

Solve Design Puzzles with Mass Integration

Mass integration provides a comprehensive methodology for targeting yields, emissions, and capacities, and designing reaction, separation, and waste-processing systems. It now is starting to play an important role industrially.

Mahmoud M. El-Halwagi,
Auburn University
H. Dennis Spriggs,
Matrix Process Integration, L.C.

The design of any industrial process must address two important dimensions: mass and energy. Mass involves the creation and routing of chemical species in reaction, separation, and byproduct/waste-processing systems. These constitute the core of the process and define a company's technology base. Energy provides the necessary heating, cooling, and shaftwork for those systems.

Design always has been part science and part art. Engineers typically start with a previous design and use experience-based rules and know-how, along with their creativity, to evolve a better design. They rely on computer-based tools such as process simulators and unit-operation design programs.

The result of this approach has been good designs that work — but that often have substantial scope for improvement.

Process integration

Starting in the 1970s, engineers began to realize that correctly assembling the process building blocks is just as important as properly selecting and designing individual components. They also discovered that fundamental principles can guide this assembly.

This led to the concept of integrated process design or process integration, which emphasizes the unity of the entire process. It addresses the big picture first using fundamental principles, and then tackles design details only after the major structural decisions have been made. This approach assures that the correct details receive attention.

The first significant success of this approach was in energy integration. So-called thermal pinch technology or pinch analysis became available to industry about 1980 and, since then, has matured and been widely applied.

Pinch analysis is the definitive way to design heat recovery networks, select process-wide utility heating and cooling levels, and establish the energy/capital tradeoff for heat recovery equipment. It also provides insight into other elements of design — but it does not address issues related to mass in the reaction, separation, and byproduct/waste-processing systems.

Since the introduction of pinch analysis in the 1980s, substantial progress has been made in the broader discipline of energy integration (1). Steam, fuel, and on-site power-generation systems now can be analyzed as integrated systems, and the interactions among them can be understood for improved design and operation.

The most recent success in process integration has been in extending it to mass. Mass integration is analogous in many ways to energy integration, but tackles the core of the process and, consequently, has a more direct and significant impact on process performance.

Mass integration (2,3) addresses the conversion, routing, and separation of mass, and deals directly with the reaction, separation, and byproduct/waste-processing systems. It guides designers in routing all species to their most desirable destinations and in establishing mass-related

cost tradeoffs. Mass integration also defines the heating, cooling, and shaftwork requirements of the process. Furthermore, it provides insight into other design issues such as debottlenecking utility systems and selecting catalysts and other material utilities. It does not directly address energy system design.

Mass and energy integration complement each other, as shown in Table 1. Together, they form the basis of a comprehensive methodology for integrated process design.

Spurring the approach

The original motivation for mass integration was pollution prevention. In 1989, El-Halwagi and Manousiouthakis (4) first suggested the use of mass exchange networks (MENs) to guide the removal of pollutants using mass separating agents (MSAs), such as solvents, adsorbents, stripping agents, and ion exchange resins. El-Halwagi and Manousiouthakis discovered that:

- There is a close analogy between MENs and heat exchange networks (HENs) at the system level. This is a macroscopic generalization of the microscopic analogy between mass transfer and heat transfer (for instance, Fick's law for diffusion vs. Fourier's law for heat conduction).

- Rich and lean streams can be formed into composite curves, resulting in a "mass pinch" analogous to the "heat pinch" discovered by Hohmann (5) and later extended by Linnhoff and Hindmarsh (6), and others.

- Rigorous performance targets can be set ahead of design for the minimum cost of MSAs and for the selection of the optimal types of MSAs.

- Notwithstanding these similarities, designing MENs is far more challenging than designing HENs. This is due to the direct contact nature of mass exchange, the more involved equilibrium relationships for mass transfer as compared with the simple equilibrium expressions for heat transfer, the need to regenerate

MSAs, the multicomponent nature of the problem, and the wide diversity of mass exchange operations and technologies.

Although useful, MEN synthesis had several limitations. It focused on separations that were induced by MSAs. It also dealt with streams that were to be discharged to end-of-pipe treatment or receiving media. Furthermore, it did not tackle how streams should be routed, reacted, and allocated throughout the process.

Over the past few years, these limitations have been addressed. The result has been the development of a general framework called mass integration (2,3). It addresses the global allocation of mass, including species utilization and recycle, species interception within the process, species conversion via chemical reactions, stream segregation, and mixing, and for the manipulation of unit operations through design and operating changes. Further, the focus of mass integration has gradually shifted from pollution prevention to the more general objectives of optimal allocation of species for yield enhancement, process debottlenecking, cost reduction, and the entire range of processing objectives. So, let's now look at how mass integration can be used today.

GENERAL CONCEPTS

Integrated process design differs fundamentally in approach — and results — from traditional design. The important distinguishing features include:

- *Problem structure.* This is the way in which the overall, complex design problem is set up and decomposed into tractable subproblems. This defines the engineers' focus and dictates the tools required to carry out design.

- *Targeting.* Quantitative performance targets are set ahead of design. Design methods then guide the engineers in developing alternative structures that meet the targets. Targeting tells the engineers where the learning curve eventually will lead and allows

them to achieve this level of performance now rather than in the future.

- *Solution fragments.* Any design is composed of many individual design decisions. And, for each decision, there are alternatives. The first step in designing to meet the performance targets is to generate these alternatives or "solution fragments" systematically. Solution fragments are not unit operations and are not necessarily mutually exclusive; rather, they constitute alternative ways to solve design subproblems.

- *Integrated solutions.* Any design problem will have more than one integrated solution, each composed of a different combination of solution fragments. Another unique feature of process integration is the systematic way in which integrated solutions are generated, incorporating consistent sets of solution fragments and screening out inappropriate ones.

Solution techniques

Clearly, given the vast number of possible solutions to any design problem, it is imperative that optimal solutions be found without having to enumerate all options. Two main approaches — structure independent and structure based — have been developed to accomplish this.

The structure independent (or targeting) approach is based on undertaking design in stages. In each stage, design targets are set and then employed in subsequent stages. These targets are fundamental and are determined ahead of design without commitment to the final system configuration. This approach relies on an array of visualization tools that help engineers both to "see" the design problem and to generate integrated solutions. Examples include mass and energy composite curves and their corresponding pinch points. Visualization tools keep engineers involved in the design process and incorporate their judgment and preferences while generating optimal solutions.

The targeting approach offers two main advantages. First, within each

stage of design, the problem scope is reduced to a manageable size. Second, it provides valuable insights into system performance and characteristics.

The structure based approach involves developing a framework that embeds all potentially interesting solutions. Examples include superstructures (7), state-space representations (8), and process graphs (9). Mathematical programming (10), typically in the form of a mixed integer nonlinear program (MINLP), is used to extract solutions from the general framework. Integer variables correspond to the existence or absence of certain

equipment in the solution, while continuous variables determine the optimal values of design and operating parameters such as flow rates, temperatures, pressures, and unit sizes.

This approach is potentially robust, but its success strongly depends upon overcoming three challenges. First, the solution framework must embed the solutions of interest; otherwise, they will not be found. Second, due to the nonlinearity of the MINLP formulation, there often are severe computational problems. (Currently, commercial optimization software cannot guarantee the global solution

of general MINLPs.) Third, the engineers' input, preferences, judgment, and insights must be incorporated during problem formulation, which can be a tedious task, before the MINLP is set up.

Both the structure independent and structure based solution techniques are used extensively in the following integrated process design procedures.

Setting up the design problem

Mass integration deals with creating desired species, minimizing undesired ones, and routing all species

Impact	Energy Integration*	Mass Integration
Addresses directly: Dictates design	Targeting: Heat recovery Utility heating/cooling requirements Heating/cooling levels Cost tradeoffs: Heat recovery equipment Energy processing systems (steam, cooling, power) System design: Heat exchange networks (HENs) Steam system Power system Cooling system (air, water, refrigeration)	Targeting: Yields Emissions, pollution prevention (air, water, solids) Capacities, debottlenecking Cost tradeoffs: Mass exchange equipment Mass processing systems (reaction, separation, byproduct/recycling, water, fuel) System design: Mass exchange networks (MENs) Material routing Reaction system Separation system Byproduct/waste-processing system Water system Fuel system Sets process energy requirements: Heating Cooling Shaftwork
Addresses indirectly: Influences design	Improving core process energy efficiency: Water system Fuel system Utility emissions	Improving energy system structure/performance: Steam system Power system Cooling system Selecting/designing catalysts
Does not address: No influence on design	System design: Reaction system Separation system Byproduct/waste-processing system Targeting/cost tradeoffs for mass processing systems	System design: Steam system Power system Cooling system (air, water, refrigeration) Targeting/cost tradeoffs for energy processing systems.

* Includes thermal pinch analysis, which focuses primarily on HENs.

(desired and undesired) to their most appropriate destinations. The focus initially is on chemical species and not on unit operations. This transforms how we think about design and how we set up the design problem, and also dictates the tools required to perform design.

The first step in performing mass integration is to represent all species in the entire process (11). This is shown in Figure 1. Here, each species initially is addressed individually and, then, all are addressed simultaneously. For each targeted species, there are sources (streams that carry the species), and process sinks (units that can accept the species). Sinks include reactors, separators, heaters, coolers, pumps, compressors, biotreatment facilities, discharge media, and the like. Streams leaving the sinks, in turn, become sources. Therefore, sinks also are generators of the targeted species. Each sink/generator can be manipulated via design or operating changes to alter the flow rates and compositions that it can accept and that it discharges. It may be necessary to modify sources' compositions to prepare them for the sinks. This is done in a network of separation units referred to as the species interception network (SPIN).

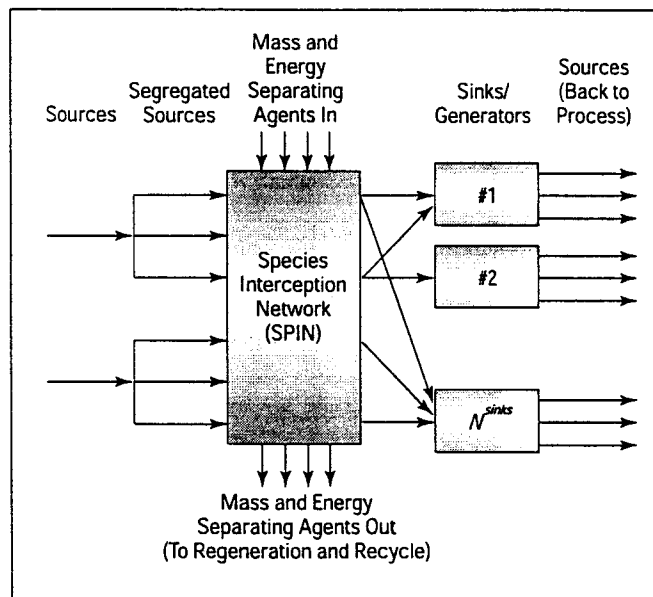
Mass integration, therefore, involves a combination of stream segregation, mixing, interception, recycle from sources to sinks (with or without interception), and sink/generator manipulation.

