

# Review of Mass Exchanger Network Synthesis Methodologies

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In this paper, a review of the methodologies that have been applied to the synthesis of mass exchange networks is presented, including their chronology. The paper also discusses the differences between a heat exchanger network and a mass exchanger network, highlighting aspects where it is difficult to establish a direct analogy between the two networks. The paper reviews the different mass exchanger network synthesis problem types that have been synthesized using the existing methods, as well as discussions on the benefits of combining the synthesis of heat and mass exchange networks, and a critical analysis of the aspects of mass exchanger network problems that still need to be investigated to fully establish the utility of this process synthesis problem.

## 1. Introduction

The attention of the chemical industry is not only focused on reducing the emission of greenhouse gases into the environment, but also ensuring that the use of scarce resources, such as water, is minimised while operating processes in a cost optimal way. Mass exchange network synthesis (MENS) can be used to achieve these sustainability and economic objectives by optimally allocating mass separating agents (MSAs), in mass exchangers, to streams rich in pollutant species to reduce their concentration to desired levels. This area of process synthesis, however has not received as much academic attention as its heat exchanger network synthesis (HENS) counterpart. The concept of MENS was first presented by El-Halwagi and Manousiouthakis (1989), by drawing analogies from HENS. Although much effort has gone into the synthesis of networks involving water minimisation, MEN problems, involving gaseous streams and other streams that are not water-based, has not been the subject of sufficient research attention. MENS also differs from the water minimisation problems in that its synthesis includes the design of the mass exchange units involved in the separation process with the goal of minimising total annual cost (TAC). Most of the synthesis methods that have been applied to MENs were developed using analogies that exist between HEN and MEN. These synthesis methods include both sequential and simultaneous techniques. In the sequential aspect, Pinch Technology has mostly been used while in the simultaneous aspect, the mass exchange equivalent of the HENS stage-wise superstructure (SWS) of Yee and Grossmann (1990) has been commonly used.

## 2. Analogies between HEN and MEN

Table 1 illustrates analogies between HENS and MENS. In HENS, there exist external hot and cold utilities contributing to the operating cost of the network. Utilities in MENS, known as mass separating agents (MSAs), are lean streams which fall into two categories: the first comprising process MSAs which are available within the process while the second are external MSAs which contribute to the operating costs.

Table 1: Analogies between HENS and MENS (Sztikai et al., 2006)

	HENS	MENS
Transported quantity	Heat	Mass
Driving force	Temperature difference	Concentration difference
Source	Hot process streams	Rich process streams
Sink	Cold process streams	Lean process streams

### 3. Pinch Technology as applied to MENS

HENS is the topic of process synthesis that has received the most attention in terms of process network optimisation. It started with the use of Pinch Technology (PT) as a sequential synthesis method, where minimum utility targets are found, and subsequent networks are designed to meet these targets (Linnhoff and Hindmarsh, 1983). This was accomplished using either a graphical approach, known as Composite Curves, or an algebraic-based approach called the Problem Table Algorithm (PTA). The synthesis of MENS followed a similar trend where the first Pinch Technology graphical-based approach was developed by El-Halwagi and Manousiouthakis (1989). The approach involves the use of the MENS Composite Curves to target the minimum MSA flows. The rich and lean Composite Curves are plotted on the Pinch diagram considering their minimum composition differences as shown in Figure 1a. This is then followed by network design to meet targets using the MENS Pinch design rules. It is worth mentioning that the Grand Composite Curves, which has found significant use in HENS, has also been developed for MENS for the determination of external MSA flows by Fraser et al. (2005). A key feature of the work of Fraser et al. (2005) is that external MSA selection should not only be based on cost per unit mass, but also on the concentration range permissible for mass load removal.

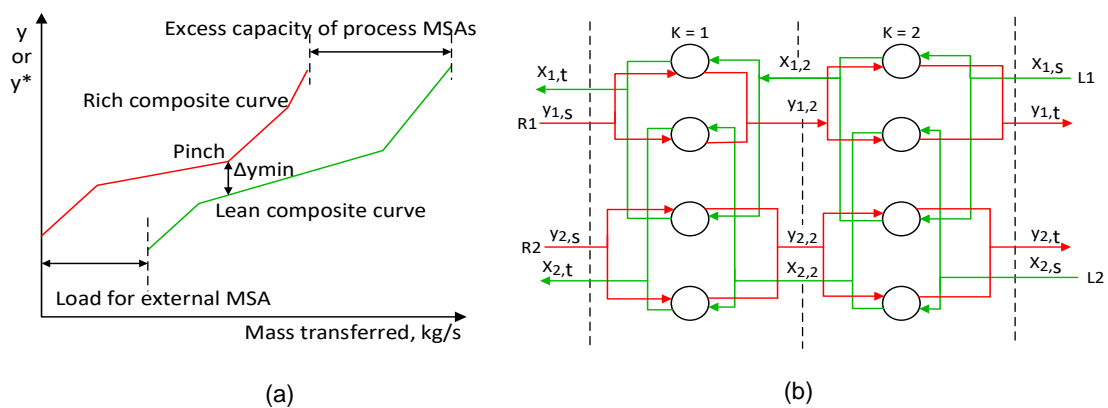


Figure 1: MENS diagrams (a) Composite curves (Hallale and Fraser, 2000b), (b) Stage-wise superstructure (Sztikai et al., 2006)

