} \quad (35)$$

$$q_{dis}^{in} = q_{dis}^{tout} + q_{dis}^{bout} \quad \forall dis \in DIS \quad (36)$$

$$q_{dis}^{in} \cdot c_{dis}^{in} = q_{dis}^{tout} \cdot c_{dis}^{tout} + q_{dis}^{bout} \cdot c_{dis}^{bout} \quad \forall dis \in DIS \quad (37)$$

where DIN_{DIS} , the subset of MX , denote the mixing node of feed to distillation unit DIS ; $TOUT_{DIS}$ and $BOUT_{DIS}$, the subsets of SP , represent respectively the splitting nodes of streams from the top and bottom of distillation unit DIS ; q_{dis}^{in} and c_{dis}^{in} denote respectively the flow rate and composition of heavy key component of the inlet feed to unit dis ; q_{dis}^{tout} and q_{dis}^{bout} denote respectively the outlet flow rate from the top and bottom of unit dis ; c_{dis}^{tout} and c_{dis}^{bout} represent the compositions of heavy key component at the top and bottom of unit dis correspondingly. In addition, the following constraint should be added to specify the presence of each distillation unit:

$$Q_{dis}^{min} \cdot w(dis) + \varepsilon \leq q_{dis}^{in} \leq Q_{dis}^{max} \cdot w(dis) + \varepsilon \quad \forall dis \in DIS \quad (38)$$

where Q_{dis}^{min} and Q_{dis}^{max} specify the minimum and maximum inlet flow rate to unit dis and the binary variable $w(dis)$ is used to denote the presence of unit dis .

b. Shortcut Design of Distillation Unit

It has been assumed in section 2 that saturated liquids are fed into the distillation unit. As a result, according to the equilibrium relation for binary mixtures, we have:

$$x_{dis}^e = c_{dis}^{in} \quad \forall dis \in DIS \quad (39)$$

$$y_{dis}^e = \frac{\alpha_{dis}^{LK, HK} \cdot x_{dis}^e}{1 + (\alpha_{dis}^{LK, HK} - 1) \cdot x_{dis}^e} \quad \forall dis \in DIS \quad (40)$$

where x_{dis}^e, y_{dis}^e denote respectively the mass fractions of the heavy key component in liquid and

vapor phase at the feed plate; $\alpha_{dis}^{LK,HK}$ is the relative volatility of the light key component to the heavy key component. By further assuming an ideal binary system, the minimum number of equilibrium stages N_{dis}^{\min} can be approximated via the Fenske Equation (McCabe et al., 2001) from the terminal concentrations of the heavy key component, i.e.,

$$N_{dis}^{\min} = \frac{\lg \left[\frac{c_{dis}^{tout} / (1 - c_{dis}^{tout})}{c_{dis}^{bout} / (1 - c_{dis}^{bout})} \right]}{\lg \alpha_{dis}^{LK,HK}} \quad \forall dis \in DIS \quad (41)$$

However, as the inlet compositions of each component cannot be known in advance, the precise calculation of the minimum reflux ratio and number of plates in distillation unit is difficult to perform. For the sake of simplicity, the minimum reflux ratio and optimal operating reflux ratio (R_{dis}^{\min} and R_{dis}) can be estimated as follows:

$$R_{dis}^{\min} = \frac{c_{dis}^{tout} - y_{dis}^e}{y_{dis}^e - x_{dis}^e} \quad \forall dis \in DIS \quad (42)$$

$$R_{dis} = 1.5 R_{dis}^{\min} \quad \forall dis \in DIS \quad (43)$$

Constraints (42) is only a roughly approximation and such relationship is only valid for the normal equilibrium curve, which is concave downward throughout its length. Similarly, a simple empirical method due to the Gilliland (McCabe et al., 2001) is used for the preliminary estimate of the number of equilibrium stages. The correlation which only requires the knowledge of minimum number of plates and reflux ratio is given as follows:

$$\frac{NP_{dis} - N_{dis}^{\min}}{NP_{dis} + 1} = 0.75 \left[1 - \left(\frac{R_{dis} - R_{dis}^{\min}}{R_{dis} + 1} \right)^{0.5668} \right] \quad \forall dis \in DIS \quad (44)$$