Segregation is simply avoiding stream mixing. Segregating streams with different compositions may make it unnecessary later to change their compositions. This can result in reduced cost and may allow streams to be recycled directly to sinks without further processing.

Mixing of process streams can be used to achieve appropriate flow rates, composition, and physical properties.

Interception is the use of separation technologies to adjust the compositions of the species-laden streams

■ *Figure 1. Mass integration from a species perspective (3,11).*



to make them acceptable for the sinks. These separations may be effected by the use of MSAs or energy separating agents (ESAs). Identifying the right combination for a SPIN can be a large and complex problem — because numerous streams typically must be processed, many separation technologies may be applicable, and, initially, it is not known how much of a species must be removed to make that stream suitable for a sink. Therefore, a systematic technique is needed to screen the candidate separating agents and separation technologies to find the optimal SPIN. This will be discussed in detail later.

Recycle refers to the routing of a source to a sink. Each sink has a number of constraints on the flow rates and compositions of feeds that it can process. If a source satisfies these constraints, it may be recycled directly to the sink. If the source violates these constraints, however, then segregation, mixing, or interception may be used to prepare the stream for recycle.

Sink/generator manipulation involves design or operating changes that alter the flow rates and compositions of sources entering or leaving the sinks. These measures can include temperature or pressure changes, unit replacement, catalyst alteration, feed-

stock and product substitution (12–14), reaction changes (15–17), and solvent substitution (12,18).

This approach to design causes engineers to ask new questions about a process. What streams contain the species of interest? Are these necessarily the streams to deal with or should they be modified? How? What are the possible destinations for these streams and are there constraints? Can these constraints be altered? What changes in compositions are required to prepare the streams for these destinations? What are the candidate technologies to use in modifying compositions? Where in the process should the streams be intercepted and modified?

All of these questions can be answered systematically through the use of mass integration tools.

VISUALIZATION TOOLS

Let's now look at some of the visualization tools. (The mathematical techniques that are used with these tools are covered in the literature, e.g., Ref. 19, and, so, will not be discussed here.)

The source-sink mapping diagram

As we've already noted, recycle involves routing a source to a sink.

The source may be recycled directly or after segregation, interception, or mixing with another source. Direct recycle (with and without segregation) always should be investigated before considering interception, because it typically is less expensive than recycle combined with interception.

The source-sink mapping diagram (2,19) is a visualization tool that can be used to determine direct recycling opportunities. As shown in Figure 2, for each species, a plot is constructed either of the flow rate or the species mass load (y axis) vs. composition (x axis).

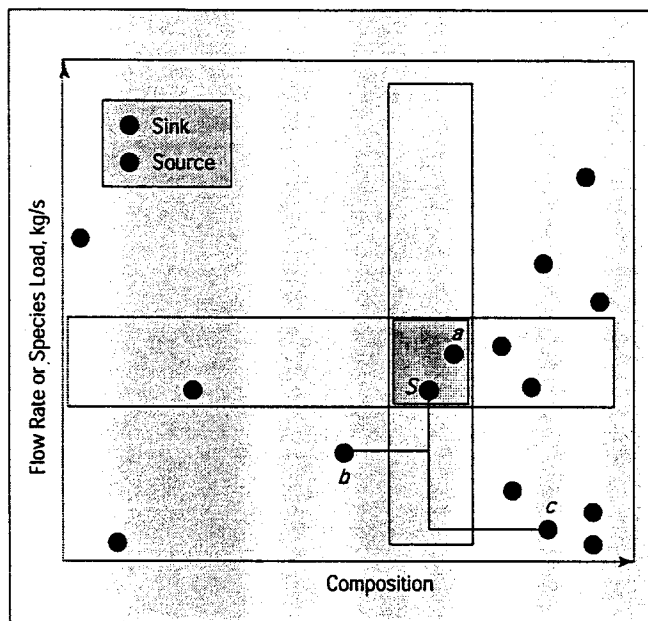
On this diagram, sources are represented by black circles and sinks by magenta circles. Typically, process constraints limit the range of species' composition and load that each sink can accept. The intersection of these two bands defines a zone of acceptable composition and load for recycle. If a source (for instance, *a* in Figure 2) lies within this zone, it can be recycled directly to the sink (for example, *S*). Moreover, sources *b* and *c* can be mixed using the lever arm principle to create a mixed stream that also can be recycled to *S*. In addition, the source-sink mapping diagram can be used to determine the extent of interception needed. If a source lies to the right of a sink, it can be intercepted to bring it within the band of acceptable recycle. We will discuss the problem of simultaneously intercepting several sources later.

Similarly, multiple components can be handled simultaneously. Figure 3 shows a three-component source-sink mapping diagram (20). On this diagram, ternary lever-arm rules can be employed to determine the extent of mixing among the various streams to satisfy sink constraints.

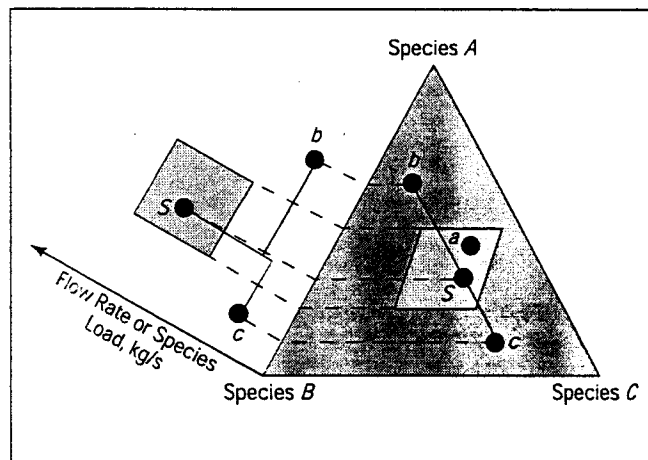
The path diagram

The path diagram (3,19) is a visualization tool that tracks the flow of a specific targeted species throughout the process. It is a plot of the load of that targeted species as a function of

■ **Figure 2.**
Recycle opportunities using the source-sink mapping diagram (2,19).



■ **Figure 3.**
A ternary source-sink mapping diagram (20).



its composition in the carrying streams. It relates the flow of the species to the performance of the different processing units through material balances and unit modeling equations. Units that do not involve the targeted species are excluded. Thus, the path diagram provides an easy way to track a species.

A path diagram is required for each targeted species in each phase (gas, liquid, and solid). Each stream is represented by a node on the diagram and these nodes are connected using composition profiles representing the changes occurring within the unit operations. The exact shape

of the composition profiles within units typically is not needed unless modifications within the units are to be considered. Therefore, these profiles can be represented by straight arrows. The direction of these arrows reflects the direction of mass flow. Because a unit may have multiple inputs and outputs, each node may be associated with multiple arrow heads and tails.

Figures 4 and 5 illustrate the construction of the path diagram. Figure 4 shows a section of a hypothetical process carrying the species of interest. Here, V_v is the flow rate in the v th gas stream or source, y is the compo-

sition of the species in the stream, and ϕ_v is the load of the species, so that:

$$\phi_v = V_v y_v \quad (1)$$

Similarly, the load of the species in the w th liquid source is given by

$$\psi_w = W_w z_w \quad (2)$$

where ψ_w , W_w , and z_w are the load, flow rate, and composition, respectively, of the w th liquid source (node).

The path diagram for the species in the gas phase is shown in Figure 5. The first processing step for the gas phase is a stripping operation in which the targeted species is transferred from a liquid stream (source $\omega = 1$) to the gas stream ($\nu = 1$). The path profile for the gas phase is represented by a straight line between inlet and outlet compositions that has a slope approximately equal to the flow rate of the stream. The gaseous stream leaving the stripper then is processed in a continuously stirred tank reactor where additional mass of the targeted species, ϕ_a , is generated by chemical reaction. Due to the complete mixing in the reactor, the concentration of the targeted species instantaneously changes from the inlet concentration, y_2 , to the outlet concentration, y_3 . And, the species loading increases to $\phi_3 (= \phi_2 + \phi_a)$. The effluent from the reactor is mixed with another process stream ($\nu = 4$) to give a resulting composition, y_5 , and mass loading, ϕ_5 . The composition y_5 can be determined graphically using the lever arm principle. The final operation involves the separation of the mixed stream into two terminal streams ($\nu = 6$ and $\nu = 7$). Though not shown, there is an equivalent liquid-path diagram for the process.

The path diagram provides a "big picture" view of a species. This is fundamentally different from the traditional equipment-oriented descriptions of a process in the form of process flow diagrams (PFDs) and piping and instrumentation diagrams (P&IDs). These are useful for certain purposes but mask the propagation and fate of individual species.

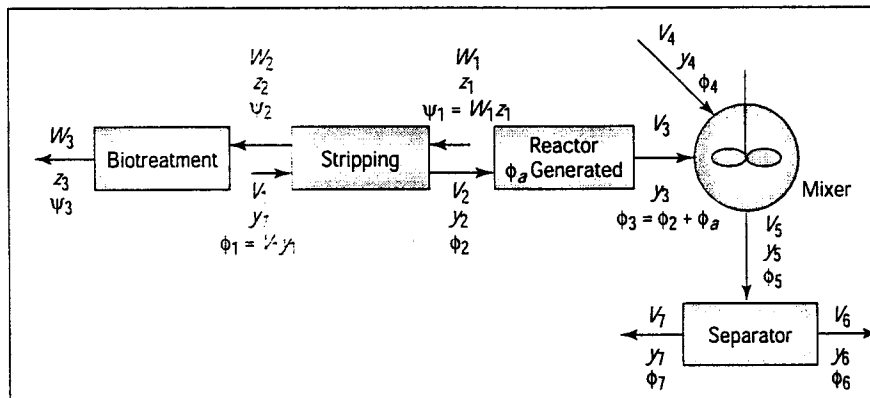


Figure 4. Loads and compositions of a targeted species throughout a process (3).

Figure 5. The gaseous path diagram (3).

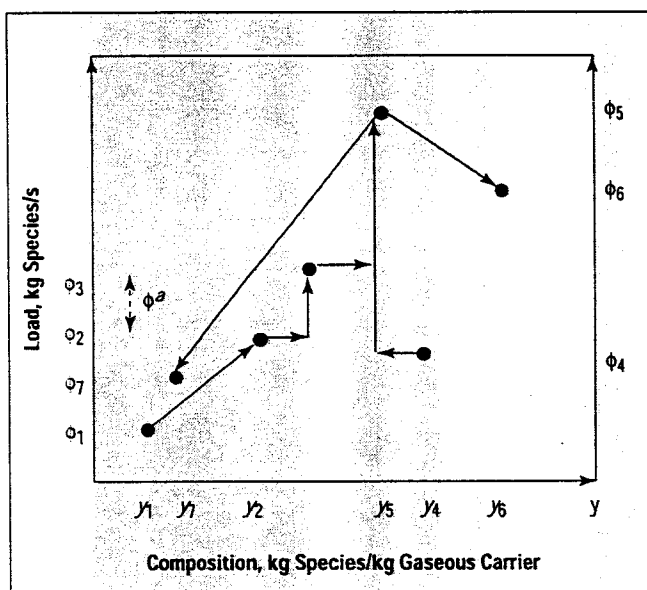
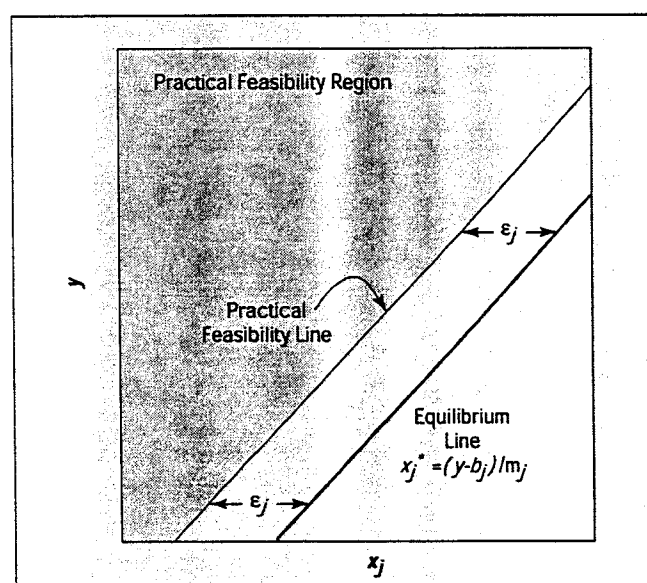


Figure 6. Establishing corresponding composition scales (19).