Automated synthesis approaches have also been applied to HENS and MENS. For HENS, Papoulias and Grossmann (1983) adopted the transshipment model to determine the minimum utilities using linear programming (LP). This was then followed using mixed integer linear programming (MILP) to determine the minimum number of units that satisfies the minimum energy targets of the LP. Again, analogies of this automated synthesis trend can be found in MENS where El-Halwagi and Manousiouthakis (1990a) also adopted the transshipment model to determine the minimum MSA flows and minimum number of units using LP and MILP.

Linnhoff and Ahmad (1990) stated that the two-stage synthesis of PT, which involves minimum utility target and network design to meet target, do not adequately trade-off capital and operating costs because potentially promising topologies may be excluded from the region of search. This shortcoming led to the development of minimum capital cost targets which are traded-off against operating cost targets at various minimum approach temperatures (Linnhoff and Ahmad, 1990). The network is then designed to meet targets using the minimum approach temperature that gives the lowest TAC target. A similar issue, as identified by Linnhoff and Ahmad (1990) in HENS, was noted in MENS by Hallale and Fraser (1998) where the authors stated that the two-step synthesis procedure of El-Halwagi and Manousiouthakis (1989), which involves identifying minimum MSA targets followed by a design to meet these targets, will not always result in the optimal network. Hallale and Fraser (1998) then included an additional synthesis step known as capital cost targeting (CCT) for MENS. The CCT entails using the composition interval diagram, and a newly developed  $y-x$  composite curve, to target the total minimum number of stages (for staged columns) and total minimum height (for packed columns), required to achieve specified mass exchange operations in a cost optimal way. Note that the  $y-x$  Composite Curves differs from the Composite Curves for MSA targeting of El-Halwagi and Manousiouthakis (1989). The Composite Curve is a plot of compositions versus mass load, while the  $y-x$  graph is a plot of composites of composition ranges of the rich streams participating in the problem versus composites of composition ranges of the lean streams in the problem. After the CCT from the  $y-x$  plot, network design techniques for MENS, developed by Hallale and Fraser (1998), which draws analogies from the vertical heat transfer concept of Linnhoff and Ahmad (1990) in HENS, are then used to meet the capital and operating cost targets in design. MSA targets and the

corresponding CCT are obtained at different values of minimum composition differences (MCD). The MCD that gives the lowest sum of minimum MSA cost target and minimum capital cost target, in what is known as MENS supertargeting, is then used to initiate network design using an updated version of the Pinch design method (Hallale and Fraser, 2000a).

According to El-Halwagi and Manousiouthakis (1989), a key difference between HENS and MENS is the absence of a one-to-one correspondence between rich and lean streams. The authors then developed Eq(1) to establish a one-to-one correspondence in all composition scales where feasible mass exchange exists.

$$y_i = m_j(x_j + \varepsilon_j) + b_j \quad (1)$$

According to Hallale and Fraser (2000b), Eq(1) can be used to establish CCT and designs to meet targets, for MENS problems involving non-overlapping MSAs, i.e. where the problem has one process MSA above the pinch and one or more external MSAs below the pinch. In this case, the Pinch is caused by the inlet composition of the process MSA. On the other hand, overlapping MSAs involve scenarios where one of the process MSAs crosses the Pinch. This implies that more than one MSA will exist on both sides of the Pinch thereby making the use of the  $y$ - $x$  graph difficult to apply in determining CCT. The reason for the difficulty is because there is no longer a single scale for  $x$ , since the MSAs have unique equilibrium relations ( $m$ ), implying that their composition scales are not equivalent. In overcoming this difficulty, Hallale and Fraser (2000b) expressed the  $x$  compositions in the lean stream for each MSA in their equivalent rich stream phases using the expression  $y^* = mx$ , thereby transferring all MSA compositions to a common basis. Using this approach implies that the minimum approach composition,  $\varepsilon$ , must be replaced by a minimum approach composition expressed in the rich phase as  $\Delta y_{\min} = y - y^*$ . Also, Hallale and Fraser (2000b) replaced the CCT tool  $y$ - $x$  with a new  $y$ - $y^*$  which accommodates problems involving overlapping MSAs. Other sequential approaches are the Pinch-based graphical method of Gadalla (2015) where target materials' composition in rich streams are plotted versus the equivalent corresponding compositions in lean streams.