where NP_{dis} denotes the number of equilibrium stages according to Gilliland relations. Since the number of plates should always be an integer and after rounding up the continuous variable NP_{dis} to its nearest integer we obtain the final number of plates (N_{dis}) needed at the operating reflux, i.e.,

$$NP_{dis} \leq N_{dis} < NP_{dis} + 1 \quad \forall dis \in DIS \quad (45)$$

Finally, the overall heat duty for reboilers and condenser (Φ_{dis}^R and Φ_{dis}^C) can be built as follows:

$$\Phi_{dis}^R = \Phi_{dis}^C = (R_{dis} + 1) \cdot q_{dis}^{out} \cdot r_{dis} \quad \forall dis \in DIS \quad (46)$$

where r_{dis} is the unit latent heat for mixtures to be separated in unit dis . Constraint 46 can be derived after some rearrangements of the basic heat balances equations, making use of the constant molal overflow assumption and neglecting the convective heat transfer and overall heat loss.

Objective Function

The objective function in this scheme is to minimize the total annualized cost (TAC), taking into account, (1) the costs of mass separating and regeneration agents, (2) the cost of ESAs (including the costs of hot and cold utilities), and (3) the installation costs of mass exchange units and distillation columns. The objective function can be written as follows:

$$\begin{aligned} Obj = & \sum_{me \in ME^{TRA}} C_{me} \cdot N_{me} + \sum_{me \in ME^{PAC}} C_{me} \cdot H_{me} + \sum_{dis \in DIS} C_{dis} \cdot N_{dis} \\ & + \sum_{pls \in PLS^{IN}} C_{pls} \cdot f_{pls}^{in} + \sum_{els \in ELS^{IN}} C_{els} \cdot f_{els}^{in} + \sum_{rls \in RLS^{IN}} C_{rls} \cdot f_{rls}^{in} \\ & + \sum_{dis \in DIS} (\Phi_{dis}^h \cdot C^h + \Phi_{dis}^c \cdot C^c) \end{aligned} \quad (47)$$

where sets PLS^{IN} and ELS^{IN} denote respectively the mixing nodes of process and external lean streams entering to DN; RLS^{IN} represent the mixing nodes of mass regenerating agents entering to DN; C_{pls} , C_{els} , C_{rls} , C_{me} , C_{dis} , C^h and C^c are all relevant annualized cost coefficients.

Application Examples

Four examples are given to illustrate the relative merits of the proposed formulation for separation network design. These examples include the traditional MEN design and the separation network design with both MSAs and ESAs. Using random initial values and perturbations, GAMS/DICOPT with CPLEX as the MILP solver and CONOPT as the NLP solver are used throughout the study.

Let us first consider the common background and corresponding parameters which are used in all examples. This process deals with the removal/recovery of phenols from four aqueous waste streams in a coal conversion plant, where the principal organic hazardous species in the liquid effluents are phenols. Detailed process description and the schematic diagram can be found in El-Halwagi and Manousiouthakis (1990b). The process data of all four phenol-rich streams are given in Table 1. Available for the dephenolization are two MSAs: light oil (S1) and activated carbon (S2). The light oil, which is made up of benzene-toluene-xylene mixture, is used on a ‘once-through’ basis, whereas the activated carbon can be regenerated and recycled after used. Unlike ‘once-through’ MSA, the inlet and outlet compositions of the regenerated MSA are not given and must be determined as part of the synthesis work. Composition and cost data for all lean streams and regenerating agent are also provided in Table 1. In all examples, the upper and lower bounds of the flow rates in DN (F_S^{\max} and F_S^{\min}) are set to 10kg/s and 0.01kg/s respectively. We also assume that the tray columns are used for the light oil, and the packed columns for absorption and regeneration for activated carbon. The cost data used in Papalexandri et al. (1994), as shown in Table 2, are applied for comparison. Furthermore, the equilibrium correlations for mass transfer between the rich streams and MSAs are:

$$\text{S1: } y = 0.71 \cdot x + 0.001$$

$$\text{S2: } y = 0.13 \cdot x + 0.001$$

On the other hand, the mass transfer equilibrium between the regenerable MSA (S2) and the regenerating agent (H1) is given by:

$$x = 1.38 \cdot z_1$$

Finally, the overall mass transfer coefficient ($K_{re} a$) and the minimum composition difference are taken to be 3.7(kg phenol $\text{m}^{-3} \text{s}^{-1}$) and 0.0001 respectively.