The path diagram can be used to determine where within the process a species should be intercepted, how much should be removed, and the global impact of manipulating sink/generators. Further, the path diagram shows the effect of manipulating any stream (represented by a node) on the rest of the process streams (nodes).

Species interception

At the heart of mass integration is the SPIN that intercepts streams and prepares them for their destinations.

Species interception can be accomplished by the use of MSAs (for instance, solvents, adsorbents, and the like) or ESAs (such as heating, cooling, and the application of pressure).

Mass transfer can be physical only or combined with reaction. The MSA can be a new material introduced into the process, or an existing process stream. If the MSA is a separate material, the network must include the regeneration of that material; if it is an existing process stream, the modified composition must be acceptable to the sink for that stream.

Physical mass-exchange networks. Let's first focus on direct-contact mass-transfer devices that use MSAs to selectively transfer species from one phase to another. The problem of synthesizing a MEN of this type has been introduced by El-Halwagi and Manousiouthakis (4) and can be stated as: Given N_R source streams rich in the species of interest and N_S candidate MSAs (streams lean in the species), design a network of mass exchangers that prepares all sources for the sinks. Examples of mass exchange falling into this category include stripping, solvent extraction, absorption, adsorption, leaching, and ion exchange.

Corresponding composition scales. Unlike heat transfer, mass transfer has a different equilibrium relationship for each MSA. And, to design a

MEN, all candidate technologies (MSAs) must be screened simultaneously. This has led to the concept of "corresponding composition scales" (CCSs). It allows designers to construct composition vs. mass exchange diagrams analogous to the temperature vs. enthalpy diagrams for heat transfer.

Consider a mass exchanger (with MSA, j) that is a candidate for removing the species of interest from a source stream, i . The following analysis applies to any type of phase equilibrium. However, to streamline the illustration, let us consider a linear equilibrium relationship, which is given by:

$$y_i = m_j x_j^* + b_j \quad (3)$$

This states that for a rich stream composition of y_i , the maximum theoretically attainable composition of the species in the MSA is x_j^* , where m_j is the slope and b_j is the intercept of the equilibrium line on a y_i - x_j diagram. This corresponds to a zero driving force. In practice, a positive driving force is needed to avoid having an infinitely large mass exchanger. This driving force is called the minimum allowable composition difference, ϵ_j . It is an optimization variable and is equivalent to the minimum driving force, ΔT_{min} , for heat transfer. The smaller the ϵ_j , the smaller the operating cost but the higher the fixed (capital) cost (4).

Therefore, the maximum allowable performance or "practical feasibility" occurs when (4):

$$y_i = m_j (x_j + \epsilon_j) + b_j \quad (4a)$$

or

$$x_j = [(y_i - b_j)/m_j] - \epsilon_j \quad (4b)$$

These relationships are depicted in Figure 6. Here, the equilibrium (Eq. 3) is plotted along with the practical feasibility line (Eq. 4) that defines the maximum allowable composition of the species in the j th MSA (x_j) for any composition of the species in the i th stream (y_i). Figure 6 also shows that the region below the practical feasibility line defines all acceptable mass-exchange operations.

Relationships of the form of Eq. 4

can be written for each MSA. These relationships define the CCSs for each of the MSAs:

$$x_j = [(y_i - b_j)/m_j] - \epsilon_j \quad (5)$$

where $j = 1, 2, \dots, N_j$

The significance of this is that, for a given set of CCSs, it is thermodynamically and practically feasible to transfer the targeted species from any rich stream into any MSA.

Constructing mass composite curves (4). The practical value of the CCSs now will be illustrated through the generation of mass composite curves.

As mass is transferred from a rich stream to a MSA, its composition decreases until the final targeted composition is reached. This relationship between composition and quantity of mass transferred can be plotted as shown in Figure 7. Here, two rich streams, R_1 and R_2 , are seen releasing mass and, in the process, reducing compositions from starting (or supply) values (y_1^s and y_2^s) to final target values (y_1^t and y_2^t). The quantities of mass transferred are MR_1 and MR_2 , respectively.

This is analogous to heat transfer where the temperatures of two streams are reduced from T_1^s and T_2^s to T_1^t and T_2^t . In the process, amounts of heat Q_1 and Q_2 are released.

The streams R_1 and R_2 of Figure 7 can be formed into a single rich composite stream as shown in Figure 8 by superposition, which involves performing a material balance in each composition interval. (Again, note the analogy between heat and mass transfer.)

Removing the targeted species from the rich streams requires one or more MSAs, all of which must be screened simultaneously based on thermodynamic feasibility and cost. This is done through the use of Eq. 5 to generate the correspondence between the rich composition scale, y , and the lean composition scales for all MSAs, the CCSs, as shown in Figure 9. Here, each MSA is represented on its own composition scale as a horizontal arrow extending between

its supply and target compositions. Feasibility of mass exchange is insured when mass is transferred from a rich stream to a lean stream that lies to its left.

While the locations of the MSAs have been determined, it still is necessary to establish their optimal flow rates. This involves finding the optimal slopes of all arrows representing MSAs on a mass vs. composition diagram. There are various graphical and mathematical techniques that facilitate this. The following is one useful technique (19).

Consider three MSAs, S_1 , S_2 and S_3 , whose operating costs (\$/kg of recirculating MSA) are c_1 , c_2 , and c_3 , respectively. These costs can be converted into \$/kg of removed species, c_j' :

$$c_j' = c_j / (x_j^i - x_j^f) \quad (6)$$

where $j = 1, 2, 3$

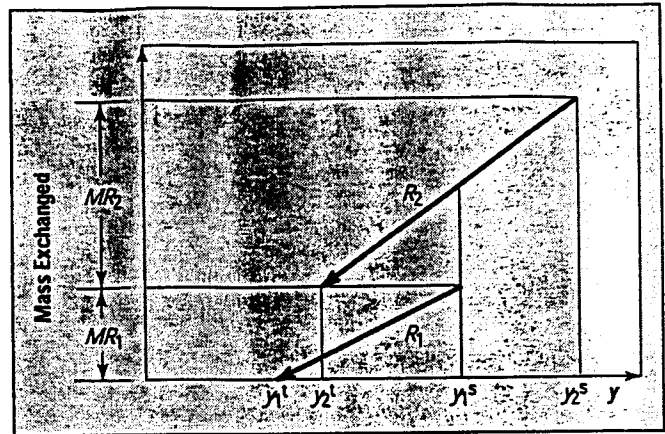
If arrow S_2 lies completely to the left of arrow S_1 , as is the case in Figure 9, and if c_2' is less than c_1' , one can eliminate S_1 from the problem, because it is thermodynamically and economically inferior to S_2 . On the other hand, if arrow S_3 lies completely to the left of arrow S_2 but c_3' is greater than c_2' , both MSAs 2 and 3 should be retained.

To minimize the operating cost of the network, separation should be staged to employ the cheapest MSA wherever feasible. Consequently, S_2 should be used to remove all of the rich load to its right, while the remaining rich load is removed by S_3 . The flow rates of S_2 and S_3 are calculated by simply dividing the rich load removed by the composition difference for the MSA.

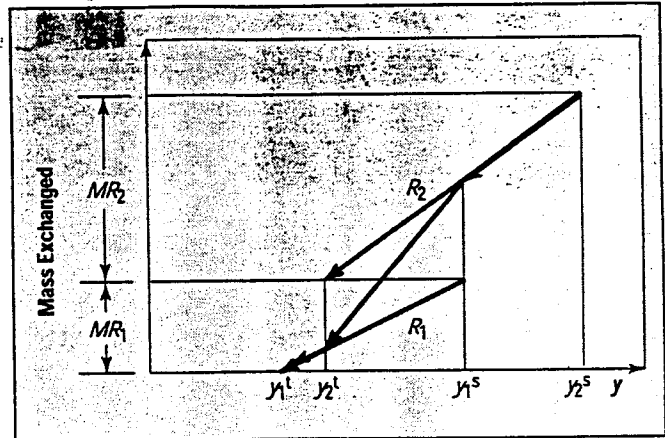
The mass pinch

Now that the MSAs have been screened and their flow rates have been determined, a lean composite curve can be constructed. This will show the mass to be removed by each MSA. See Figure 10. The sum of the operating costs of the selected MSAs is the minimum operating cost for the system. This is a rigor-

■ *Figure 7. Representation of mass exchanged by two rich streams.*



■ *Figure 8. Constructing a rich composite stream using superposition.*



ous target that has been determined ahead of design and without commitment to the final network configuration. It is important to emphasize that the fixed cost of the system is not overlooked. The identified target is based on a set of minimum allowable composition differences (ϵ_j). The parameter ϵ_j is an optimization variable that is iteratively varied to trade off the operating cost vs. the fixed cost.

The point where the two composite lines touch is termed the mass-exchange pinch point (4). (Hence, this type of drawing is called a "mass-exchange pinch diagram.") This point corresponds to the most thermodynamically constrained region of design and operation. At this point, all mass exchange takes place with a driving force equal to ϵ_j .

The mass-exchange pinch diagram can be used to identify and evaluate

structural changes to the MEN ahead of design. For example, the pinch location and, consequently, the minimum operating cost can be altered using three strategies:

1. The shape of the composite lines can be changed — for example, by altering the flow rates or compositions of the rich streams. This provides the designer with information about how the design parameters or operating conditions of the process should be modified to reduce cost.

2. The equilibrium relationships can be varied — for instance, by altering temperatures. This brings energy integration into the analysis.

3. ϵ_j can be changed. This provides a way of trading off capital and operating costs with the objective of minimizing the total annualized cost.