#### 4. Mathematical Programming as applied to MENS

Simultaneous mathematical programming (MP) approaches have also found use in MENS. The first application of MP to MENS, which accommodates multi-component problems, was presented by Papalexandri et al. (1994) where a hyperstructure was modelled as a mixed integer non-linear program (MINLP). This was then followed by another MINLP hyperstructure-based approach which accommodates multi-period profiles and network flexibility (Papalexandri and Pistikopoulos, 1994). Although the hyperstructure approach simultaneously optimizes operating and capital costs, it is not straightforward to solve, and does not guarantee global optimality due to the highly non-convex nature of the MENS problem. This shortcoming led to the development of a more simplified MINLP formulation by Chen and Hung (2005a) found by drawing analogies from the stage-wise superstructure (SWS) synthesis method for HENS by Yee and Grossmann (1990). Figure 1b illustrates the SWS for MENS. This was later followed by a host of other SWS approaches, such as that of Szitkai et al. (2006), which handles multi-component problems and the interval based MINLP approach of Isafiade and Fraser (2008), where the superstructure is defined using the supply and target compositions of either the rich or lean streams. Other variants of the SWS for MENS are the methods proposed by Azeez et al. (2012), where the superstructure is defined using the supply of the rich streams and target compositions of the lean streams, and the supply-based superstructure technique of Azeez et al. (2013) where the superstructure is defined using the supply compositions of the rich and lean streams. Velázquez-Guevara (2018) also adopted an MINLP based approach using disjunctions.

Some of the key shortcomings of the PT approach include tediousness in applying the procedure, lack of a simultaneous trade-off between operating and capital costs as each step is dependent on the previous, and difficulty in solving large problems at the design stage. Hallale (1998) noted that in some cases TAC target values may beat actual design TAC values and vice versa. In the aspect of mathematical programming, especially when using the simultaneous based approach, getting feasible solutions remains a challenge due to the non-convex formulations.

#### 5. Other applications of MENS

It is worth mentioning that MENS has not been as well developed as its HENS counterpart despite the myriad of scenarios in the process and allied industries that requires its application. A common feature among all the works reviewed above is that the problems considered have few rich and lean streams. The largest problem considered in the literature so far involves five rich streams from which ammonia is to be removed and three water-based lean streams, where two of the lean streams are process MSAs while the third is an external MSA. This problem, which involves packed columns, was first presented by Hallale (1998) using PT, and it has been

solved using the fairly linear SWS MINLP formulation of Szitkai et al. (2006). It has also been solved using other methods such as the hybrid approach involving integer cuts and bounds by Emhamed et al. (2007), the interval-based approach of Isafiade and Fraser (2008), the supply and target-based approach of Azeez et al. (2012) and the supply-based method of Azeez et al. (2013). It is worth stating that methodologies for solving larger problems involving many rich and many lean streams, where both packed and stage-wise columns may be required, has yet to be developed. This is essential to handle problems that may require interplant mass integration as is the case with total site heat integration in HENS.

### **5.1 MEN retrofit**

In retrofit, very few papers have been presented. The first paper to address the retrofit of MENS is that of Fraser and Hallale (2000), where three stages utilizing PT was adopted. The stages include establishing retrofit targets for MSA savings and capital investment, generating a savings investment plot, and designing the network to meet targets. In mathematical programming, only two methods have been presented, the SWS based approach of Chen and Hung (2005b) and the reduced superstructure synthesis approach of Isafiade (2018). These three papers considered small examples involving two rich streams and two lean streams. New methods, which can handle realistic retrofit MENS problems involving large numbers of rich and lean streams need to be developed.

### **5.2 Multicomponent problems**

Another complication involved in MENS, unlike HENS, is the presence of multiple components to be preferentially transferred from rich streams to lean streams. Problems of this nature are important in CO<sub>2</sub> rich streams where other species may be present. Few synthesis techniques have been developed in this area. The methods include the pinch based technique of El-Halwagi and Manousiouthakis (1998), the CCT pinch based method of Hallale (1998), the hyperstructure based approach of Papalexandri et al. (1994), the MINLP SWS based approach of Chen and Hung (2005a), the fairly linear MINLP SWS based approach of Szitkai et al (2006) and the superstructure based method of Linlin et al. (2013), which does not involve the selection of a key component. All the methods were applied to the popular coke-oven gas sweetening problem (El-Halwagi and Manousiouthakis, 1989). The problem, which involves simultaneously removing CO<sub>2</sub> and H<sub>2</sub>S, has just two rich streams and two lean streams. The existing methods need to be extended to larger problems having more than two components, or new methods should be developed to make MENS realistic for industrial application.