Example 1

The first example is addressed to demonstrate the effectiveness of our method in traditional MEN design. Let us first consider the aforementioned MEN design problem which has already been solved by several authors (El-Halwagi and Manousiouthakis, 1990b; Hallale and Fraser, 2000d; Papalexandri et al., 1994; Chen and Hung, 2005a; Isafiade and Fraser, 2008). The solution methods adopted and corresponding results are summarized in Table 3. In this example, the original MEN problem is solved with our proposed simultaneous solution strategy. To be able to compare different strategies on the same basis, the multi-stream mixing is forbidden and the corresponding model developed is slightly modified. Specifically, constraints for PO-ESA are removed and only costs of MEN are considered in the objective function. Solving the resulting MINLP model with the minimum number of internal junctions yields the minimum TAC network structure (see Figure 2). In this figure, numerical values denote respectively the flow rates and compositions, while the mass transfer load and the size of each exchange unit are shown in Table 5. The resulting network features a TAC of \$670,586 out of which the total capital cost (TCC) and total operating cost (TOC) are found to be \$77,323 and \$593,263 respectively. Here, the cost reduction with respect to the designs reported in Table 1 can be attributed to the overall network improvements, which are provided by additional splitting and mixing opportunities in the state-space superstructure. More specifically, stream splitting of S1 from unit 3 and 4 provides the most appropriate match opportunities and driving forces, so that the trade-offs between operating and investment costs in MEN design can be balanced more effectively. In fact, such mixing opportunity has never been considered by any of the previous method.

Example 2

To investigate the effects of multi-stream mixing, let us further assume that the four rich streams (R1-R4) are allowed to be merged. A typical state-space framework is constructed and every pair of splitter and mixer for rich streams is connected. The resulting optimal network and detailed design specifications of each mass exchanger are presented in Figure 3 and Table 5. The optimal TAC, TCC and TOC in this case can be further reduced to \$625,320, \$31,757 and \$593,563 respectively. The optimal number of mass exchange unit has also been cut down to four as a mass exchanger is not needed for direct mixing between streams. In terms of the utility cost, although consumption of S1 has been slightly increased, the demand of regenerating agent H1 have decreased considerably so as to reduce the overall cost of MSAs. Detailed comparisons of costs and specific designs with the previous one are listed in Table 4 and 5. It is apparent that both capital and utility costs can be lowered if multi-stream mixing can be considered as an added option in the MEN design.

Example 3

In this example, ESA is introduced as an alternative option for the lean stream regeneration. To facilitate the overall separation network design, an additional set of parameters are provided. Specifically, ESA is introduced to recover light oil (S1) from the spent mixtures, which are constituted of benzene, toluene, xylene and phenol. In the distillation scheme, xylene and phenol are chosen respectively as the light and heavy components and the relative volatility, which is 1.376, can be calculated according to the heuristic estimation proposed by Nadgir and Liu (1983). Furthermore, the latent heat of the benzene-toluene-xylene-phenol mixture is chosen to be 370kJ/kg and the annualized costs of hot and cold utilities are set to \$2,280s/kg and \$570s/kg respectively. Finally, the upper and lower bounds for inlet flow rate to distillation units (Q_{dis}^{max} , Q_{dis}^{min}) are taken to be 10kg/s and 0.1kg/s.

By constructing the state-space superstructure and solving the resulting MINLP, one can then generate the optimal structure in Figure 4. Notice that this network is assembled with four mass exchangers and one distillation unit. Notice also that fresh S1 has been recovered from the top of distillation unit, whereas the pure phenols are obtained from the bottom and can be sold directly. The corresponding minimum TAC of the separation network is reduced significantly to \$380,405, which consists of a TCC and TOC of \$268,568 and \$111,837. More specifically, although the capital investments and cost of ESAs are larger than those in the former studies, the utility costs of MSAs, namely S1 and S2, are reduced dramatically to \$13,493 and \$0 respectively. Accordingly, regenerating agent H1 is also not employed in the optimal separation scheme. Detailed designs of each separation unit and the corresponding costs of the network are summarized in Table 4 and 5. In addition to the economic advantages, another important feature is that our conceptual designs have less environmental impact, as the mass regenerating agent H1 is replaced by the more environmentally friendly ESAs. In particular, since rich and lean streams are directly contacted, using ESAs can prevent the process streams from being polluted by the undesirable species in regenerating agents. Also, on this note, further investment in recovering or disposing the waste regeneration agents can be avoided.