The subject of physical MENs has been addressed extensively. This includes MENs with a single transfer-

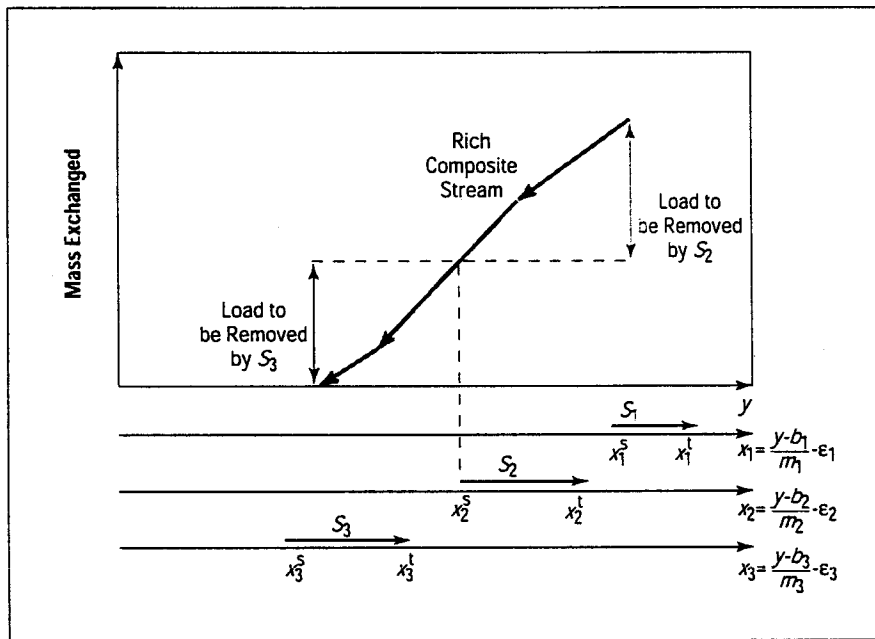


Figure 9. Screening MSAs (19).

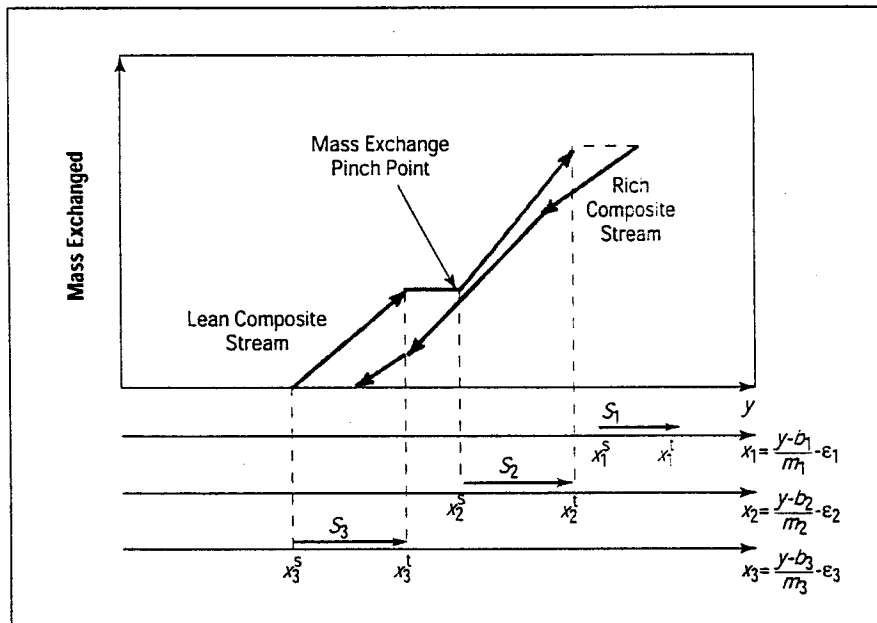


Figure 10. Constructing the mass-exchange pinch diagram.

able component (21), those with multiple transferable components (22), those involving regeneration of the MSAs (23), mass exchange combined with heat exchange (24), removal of fixed loads (25), MENs providing flexible performance (26,27), controllable MENs (28), and MENs with a

single lean stream (water) with the objective of minimizing water use (29). Furthermore, fixed cost targets have been identified graphically by developing a rich-composition/lean-composition composite diagram that optimizes the mass-exchange driving forces throughout the pinch diagrams

and predicts minimum column size ahead of detailed design (30).

Many industrial applications of species interception using MENs also have been published, including for pulp and paper (31), synthetic fuels (18,32), petrochemicals (33,34), polymers (35), and metal finishing (21). In addition, numerous examples illustrating the detailed application of MENs and mass integration to pollution prevention have been given in a recent textbook (19).

Reactive mass-exchange networks

In addition to physical mass exchange, reactive mass exchange often is an option for species interception. Indeed, Srinivas and El-Halwagi have pointed out that physical and reactive MSAs can be teamed to remove targeted species from the rich streams and to convert them into salable, reusable, or benign species (24,36,37). This involves combining mass-transfer equations with chemical reaction equilibria to develop equilibrium expressions of the form:

$$y_i^* = f_j(x_j^*) \quad (7)$$

where i is the rich phase, j is the lean phase, y_i^* is the rich-phase equilibrium composition of the targeted species, and x_j^* is the lean-phase equilibrium composition of all species resulting from the chemical reactions of the targeted species. Incorporating the minimum allowable composition differences for the rich and the lean phases (ϵ_j^R and ϵ_j^S , respectively) to trade off capital vs. operating cost gives:

$$y = f(x_j + \epsilon_j^S) \quad (8a)$$

where

$$y = y_i - \epsilon_i^R \quad (8b)$$

Equations 8a and 8b provide the relationship between the rich and lean composition scales for which reactive mass exchange is practically feasible. This is the reactive equivalent to Eqs. 4a and 4b used for establishing the corresponding composition scales for physical MENs.

These concepts now can be used to develop a reactive mass-exchange

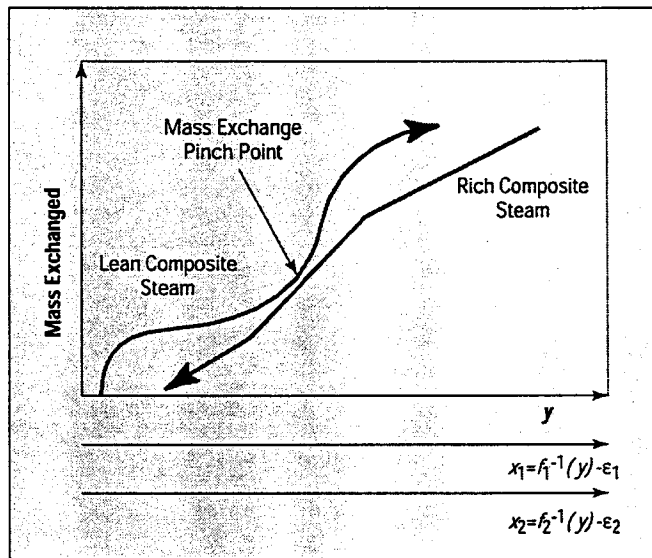


Figure 11. The reactive mass-exchange pinch diagram.

pinch diagram (Figure 11) for screening all candidate MSAs to identify the lowest cost target for reactive interception networks.

Heat-induced separation networks

Another class of separation networks relies on ESAs to induce separation. These heat induced separation networks (HISENs) involve heating or cooling to effect separation through a phase change such as condensation, evaporation, or crystallization (38-43). Other examples of ESA-induced interception include pressure-driven membrane separations (44,45), and distillation sequences (46,47).

To design HISENs, we first must transform mass separation into a heat-transfer task. This can be done by relating phase change to temperature. For instance, as a gas laden with a volatile organic compound (VOC) is cooled, the composition of the VOC remains constant until condensation starts at the dew point, T^c , after which a phase change occurs. For a dilute system, the composition of the VOC can be expressed as a function of temperature:

$$y(T) = y^s \text{ if } T > T^c \quad (9a)$$

$$y(T) = p^o(T) / [P^{total} - p^o(T)] \text{ if } T \leq T^c \quad (9b)$$

where y^s is the supply (inlet) mole

fraction of the VOC in the gas, $p^o(T)$ is the vapor pressure of the VOC expressed as a function of the gas temperature, T , and P^{total} is the total pressure of the gas. For a given target composition of the VOC, y^t , the VOC recovery task is equivalent to cooling the stream to a separation target temperature, T^* , which is calculated via:

$$y^t = p^o(T^*) / [P^{total} - p^o(T^*)] \quad (10)$$

For concentrated multicomponent VOC systems, nonideal equilibrium calculations (20) can be performed in lieu of Eqs. 9 and 10.

VOC recovery now has been con-

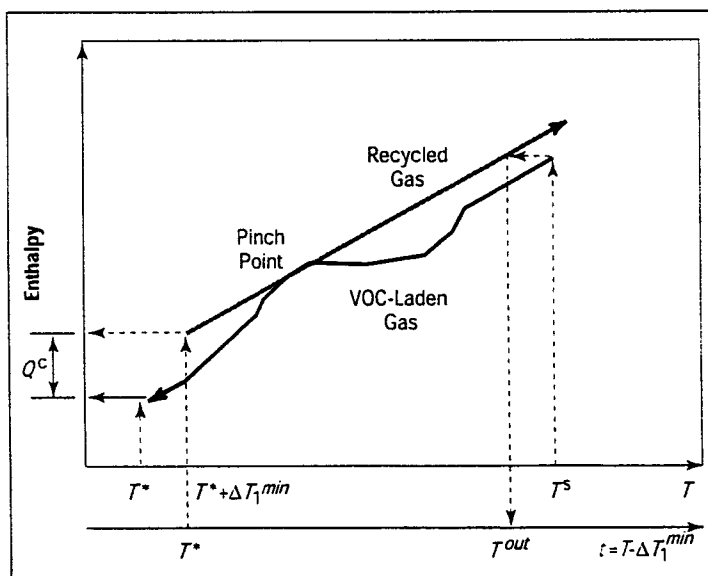
verted into a heat-transfer task. The target for the minimum cooling/refrigeration duty, Q^c , can be found using a thermal pinch diagram as shown in Figure 12 (48). For multiple rich streams and multiple cooling utilities, similar pinch representations can be developed (40).

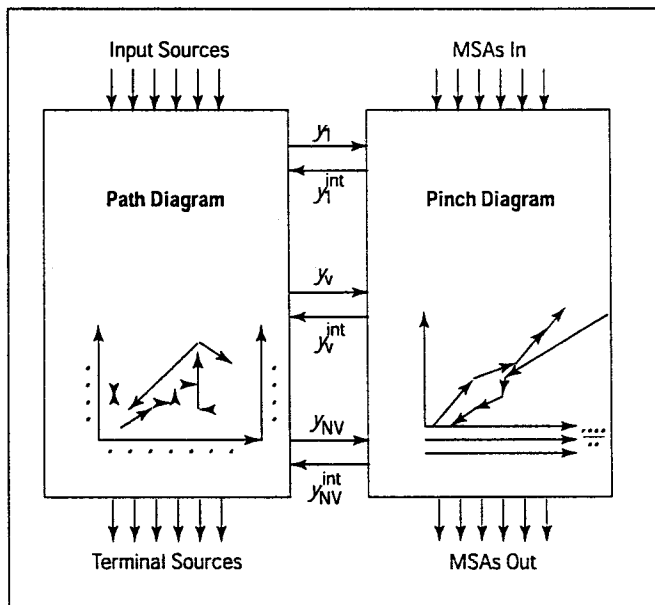
Integrating path and pinch diagrams

So far, we have assumed that the designer knows which streams should be intercepted and what extent of separation is needed. These questions can be answered systematically by combining the concepts of the path and mass pinch diagrams. The path diagram tracks the consequences of any interception. On the other hand, the pinch diagram provides insights on the optimal interception strategy for given streams and separation tasks. Put together, the two diagrams can clarify which streams are to be separated and the extent of separation. The relationship between the path and pinch diagrams is shown in Figure 13, which illustrates the back-and-forth passage of stream information between the two diagrams.