### **5.3 CHAMENS involving regeneration**

Combined heat and mass exchange network synthesis (CHAMENS) is beneficial in that absorption is improved at lower temperatures while regeneration using stripping and other methods is improved at higher temperatures. Regenerating external MSAs may also be beneficial for a variety of reasons such as cost, environmental regulations, etc. This implies that integration methods that adequately trade-off the multiple competing variables in multiple systems such as CHAMENS need to be developed. However, very few papers have been presented in this application area. El-Halwagi and Manousiouthakis (1990b) integrated regeneration with primary MENS, however the authors did not include heat integration. The first set of papers that included heat integration are the works of Srinivas and El-Halwagi (1994) where a two-stage mathematical programming targeting approach was adopted and the hyperstructure-based formulation of Papalexandri and Pistikoupoulos (1994). Other works in this area include the pinch based method of Isafiade and Fraser (2007), the interval-based approach of Isafiade and Fraser (2009), the combined mass-pinch and pseudo T-H diagrams of Liu et al. (2013) and the non-linear programming (NLP) SWS based method of Liu et al (2015). A key variable missing from these approaches has been that the temperature at which the regeneration occurs has always been fixed, or different temperatures investigated sequentially, as none of the models included variable mass transfer coefficients. Recently, Isafiade (2017) extended CHAMENS to include integration with solar thermal and heat storage, using a simplified multi-period profile to capture solar irradiation availability. This application area of MENS still requires many contributions considering integration with multiple types of renewable energy, detailed multi-period profiles that capture the availability of the renewable energy sources, and heat storage whose dynamic profile is adequately accounted for in the overall network.

### **5.4 Multi-period and flexible MENS**

Supply and target compositions of rich and lean streams may fluctuate around nominal values due to changes in environmental conditions, feedstock or product quality demand changes, upstream process upsets, etc. The few methods that have been developed for the multi-period/flexible scenario in MENS include the hyperstructure of Papalexandri and Pistikoupoulos (1994), the interval-based approach of Isafiade and Fraser (2009b), the SWS based method of Chen and Hung (2007) and the supply-based approach of Isafiade and Short (2016). More robust methodologies, extendable to larger numbers of periods, need to be developed.

## 5.5 MENS involving detailed unit designs

As is the case in HENS, where most papers have simplified the design of individual heat exchangers in the network, so also it is in MENS. Most papers in MENS have used fixed column diameters, and simplified capital cost functions where column capital costs are based only on number of trays and height for stage-wise and continuous contact columns. Mass absorber design needs to consider other issues such as flooding, pressure drop, packing characteristics, stream-dependent mass transfer coefficients, etc. The few papers that have considered one or more of the aforementioned design parameters, are the papers by Hallale and Fraser (2000c) where the pinch based CCT was adopted, the supply-based approach of Isafiade and Short (2016) and the paper by Short et al. (2018) which applied a two-step hybrid optimization procedure involving a nonlinear programming optimization of a detailed packed column model. It is worth mentioning that the method of Hallale and Fraser (2000c) is fraught with the tediousness associated with PT based approaches while that of Isafiade and Short (2016) may not be able to guarantee global optimal solutions due to the highly non-convex nature of the model equations involved when detailed column design is considered. The paper by Short et al (2018) may not guarantee global optimal solution as the NLP step is dependent on solutions obtained at the MINLP step.

## 6. Conclusions

This paper has presented a mini review of the methods that have been used in MENS including analogies with HENS. Despite the fewer papers that have been published in this area of process synthesis, when compared to HENS, a relatively high number of applications have been addressed. Most work in MENS has addressed networks involving gas-liquid streams running through tray and packed columns. More work needs to be done to address problems involving liquid-liquid streams, and other kinds of separation processes such as stripping, adsorption, membrane separations, and a combination of these options. Problems involving many rich and lean streams also need to be studied as this will be useful in scenarios involving mass integration over multiple plants or integrating the resource utilisation networks of industries with residential areas and agricultural sectors. In the area of multicomponent systems that require retrofitting, robust methodologies, that are able to handle streams having more than two components, also need to be developed as this will be useful in the purification of gases, such as biogas, to streams of higher grade. Additionally, it is vital that, to make MENS a possibility in industrial applications, more detailed design aspects are included at the network synthesis level. Specifically, the inaccuracies of the shortcut models used in the current network formulations need to be addressed (fixed diameters, fixed mass transfer coefficients, etc.) as these lead to vastly different networks, with implications on costing.

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