Example 4

The last example is the same as the third one, except that all rich streams are allowed to be mixed. Under this condition, the optimal network structure obtained is shown in Figure 5 and the TAC is now reduced to \$317,188. The corresponding capital and operating costs have decreased to \$227,600 and \$89,588. All other main design parameters of this network are provided in Table 4 and 5. As evident from Figure 5, both rich and lean streams go through mass exchangers in series and this constitutes a

significant departure from the previous case. Furthermore, instead of directly mixing with the original S1, as is the case in example 3, the regenerated S1 has matched with the rich stream discharged from unit 1 before mixing with the initial stream at an equal composition level. All these desirable characteristics should be ascribed to the multi-stream mixing options and state-space representation.

Computational Results

Table 6 shows the size of each example as well as the CPU time required to solve them with appropriate initial values in a 2.2GHz Intel Core Duo Processor. One can notice that computing times are relatively small. From the designs produced above, it can be concluded that our method, which enables the optimal selection of separating agents and arbitrary mixing and splitting, is indeed suitable for obtaining cost-optimal designs for separation network. However, we have to mention that the global optimal solutions of all cases cannot be guaranteed, because of the non-linearity and non-convexity of the proposed mathematical model.

Conclusions

An MINLP model has been presented in this work for one-step optimization of separation network with both MSAs and ESAs. The selection of MSAs and ESAs is rendered possible by resorting to the modified state-space superstructure, where arbitrary mixing and splitting options are easily incorporated. To illustrate the advantages and various aspects of our approaches, four cases were studied. From the results obtained so far, it can be clearly observed that the resulting networks are superior to those generated with other conventional methods. Better overall designs are brought about not only by the financial savings but also by the potential environmental benefits.

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Nomenclature

Sets and Indices

RS^{IN} = initial inlet nodes of rich streams to DN

PLS^{IN} = initial inlet nodes of process lean streams to DN

ELS^{IN} = initial inlet nodes of external lean streams to DN

RLS^{IN} = initial inlet nodes of regenerating agents to DN

RS^{OUT} = final outlet nodes of rich streams from DN

PLS^{OUT} = final inlet nodes of process lean streams to DN

ELS^{OUT} = final inlet nodes of external lean streams to DN

RLS^{OUT} = final inlet nodes of regenerating agents to DN

ME = set of mass exchange units (including regenerating units) in the system

DIS = set of distillation columns in the system

RIN_{ME} = mixing node of rich streams leading to the inlet of mass exchange unit me

$ROUT_{ME}$ = splitting node of the rich stream from the outlet of mass exchange unit me

LIN_{ME} = mixing node of lean streams (including regenerating agents) leading to the inlet of mass exchange unit me

$LOUT_{ME}$ = splitting node of the lean streams (including regenerating agents) from the outlet of mass exchange unit me

DIN_{DIS} = node denoting the inlet of the distillation columns *dis*

$TOUT_{DIS}$ = node denoting the outlet from the top of distillation unit *dis*

$BOUT_{DIS}$ = node denoting the outlet from the bottom of distillation unit *dis*

MX = all mixing nodes in the system,

$$MX = RS^{OUT} \cup PLS^{OUT} \cup ELS^{OUT} \cup RLS^{OUT} \cup RIN_{ME} \cup LIN_{ME} \cup DIN_{DIS}$$

SP = all splitting nodes in the system,

$$SP = RS^{IN} \cup PLS^{IN} \cup ELS^{IN} \cup RLS^{IN} \cup RIN_{ME} \cup LIN_{ME} \cup TOUT_{DIS} \cup BOUT_{DIS}$$

N_{SP} = all forbidden mixing nodes of stream from splitting node *sp*

Parameters

F_S^{\max}, F_S^{\min} = upper and lower bounds of the flow rates in DN

$M_{me}^{\max}, M_{me}^{\min}$ = upper and lower bounds of the mass exchanged in unit *me*