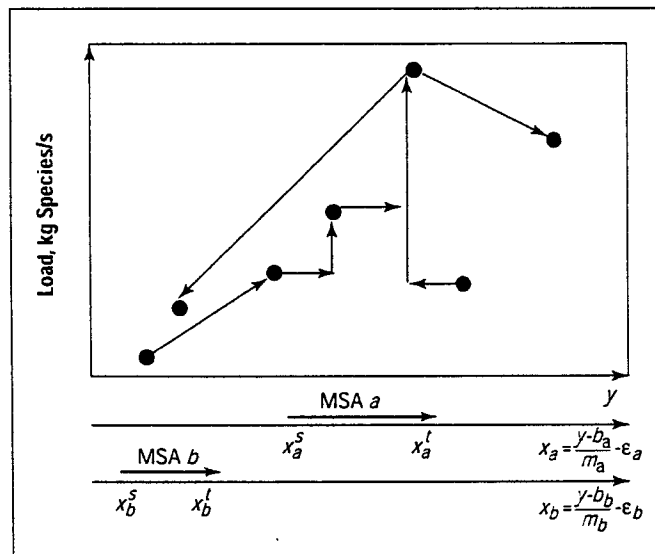
The path and pinch diagrams can be integrated via a MINLP. But, a short-cut graphical method, as de-

Figure 12. Pinch diagram for a VOC condensation system (48).





■ **Figure 13.**
Integration of the path and pinch diagrams (3).



■ **Figure 14.**
Using a hybrid of the path and pinch diagrams to screen MSAs for intercepting sources (3).

picted in Figure 14, is a handy alternative. After plotting the path diagram for the sources, the pinch diagram is superimposed through use of its CCSs (for instance, Eq. 5). Each MSA is represented on its own scale as a horizontal arrow extending between its supply and target compositions.

Useful insights can be gained from this hybrid diagram. Consider two MSAs, *a* and *b*, whose costs (\$/kg of recirculating MSA) are c_a and c_b , respectively. These costs can be converted into \$/kg of removed species,

c_a' and c_b' , using Eq. 6. If the arrow for MSA *b* lies completely to the left of the arrow for MSA *a* and c_b' is less than c_a' , MSA *b* is chosen in favor of MSA *a*, because it is thermodynamically and economically superior. In addition, the relative locations of the sources (nodes) and the MSAs provide helpful information. If a MSA lies to the right of a source node, that MSA is not a candidate for intercepting this node, because mass exchange is not feasible. Analogous observations can be made for systems employing ESAs (41).

APPLICABILITY BEYOND DESIGN

While this article focuses on design-related applications of mass integration, the approach can be employed effectively in other areas such as process operation and simplification.

Process operation

Mass integration tools can be revised to tackle operational issues (26–28,49), including cost reduction and performance improvement. For instance, on-line optimization strategies can be based on a holistic understanding of process performance (instead of the typical approach of using pre-set criteria). Mass integration, therefore, can lead to optimal response to continuous process disturbances and variations (49). It also can improve the dispatch and management of process resources such as raw materials, fuels, material utilities, electric power, and MSAs.

Process simplification

Developing conceptual flowsheet alternatives that employ the least number of processing steps is an important objective during research and development or front-end engineering design (FEED). This exercise can yield useful feedback to the R&D team, preventing the generation of poor process configurations, while incorporating techno-economic issues early in the design cycle. For FEED, it safeguards against the automatic replication of the previous design and provides a fresh perspective into the process. Mass integration tools can be used effectively to generate the simplest process alternatives (50).

One of the key concepts is the notion of maximum extendible zones (MEZs), which define the largest design and operating space of a technology. They can be developed based on fundamental principles of chemical engineering, such as the practical feasibility region for mass exchange (see Figure 6)

Stream	Description	Supply Composition x_j^s , ppmw	Target Composition x_j^t , ppmw	m_j	ϵ_j , ppmw	C_j , \$/kg MSA	C_j^r , \$/kg CE removed
SV_1	Polymeric resin	2	10	0.03	5	0.08	10,000
SV_2	Activated carbon	5	30	0.06	10	0.10	4,000
SV_3	Oil	200	300	0.80	20	0.05	500

Stream	Description	Supply Composition X_j^s , ppmw	Target Composition X_j^t , ppmw	M_j	ϵ_j , ppmw	C_j , \$/kg MSA	C_j^r , \$/kg CE removed
SW_1	Zeolite	3	15	0.09	15	0.70	58,333
SW_2	Air	0	10	0.10	100	0.05	5,000
SW_3	Steam	0	15	0.80	50	0.12	8,000

and for ideal/nonideal distillation (46,47), and the attainable regions for reactors (16). MEZs can be established ahead of detailed design without specifying the process configuration.

Once these zones are developed, the task of synthesizing the simplest process can be transformed into identifying a scheme with the least number of "jumps" that connect the raw materials to the products, byproducts, and all acceptable process outlets via these MEZs. (See Figure 15.) First, only the key process species are considered. Later, the other species are added. Graphical and mathematical tools can be designed to determine these configurations (50). Once the process with the least number of processing steps is established, it is eliminated from the analysis, and the next-to-simplest alternative then is generated. Repeating this exercise provides a roster of the simplest alternatives. Next, simulation, cost optimization, and detailed design studies are undertaken to refine the conceptual alternatives.

USING THE APPROACH

Let's now look at how to actually apply mass-integration design tools by taking the ethyl chloride process (3,19) as an example.

Ethyl chloride (C_2H_5Cl) can be manufactured by catalytically reacting ethanol and hydrochloric acid as shown in Figure 16. This involves two integrated processes, for ethanol and ethyl chloride.

Ethanol is made by the catalytic hydration of ethylene. Compressed ethylene is heated with water and reacted to form ethanol. Ethanol is separated using distillation followed by membrane separation (pervaporation). Aqueous waste is fed to a biotreatment facility.

Purified ethanol is reacted with hydrochloric acid in a multiphase reactor to form ethyl chloride. The reaction takes place primarily in the liquid phase. Chloroethanol (CE), C_2H_5OCl , is generated as a byproduct according to the following zero-order reaction:

$$r_{\text{oxychlorination}} = 6.03 \times 10^{-6} \text{ kg CE/s} \tag{11}$$

Off-gas from the reactor is scrubbed with water in two scrubbers. The first recovers the majority of unreacted ethanol, hydrogen chloride, and CE. The second removes traces of unreacted materials to yield product ethyl chloride.

The aqueous streams leaving the scrubbers are mixed and recycled to the reactor, where a fraction of the recycled CE is reduced to ethyl chlo-

ride. The rate of CE depletion in the reactor is approximated by:

$$r_{\text{reduction}} = 0.090 z_5 \text{ kg CE/s} \tag{12}$$

where z_5 is the mass fraction in the liquid phase.

The distribution of CE between the gaseous and liquid phases in the ethyl chloride reactor is given by the following distribution relationship:

$$y_1/z_6 = 5 \tag{13}$$

The design problem

Ethyl chloride is one of the least toxic of all chlorinated hydrocarbons. CE, however, is a toxic pollutant that is causing significant problems for the biotreatment facility. The design

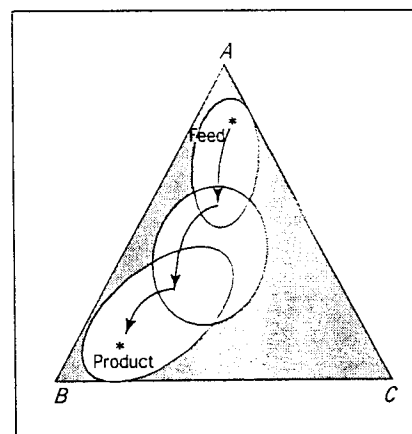
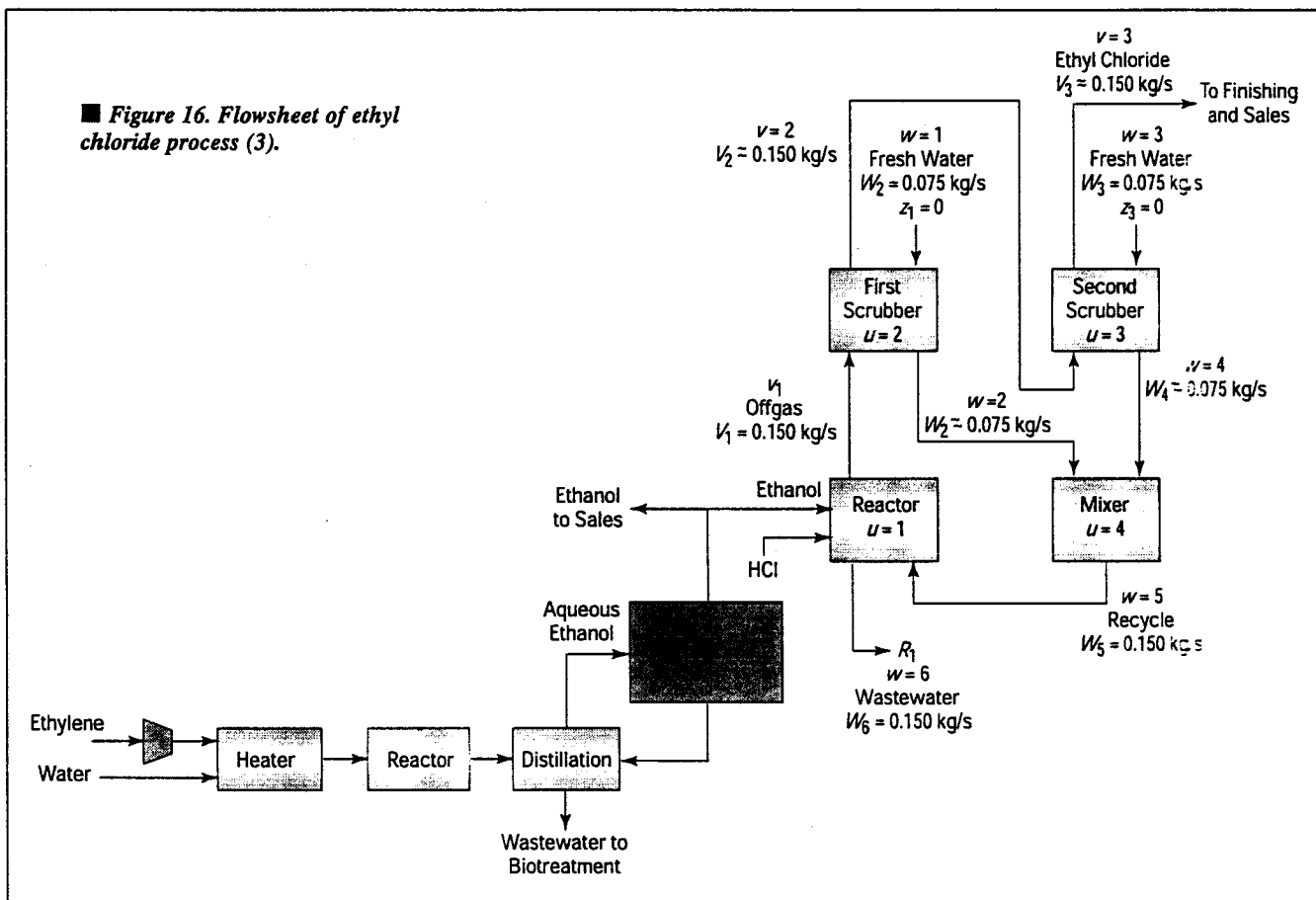


Figure 15. Least number of "jumps" among maximum extendible zones (50).

■ Figure 16. Flowsheet of ethyl chloride process (3).



objective is to reduce the CE content of R_1 (Figure 16) to meet the following targets:

composition:

$$z_6^{int} = z_1^{terminal\ target} = 7 \text{ ppmw} \quad (14)$$

load:

$$\text{CE in } R_1 = 1.05 \times 10^{-6} \text{ kg/s} \quad (15)$$

This design problem involves the reactor, first scrubber, second scrubber, and mixer (u_1 , u_2 , u_3 , and u_4), three gaseous sources/nodes (v_1 , v_2 , and v_3), and six liquid sources/nodes (w_1 , w_2 , w_3 , w_4 , w_5 , and w_6). The flow rates of all gaseous and liquid sources are shown on Figure 16.

Six MSAs have been suggested as candidates for removing CE — three for gaseous streams, and three for liquid streams. Tables 2 and 3 provide the data.

The first step in finding a solution is to develop the path diagram. The path diagram equations can be de-

rived from basic unit performance considerations as shown by El-Halwagi and coworkers (3). These are summarized as follows (with all compositions in ppmw):

For $u = 1$:

$$0.180 y_1 - 6.030 = 0.060 z_5 \quad (P1)$$

$$y_1 - 5 z_6 = 0$$

For $u = 2$:

$$2 y_2 + z_2 = 2 y_1$$

$$y_2 = 0.10 y_1$$

For $u = 3$:

$$2 y_3 + z_4 = 2 y_2$$

$$y_3 = 0.10 y_2$$

For $u = 4$:

$$2 z_5 = z_2 + z_4$$

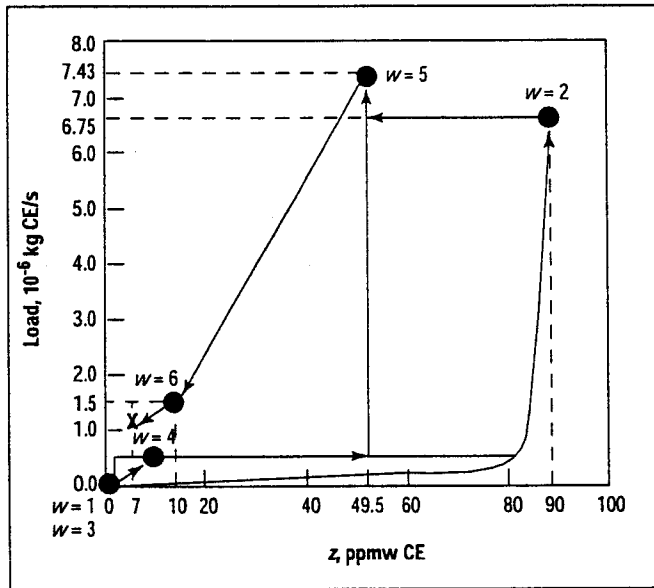
Figure 17 is the resulting path diagram for CE in the liquid phase.

The objective is to move the terminal wastewater node ($w = 6$) to the targeted location (designated by an "X"). The challenge is to determine how the other nodes should be ma-

nipulated (intercepted) to meet this target at minimum cost.

Interception is incorporated by constructing a hybrid path-pinch diagram as shown in Figure 18. Thermodynamic feasibility and cost are used to prescreen interception strategies for each node. For example, as air lies to the left of steam and has a lower cost (per kg CE removed), it is chosen over steam. Nodes lying to the right of air ($w = 2$, $w = 5$) can be intercepted down to 10 ppmw CE using air stripping. If a lower level is required, CE first should be removed by air (until 10.0 ppmw CE is reached), followed by zeolite. Similarly, nodes $w = 4$ and $w = 6$ can be intercepted by zeolite down to 1.6 ppmw CE.

Figure 19 shows the combined path-pinch diagram for the gaseous nodes and candidate MSAs. Because



■ **Figure 17.** The liquid path diagram for CE with "X" showing the waste reduction target (19).

screened candidate MSAs with the path diagram. This is done by revising the path diagram equations (P1) to include potential interception at all nodes. The result (see Ref. 19) is:

$$0.180 y_1 - 6.030 = 0.0060 z_5^{int} \quad (P2)$$

$$y_1 - 5 z_6 = 0$$

$$2 y_2 + z_2 = 2 y_1^{int}$$

$$y_2 = 0.10 y_1^{int}$$

$$2 y_3 + z_4 = 2 y_2^{int}$$

$$y_3 = 0.10 y_2^{int}$$

$$2 z_5 = z_2^{int} + z_4^{int}$$

These equations can be solved using an optimization program to minimize the cost of meeting environmental requirements. The solution will identify which nodes are to be intercepted, which MSAs are to be used, and how much mass should be removed from each node.

it is not thermodynamically feasible, oil cannot be used to intercept any node. Activated carbon should be used to intercept nodes $v = 1$ and $v = 2$ down to 0.9 ppmw CE. Any inter-

ception below 0.9 ppmw CE can be handled by polymeric resin, which can reduce the CE content to 0.21 ppmw, if required.

We now must integrate the pre-

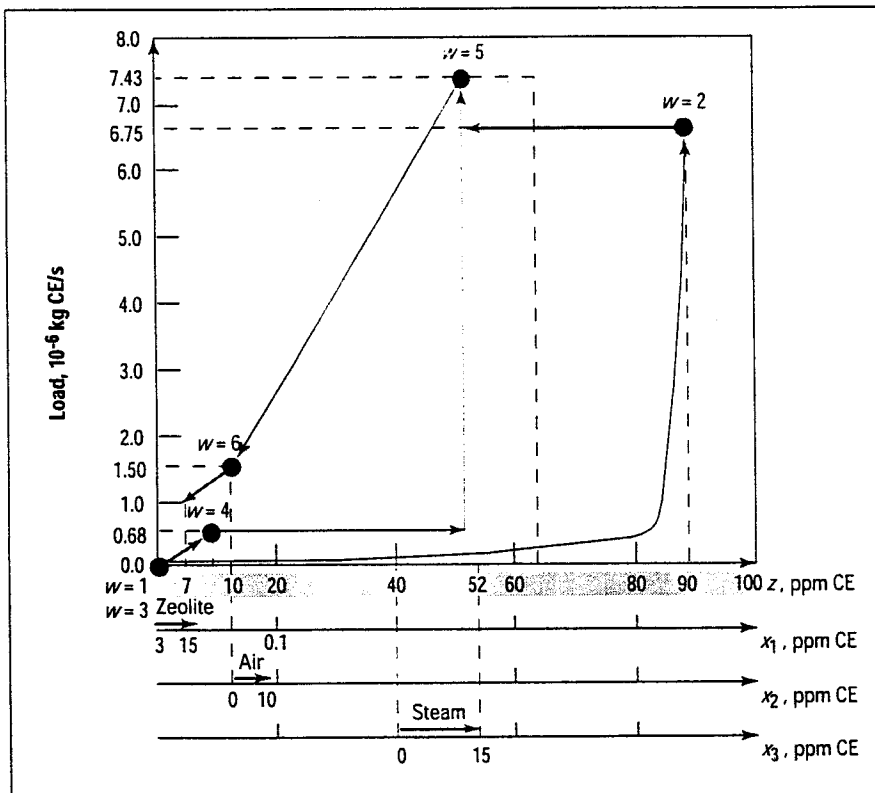
This procedure can be illustrated graphically for the case where only one node is intercepted at a time. (Simultaneous interception of multiple sources may be considered by allowing intercepted compositions of multiple nodes to be treated as optimization variables in the same formulation. See Ref. 3 for more details.) For example, if the gaseous node $v = 1$ (reactor offgas) is to be intercepted, its composition must be reduced to 4.6 ppm CE to achieve a 7 ppm composition of CE in the terminal wastewater stream. Using Figure 19, activated carbon is found to be the optimal MSA for this purpose. Based on a material balance and the data provided in Table 2, the cost of this interception is approximately \$576,000/y.

The procedure can be repeated for the other gaseous and liquid nodes. By comparing the costs of all alternatives, the optimal solution is found to be activated carbon adsorption to remove 4.57×10^{-6} kg CE/s from the reactor off-gas at a cost of \$576,000/y. An ordered list of next-to-best solutions also can be generated.

Key observations

This example points up several important facets always to keep in mind:

- Chemical processing is a multi-



■ **Figure 18.** Prescreening MSAs for intercepting the liquid path diagram (19).

media (multiphase) problem. Removing a species in one medium can be less expensive than in another medium. For example, a given quantity of chloroethanol may be eliminated at a lower cost from an in-process gaseous stream than from wastewater. Indeed, the optimal solution to a wastewater problem may lie in the gas phase.

- In-plant interception often is superior to terminal waste separation. For instance, separating CE from the terminal wastewater stream incurs an annual operating cost of \$828,000 — 44% more than the best solution.

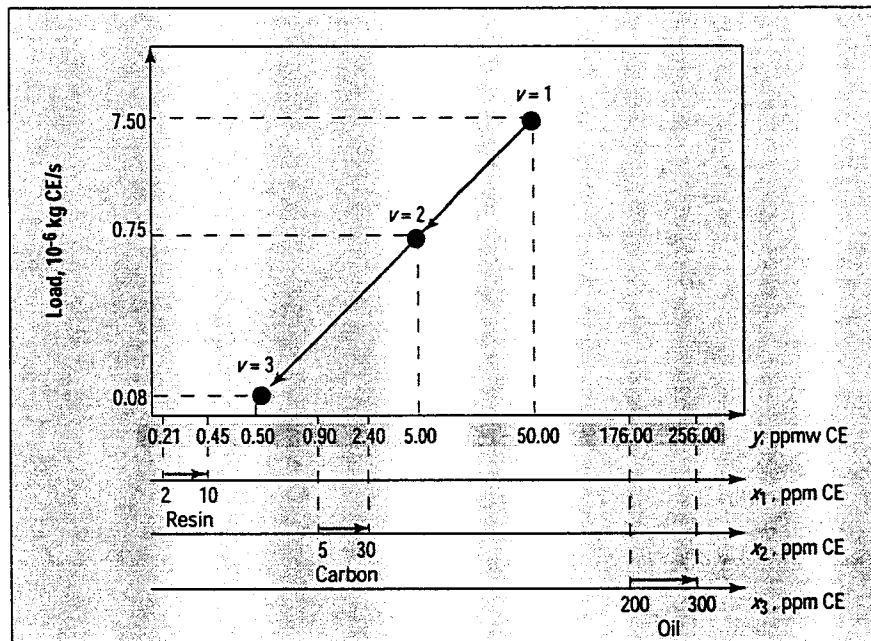
- Removing more mass needn't mean higher cost. For example, intercepting $v = 1$ involves eliminating ten times more CE than the more expensive interception of $w = 6$ (4.57×10^{-6} kg CE/s vs. 0.45×10^{-6} kg CE/s, respectively). This is because removing the pollutant from more concentrated streams is thermodynamically favored. It generally is less expensive to use low cost MSAs to eliminate the species from in-plant streams than to employ high cost MSAs to remove traces of the species from the dilute terminal streams.