ΔC_{me}^{\min} = minimum composition difference for unit *me*

h_{me}, b_{me} = Henry coefficient and constant in mass exchange unit *me*

$K_{re} a$ = overall mass transfer coefficient

$\alpha_{dis}^{LK, HK}$ = relative volatility of the light key component to the heavy key component

r_{dis} = unit latent heat of the mixtures in distillation unit *dis*

$Q_{dis}^{\max}, Q_{dis}^{\min}$ = maximum and minimum inlet flow rate to unit *dis*

$C_{els}, C_{els}, C_{rls}$ = annualized cost coefficients for mass separating and regenerating agents

C_{me}, C_{dis} = annualized cost factors for mass exchange unit *me* and distillation unit *dis*

C^h, C^c = annualized cost coefficients for hot and cold utilities

Continuous variables

f_{mx}^{out} = total outlet flow rate from mixing node *mx*

c_{mx}^{out} = composition at mixing node mx

f_{sp}^{in} = total inlet flow rate to splitting node sp

c_{sp}^{in} = composition at splitting node sp

$f_{sp,mx}$ = flow rate from splitting node sp to mixing node mx

f_{me} = flow rate of the rich stream passing through mass exchange unit me

cr_{me}^{in} = composition of the rich stream leading to the inlet of mass exchange unit me

cr_{me}^{out} = composition of the rich stream from the outlet of mass exchange unit me

fl_{me} = flow rate of the lean stream passing through mass exchange unit me

cl_{me}^{in} = composition of the lean stream leading to the inlet of mass exchange unit me

cl_{me}^{out} = composition of the lean stream from the outlet of mass exchange unit me

m_{me} = mass exchanged of unit me

$\Delta c_{me}^1, \Delta c_{me}^2$ = composition driving forces at both ends of the mass exchanger me

Δc_{me} = logarithmic mean composition difference for mass exchanged of unit me

A_{me} = the area of mass exchange unit me

NTU_{me}, HTU_{me} = the number of transfer units and the height of a transfer unit for packed column me

H_{me} = the height of packed column me

NT_{me} = number of the trays in the tray column me

q_{dis}^{in} = inlet flow rate to distillation unit dis

c_{dis}^{in} = composition of the heavy key component in stream to distillation unit dis

q_{dis}^{tout} = outlet flow rate from the top of distillation unit dis

c_{dis}^{tout} = composition of the heavy key component in stream from the top of distillation unit dis

q_{dis}^{bout} = outlet flow rate from the bottom of distillation unit dis

c_{dis}^{bout} = composition of the heavy key component in stream from the bottom of distillation unit dis

N_{dis}^{min} = minimum number of equilibrium stages of distillation unit dis

NP_{dis} = number of equilibrium stages of distillation unit dis

R_{dis}^{min} = minimum reflux ratio of the of distillation column dis

R_{dis} = actual reflux ratio of distillation column dis

x_{dis}^e, y_{dis}^e = the mass fractions of the heavy key component in liquid and vapor at the feed plate in distillation unit dis

$\Phi_{dis}^R, \Phi_{dis}^C$ = the heat duty for reboilers and condenser for unit dis

Binary and integer variables

$nfs(sp, mx)$ = binary variables denoting the existence/nonexistence of the flow rate between nodes

sp and mx

$w(me), w(dis)$ = binary variables denoting the existence/nonexistence of the mass exchange unit

me and distillation unit dis

N_{me} = final number of trays after rounding up NT_{me} to the nearest integer

N_{dis} = final number of plates after rounding up NP_{dis} to the nearest integer

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Table 5. Design specifications of each separator in examples 1-4

Table 6. Problem size and computing time for each example

Table 1 Stream Data for examples 1-4

stream	Description	Flow rate (kg/s)	Inlet Conc.	Outlet Conc.	Cost (\$/kg)	Annual cost (\$/kg year)
R1	wastewater	3.3	0.05	0.0015	NA	NA
R2	condensate	0.6	0.07	0.003	NA	NA
R3	waste stream	1.4	0.02	0.003	NA	NA
R4	wastewater	0.2	0.03	0.002	NA	NA
S1	light oil	10	0.0013	0.025	0.01	58680
S2	active carbon	10			0.07	417060
H1	caustic soda	10	0	0.005	0.015	88020

NA= nonapplicable

Table 2 Capital cost data for examples 1-4 (with fixed diameter 1m)

Plate mass exchanger	4552N\$/year	For examples 1-4
Packed mass exchanger	4245H\$/year	For examples 1-4
Plate distillation unit	4552N\$/year	For examples 3 and 4

Table 3 Summary of previous results for the original MEN in example 1

	Methods	TAC(\$/year)
Papalexandri et al. (1994)	hyperstructure	957,000
Hallale and Fraser (2000d)	supertargeting	706,000
Chen and Hung (2005)	stage-wise superstructure	694,000
Isafiade and Fraser (2008)	IBMS	689,300

Table 4 Summary of the cost of each item in examples 1-4

	TAC(\$/year)	TCC(\$/year)	Costs of MSAs (\$/year)		Costs of ESAs (\$/year)	
			S1	H1	Hot utility	Cold utility
Example 1	670,586	77,323	564,565	28,698	0	0
Example 2	625,320	31,757	564,516	29,047	0	0
Example 3	380,405	268,568	13,493	0	78,675	19,669
Example 4	317,188	227,600	13,493	0	60,876	15,219

Table 5 Design specifications of each separator in examples 1-4

	Tray column			Packed column			Distillation column	
	No.	Number of stages	Mass load	No.	Height	Mass load	Number of plates	Reflux ratio
Example 1	1	7	0.158	5	0.778	0.002		
	2	3	0.040	6	0.280	0.002		
	3	3	0.024					
	4	3	0.006					
Example 2	1	4	0.225	3	0.770	0.002		
	2	2	0.004	4	0.277	0.002		
Example 3	1	8	0.160				40	4.040
	2	4	0.040					
	3	4	0.024					
	4	3	0.006					
Example 4	1	7	0.140				39	4.086
	2	1	0.085					
	3	3	0.004					

Table 6 Problem size and computing time for each example

Problem	Constraints	Binary Variables	Continuous variables	CPU time (sec)
Example 1	2544	634	838	15.34
Example 2	1123	294	420	7.27
Example 3	1344	405	539	4.36
Example 4	1248	405	539	5.78