- Propagation of mass through the process is nonlinear. For a specified reduction from a terminal stream, different masses may be removed from different in-plant streams. This can work to the designers' advantage.

Extending the scope

Our example has focused on in-plant interception to illustrate using a combined path-pinch diagram. Let's now see how other mass-integration strategies, including stream segregation, mixing, and direct recycle, can be used to generate even better solutions.

Suppose we wish to reduce the composition of CE to one-third its current value (that is, to 2.3 ppmw rather than 7.0 ppmw), while simultaneously decreasing the flow rate of the discharged wastewater to one-half its present value — an even more challenging problem than the one just solved.



■ Figure 19. Prescreening MSAs for intercepting the gaseous path diagram (19). (Note: not to scale.)

The solution is generated using the general framework shown in Figure 1. This brings in stream segregation, recycle, and sink/generator manipulation as additional design tools (19).

The resulting solution (see Figure 20) is to segregate the effluents of the two scrubbers, recycle the aqueous effluent of the reactor to the first scrubber, recycle the distillation bottoms to the second scrubber, and use activated carbon to intercept the gaseous stream from the first scrubber ($v = 2$) to reduce its composition to $y_2^{int} = 1.8$ ppm CE. The annual operating cost of this solution is approximately \$32,000.

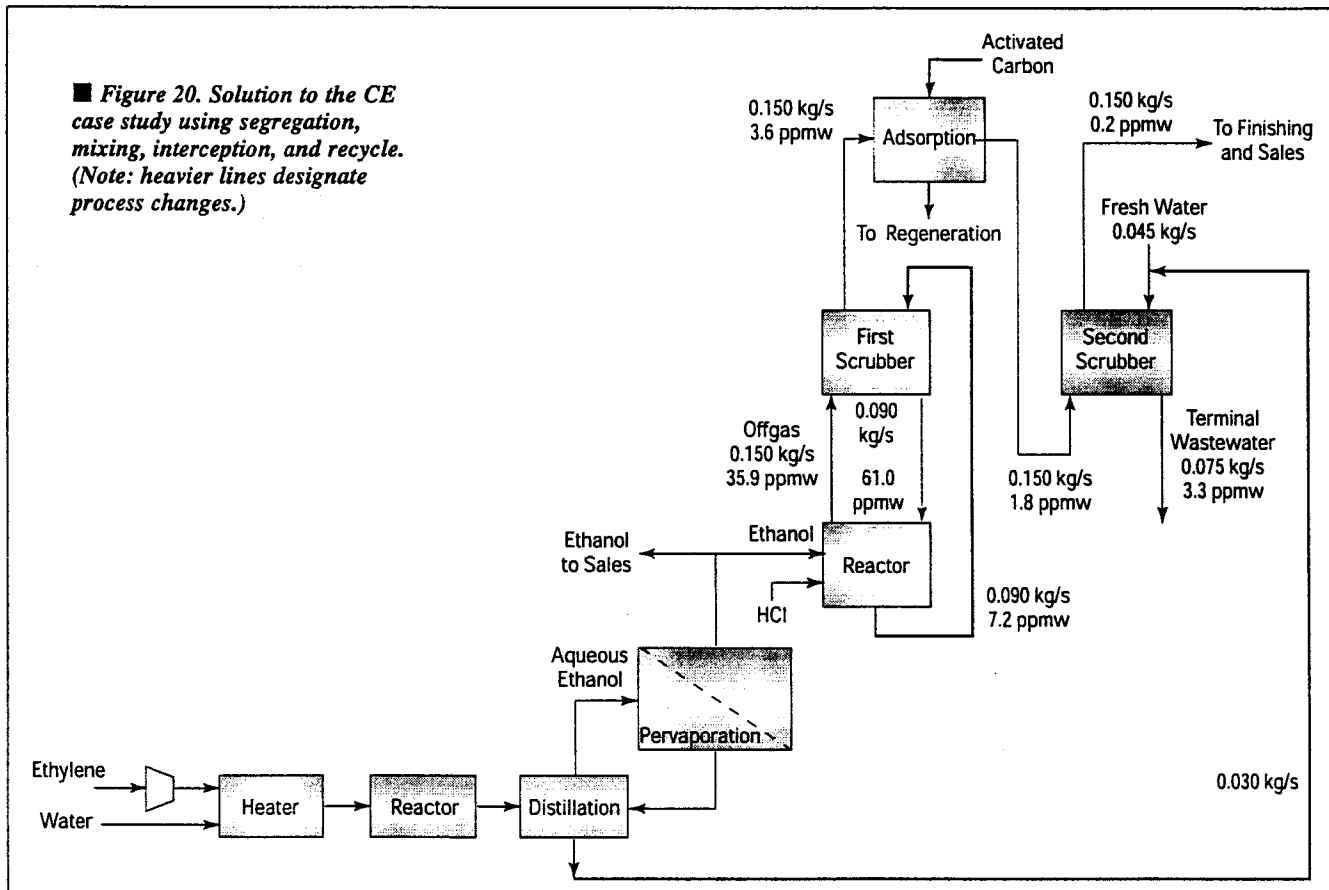
This can be compared with other solutions, including end-of-pipe treatment. Suppose we retain the segregation, mixing, and direct recycle strategies shown in Figure 20, but intercept the wrong stream. For example, if the terminal wastewater stream is intercepted rather than the gaseous stream from the first scrubber, the least expensive MSA that is practically feasible is zeolite (as can be seen from Figure 18). This end-of-pipe separation (after segregation, mixing, and recy-

cle) requires an annual operating cost of \$441,500, or 14 times more than the best solution. If end-of-pipe separation is employed without segregation, mixing, and recycle, the targeted limits for CE can be met by using zeolite (again see Figure 18) — but at an operating cost of about \$2.3 million/yr. While these solutions meet the CE loading target and provide the same waste-reduction effect as the optimal solution, they are significantly inferior to the solutions obtained using the full range of mass integration techniques.

Proven in practice

Our company, Matrix Process Integration, has directed numerous industrial applications of the integration methods described in this article. We have worked on projects for improving profitability and cutting operating and capital costs; developing process technology and enhancing process reliability and flexibility; increasing product yields, capacity, and quality; reducing the use of water, energy, materials, and solvents; and achieving environmental compliance. Most of these projects involved meeting

■ **Figure 20. Solution to the CE case study using segregation, mixing, interception, and recycle.** (Note: heavier lines designate process changes.)



many of these goals simultaneously.

Three of these projects are described in the accompanying box, to provide a more complete picture of typical projects, including scope, customer requirements, and results.

Our experience clearly has demonstrated that:

- Integration methods are highly useful for a wide range of processing facilities. We have applied the techniques in a broad variety of sectors within the chemical process industries (CPI) — including the chemical, petrochemical, refining, pulp and paper, pharmaceutical, food and beverage, and related industries — with uniform success.

- The techniques support both business and technical objectives. This makes them valuable tools both for management, who are interested in planning, profit improvement, cost reduction, and better utilization of capital and other resources, and for engineers, who must design and oper-

ate facilities to support their company's strategic direction.

- All projects have multiple objectives. This illustrates the integrated nature of problems and their solutions. And, it emphasizes the importance of having tools that allow a global understanding of these complex facilities and their design and operating issues.

- Integration significantly outperforms traditional approaches to design. It leads to a better selection of process improvement projects, and pinpoints those with the best payback.

THE OPPORTUNITIES

Engineers long have striven to design plants that best address all important technical and economic issues. Their design tools (simulators and other programs) have advanced significantly over the last two decades. So, do substantial process improvement opportunities still exist? Our experience dramatically shows

that they do — for three main reasons: organization, changing business requirements, and integration tools.

Organization. CPI manufacturing facilities are capital intensive and complex. And, complexity naturally leads to a subdivision of work to allow individuals or teams to focus on small, manageable portions of the overall mission. This is true of management and of technology, as well.

The management of a company typically is organized both along product lines and by functions. An integrated production facility might have several product lines on-site, each directed by a separate business team. Staff running the plant usually are responsible for specific functions, such as operations, maintenance, plant engineering, utilities, and health, safety, and the environment.

The technologies employed are supported by technical specialists who are experts in narrow fields. In a

refinery, for instance, one or two individuals might understand the cracking furnaces, another the refining section, another the biotreatment facility, and yet another the boiler house.

The consequence of this is that both the management and the technology of a company are understood only as the sum of many small component parts. No one comprehends the whole and the interactions among the parts. Op-

portunities are lost at the interfaces: a new biotreatment facility is constructed although process changes could have eliminated the need; a new boiler is purchased although improved heat recovery could have made it unnecessary; and a new take-or-pay contract for fuel is negotiated although the need for more fuel could have been eliminated through process efficiencies.

Changing business requirements.

Production sites are dynamic, ever-evolving entities. Change is constant.

New processes continually are being planned, designed, and constructed. Old product lines are being discontinued. Existing processes are being expanded and debottlenecked. And, actions are being taken to meet environmental and other regulations.

Even if a site is properly integrated and optimized today, it will not be tomorrow. Change creates a requirement to maintain the total site as an integrated system. This is typically not done, resulting in inefficiencies and unrealized opportunities.

Integrated tools. Processes and sites are complex, integrated systems. Understanding them as such requires tools other than the traditional process simulators and unit-operations design programs.

The mass and energy integration tools described in this article (especially those for mass integration) are just becoming available for widespread use. They allow engineers and others to see the big picture, to understand the interactions, and to devise design and operating strategies that realize the intrinsic opportunities.

Areas of application

The use of process integration methods should be viewed strategically. After all, they address the entire range of a company's processing objectives and, so, should play a key role in the development and implementation of corporate strategy. We have found the following to be particularly rich areas of application.

- *Aligning technology with corporate strategy.* There often is a gap between a company's strategic direction and its technology base. Strategy and technology must be developed together. This frequently is a chicken-and-egg problem. Developing strategy requires an understanding of the technical possibilities, while developing technology demands a clear statement of strategy.

- *Planning site development.* Once the overall strategy has been decided, it is necessary to implement it effective-

Some successful applications

Large integrated petrochemical complex

Project scope:

- Develop strategic plans for site expansion and infrastructure development, including electricity supply/distribution, fuel-gas/natural-gas supply/distribution, vents/purges handling, and feedstock supply.
- Reduce costs.