Legend of Figures

Figure 1. The improved state-space superstructure

Figure 2. Optimal network configurations for example 1

Figure 3. Optimal network configurations for example 2

Figure 4. Optimal separation network designs in example 3

Figure 5. Optimal separation network designs in example 4

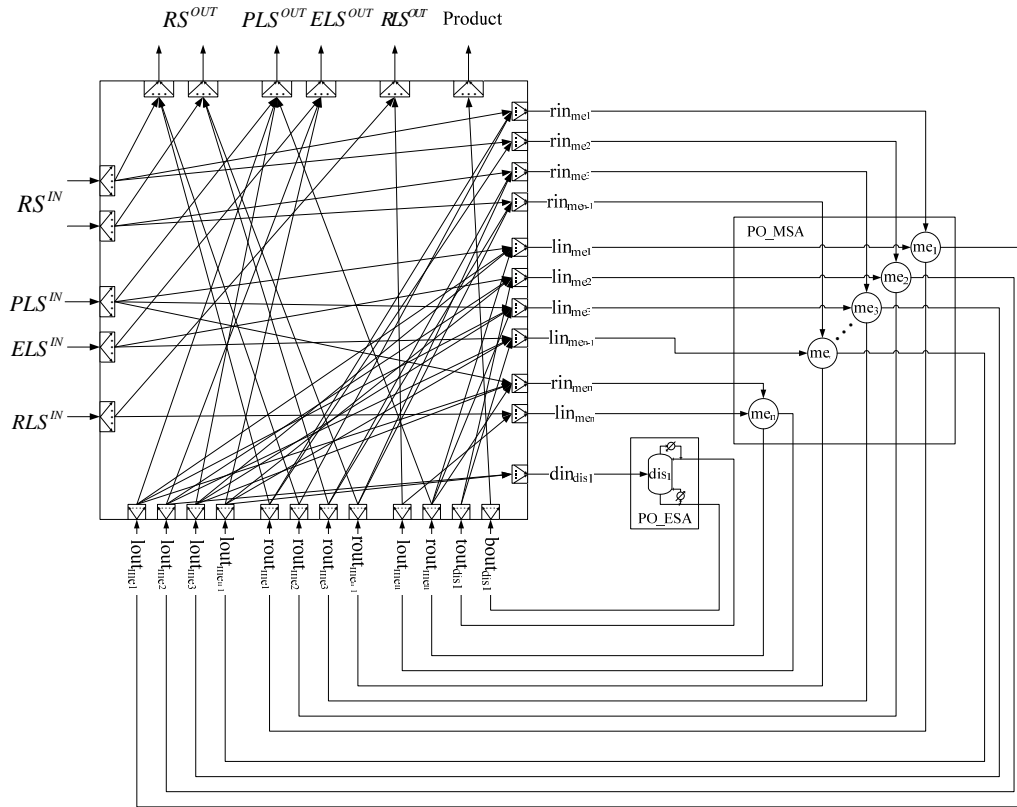


Figure 1. The improved state-space superstructure

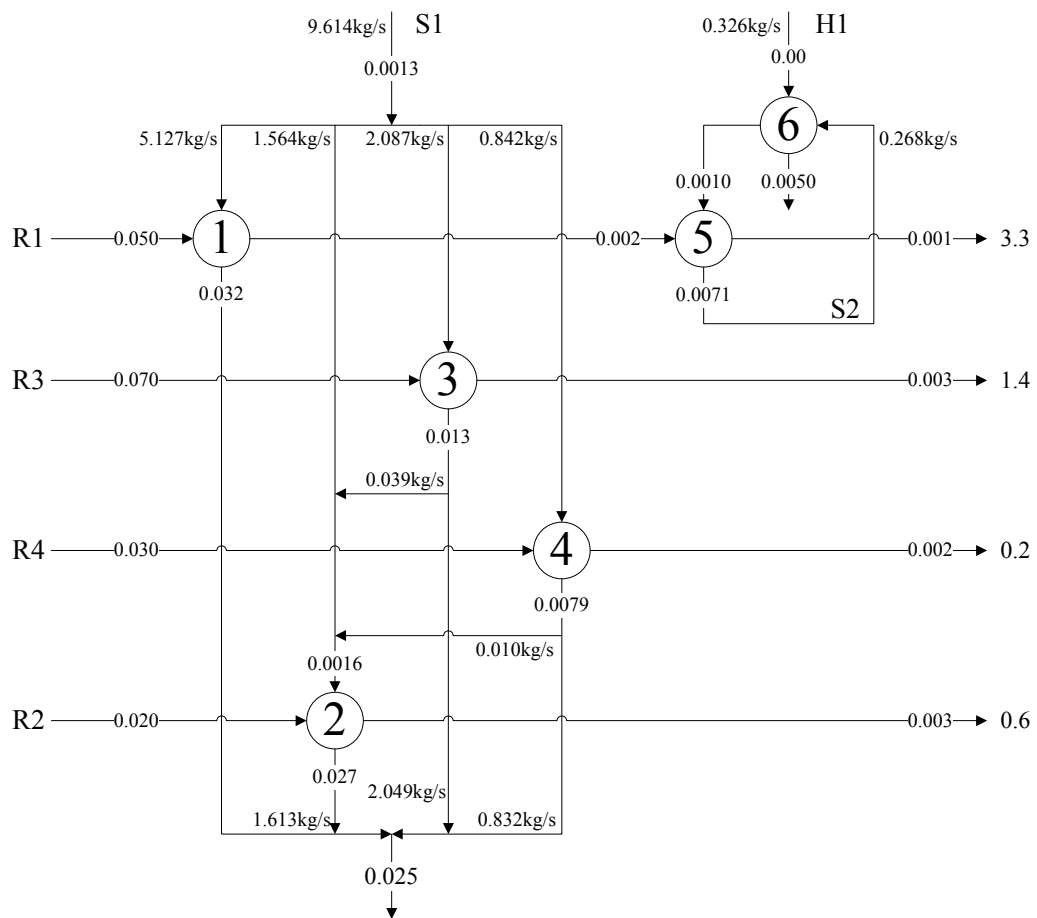


Figure 2. Optimal network configurations for example 1

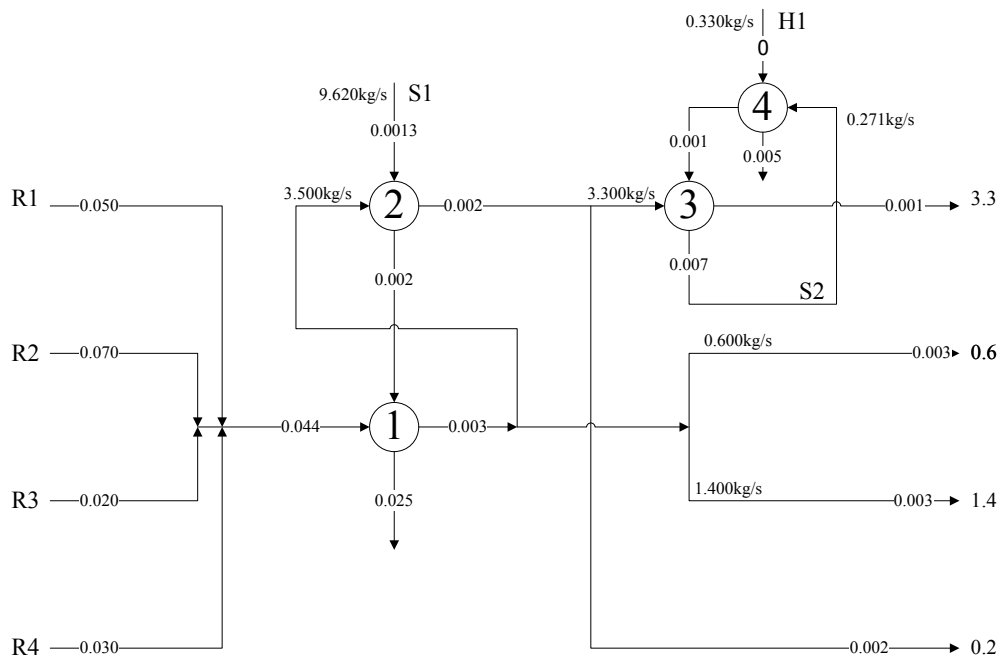


Figure 3. Optimal network configurations for example 2

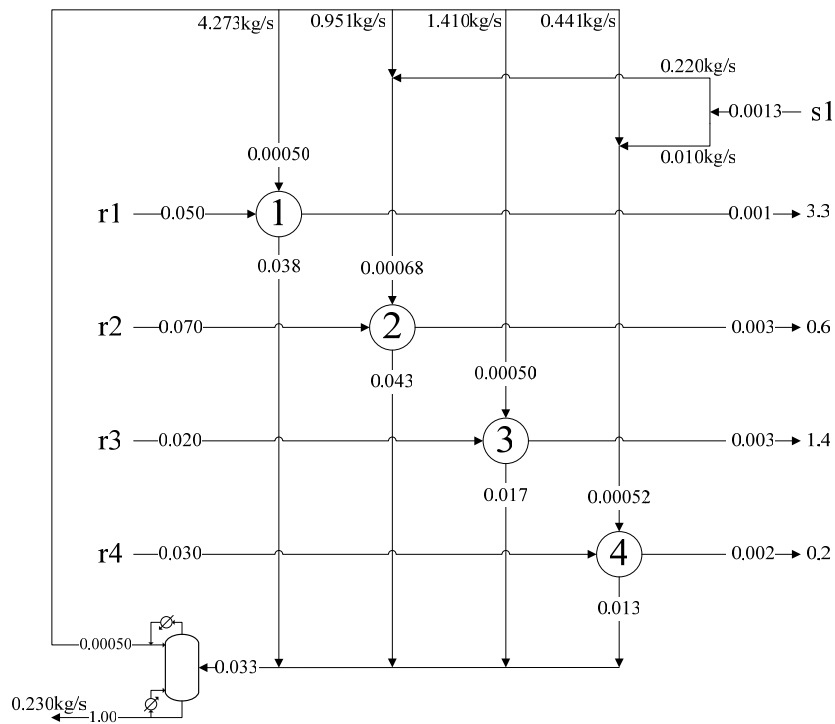


Figure 4. Optimal separation network designs in example 3