Requirement: The expansions included debottlenecking existing processing equipment, shutting down an obsolete unit, and adding a new product line. In addition, site infrastructure improvements to accommodate the expansions had to reduce overall production costs, meet all environmental regulations, allow for startups and shutdowns of individual units, be robust, and provide a path for future expansions over a 10-y horizon.

Results: Alternative strategies were developed addressing all requirements. Strategies ranged from minimum cost/minimum benefit to more substantial investments to achieve the maximum benefits. The company now is implementing the site development plan that meets its goal of being the low cost producer.

Specialty chemicals plant

Project scope:

- Reduce water use.
- Decrease organic load to biological treatment.
- Increase product capacity.
- Debottleneck cooling system.
- Cut solvent loss.

Requirement: The unit under investigation was the largest polluter on-site and would have to pay for a large portion of a new water-treatment facility. It was desired to reduce both the volume of water and the contaminant load, increase capacity, which was limited by a brine cooling system, and cut yield losses. The project was combination of pollution prevention, reduction of solvent and product loss, yield improvement, and energy integration. All of these were interrelated.

Results: A strategy was developed that involved: (1) energy storage to recover and reuse refrigeration; (2) recycle and recovery of solvents and product; and (3) water conservation and reuse. Yield was increased by 7.5%, the brine system was debottlenecked, solvent loss was cut by one-quarter, and water use trimmed by one-third. Payback was substantially less than one year.

Chemical and polymers complex

Project scope:

- Meet regulations.
- Decrease thermal discharge.
- Recover VOCs.

Requirements: Two interrelated problems had to be addressed: (1) the heat load going to biological treatment was too high, resulting in discharge temperatures exceeding local regulations; and (2) VOCs were leaving the process in vent and water streams, limiting the ability to expand capacity. The company wished to meet regulations, recover heat and VOCs, and allow expansion. Ideally, it hoped that the recovered heat and VOCs would pay for the required investment.

Results: The thermal load was reduced by over 90% and VOC losses were cut by over 50% through a combination of operating changes and retrofit projects. The package had a simple payback of 1-1½ y.

ly. This involves integrating new/revised processes into the site from a material flow perspective and modifying site infrastructure (for instance, the fuel, steam, power, water, and plant air systems) to accommodate the revised production requirements.

- *Developing new process technologies.* Integration methods can be used to plan and direct research and development, and to design processes as they emerge from R&D. This will assure that the next generation of technologies is strong and competitive.

And, it will materially shorten the process-development time cycle.

- *Designing expansions and improving existing processes.* Current capabilities must be continually upgraded. This involves many small to large projects to enhance yields, increase capacities, improve product quality, reduce operating costs, boost profits, and the like.

- *Meeting regulations.* Environmental and other regulations require that companies develop and implement strategies to stay in compliance now —

and over the long term. This may involve reducing gaseous emissions (VOCs, NO_x, SO_x), water use, water flow and load to biotreatment, deep-well injection, waste disposal via boilers, and the amount of sludge and solid waste sent to landfills.

- *Selecting and conserving resources.* The use of resources (such as feedstocks, solvents, water, and energy) is both an economic and a sustainability issue. They must be selected and used wisely for the short- and long-term viability of the business.

Literature Cited

1. Linnhoff, B., "Use Pinch Analysis to Knock Down Capital Costs and Emissions," *Chem. Eng. Progress*, **90** (8), pp. 32-57 (Aug. 1994).
2. El-Halwagi, M. M., and H. D. Spriggs, "An Integrated Approach to Cost and Energy Efficient Pollution Prevention," *Proceedings, 5th World Congr. of Chem. Eng. (San Diego)*, III, AIChE, New York, pp. 344-349 (1996).
3. El-Halwagi, M. M., A. A. Hamad, and G. W. Garrison, "Synthesis of Waste Interception and Allocation Networks," *AIChE J.*, **42** (11), pp. 3,087-3,101 (Nov. 1996).
4. El-Halwagi, M. M., and V. Manousiouthakis, "Synthesis of Mass Exchange Networks," *AIChE J.*, **35** (8), pp. 1,233-1,244 (Aug. 1989).
5. Hohmann, E. C., "Optimum Networks for Heat Exchanger," PhD Thesis, Univ. of Southern California, Los Angeles (1971).
6. Linnhoff, B., and E. Hindmarsh, "The Pinch Design Method for Heat Exchanger Networks," *Chem. Eng. Sci.*, **38** (5), pp. 745-763 (1983).
7. Floudas, C. A., A. R. Ciric, and I. E. Grossmann, "Automatic Synthesis of Optimum Heat Exchange Network Configurations," *AIChE J.*, **32** (2), pp. 276-290 (Feb. 1986).
8. Bagajewicz, M. J., and V. Manousiouthakis, "Mass-Heat Exchange Network Representation of Distillation Networks," *AIChE J.*, **38** (11), pp. 1,769-1,800 (Nov. 1992).
9. Friedler, F., J. B. Varga, and L. T. Fan, "Algorithmic Approach to the Integration of Total Flowsheet Synthesis and Waste Minimization," *AIChE Symp. Ser.*, **90** (303), AIChE, New York, pp. 86-97 (1995).
10. El-Halwagi, M. M., "Introduction to Numerical Optimization Approaches to Pollution Prevention," in "Waste Minimization through Process Design," A. P. Rossiter, ed., McGraw-Hill, New York, pp. 199-208 (1995).
11. Garrison, G. W., H. D. Spriggs, and M. M. El-Halwagi, "A Global Approach to Integrating Environmental, Energy, Economic and Technological Objectives," *Proceedings, 5th World Congr. of Chem. Eng. (San Diego)*, I, AIChE, New York, pp. 675-680 (1996).
12. Joback, K. G., and G. Stephanopoulos, "Designing Molecules Possessing Desired Physical Property Values," *Proceedings, 3rd Conf. on Found's of Comp. Aided Proc. Des.*, pp. 363-387, Elsevier, New York (1989).
13. Constantinou, L., C. Jacksland, K. Bagherpour, R. Gani, and L. Bogle, "Application of Group Contribution Approach to Tackle Environmentally-Related Problems," *AIChE Symp. Ser.*, **90** (303), pp. 105-116 (1995).
14. Vaidyanathan, R., and M. M. El-Halwagi, "Computer-Aided Synthesis of Polymers and Blends with Target Properties," *Ind. Eng. Chem. Res.*, **35**, pp. 627-634 (1996).
15. Crabtree, E. W., and M. M. El-Halwagi, "Synthesis of Environmentally Acceptable Reactions," *AIChE Symp. Ser.*, **90** (303), AIChE, New York, pp. 117-127 (1995).
16. Hildebrandt, D., and L. T. Biegler, "Synthesis of Chemical Reactor Networks," *AIChE Symp. Ser.*, **91** (304), pp. 52-67 (1995).
17. Rotstein, E., D. Resasco, and G. Stephanopoulos, "Studies on the Synthesis of Chemical Reaction Paths — I. Reaction Characteristics in the ($\Delta G, T$) Space and a Primitive Synthesis Procedure," *Chem. Eng. Sci.*, **37** (9), pp. 1,337-1,352 (1982).
18. Hamad, A. A., and M. M. El-Halwagi, "Simultaneous Synthesis of Mass Separating Agents and Interception Networks," *Trans. I.Chem.E.*, **76**, Part A, pp. 376-388 (1998).
19. El-Halwagi, M. M., "Pollution Prevention through Process Integration: Systematic Design Tool," Academic Press, San Diego (1997).
20. Parthasarathy, G., and M. M. El-Halwagi, "Mass Integration for Multicomponent Nonideal Systems," presented at AIChE Annual Meeting (Los Angeles), AIChE, New York (Nov. 1997).
21. El-Halwagi, M. M., and V. Manousiouthakis, "Automatic Synthesis of Mass Exchange Networks with Single-Component Targets," *Chem. Eng. Sci.*, **45** (9), pp. 2,813-2,831 (1990).
22. El-Halwagi, M. M., and V. Manousiouthakis, "Design and Analysis of Mass Exchange Networks with Multicomponent Targets," presented at AIChE Annual Meeting (San Francisco), AIChE, New York (Nov. 1989).
23. El-Halwagi, M. M., and V. Manousiouthakis, "Simultaneous Synthesis of Mass Exchange and Regeneration Networks," *AIChE J.*, **36** (8), pp. 1,209-1,219 (1990).
24. Srinivas, B. K., and M. M. El-Halwagi, "Synthesis of Combined Heat and Reactive Mass-Exchange Networks," *Chem. Eng. Sci.*, **49** (13), pp. 2,059-2,074 (1994).
25. Kiperstok, A., and P. N. Sharratt, "On the Optimization of Mass Exchange Networks for Removal of Pollutants," *Trans. I.Chem.E.*, **73**, Part B, pp. 271-277 (Nov. 1995).
26. Zhu, M., and M. M. El-Halwagi, "Synthesis of Flexible Mass-Exchange Networks," *Chem. Eng. Comm.*, **138**, pp. 193-211 (1996).

- *Operating processes.* As already noted, designing processes and sites as integrated systems creates many benefits. But, once constructed, these facilities also must be operated as integrated systems to fully capitalize on the benefits. Process integration methods can be used effectively to devise operating strategies and to develop process monitoring and control procedures.

Essential changes

Integration requires that companies and individuals make changes. These

will be reflected in the organization, in the skills of the employees, and in the tools they use.

Organization. Integration, if it is to be effective, must become an integral part of how work is performed.

Integration changes the relationship between strategy and technology and, therefore, the relationship between senior management and engineers. It alters the types of problems that are addressed, and how those problems are structured and solved. It changes the data and other informa-

tion required as solution alternatives are generated and screened. And, it greatly reduces the time between problem statement and solution.

Integration, therefore, cannot be just an add-on to the current organization and work processes. It requires commitment for the long term and a systematic rethinking of a company's organization and internal procedures. The result of this rethinking will be a company-specific plan treating integration as a source of competitive advantage.

Skills. Engineers will require train-

27. Papalexandri, K. P., and E. N. Pistikopoulos, "A Multiperiod MINLP Model for the Synthesis of Heat and Mass Exchange Networks," *Comp. Chem. Eng.*, 18 (12), pp. 1,125-1,139 (1994).
28. Huang, Y. L., and L. T. Fan, "Intelligent Process Design and Control for In-Plant Waste Minimization," in "Waste Minimization Through Process Design," A. P. Rossiter, ed., McGraw-Hill, New York, pp. 165-180 (1995).
29. Wang, Y. P., and R. Smith, "Wastewater Minimization," *Chem. Eng. Sci.*, 49 (7), pp. 981-1,006 (1994).
30. Hallale, N., and D. M. Fraser, "Synthesis of Cost Optimum Gas Treating Process Using Pinch Analysis," *Proceedings, Top. Conf. on Sep. Sci. and Techs.*, W. S. Ho, and R. G. Luo, eds., Part II, AIChE, New York, pp. 1,708-1,713 (1997).
31. Hamad, A. A., V. Varma, G. Krishnagopalan, and M. M. El-Halwagi, "Application of Mass Integration to Reduce Methanol and Effluent Discharge in Pulp Mills," *TAPPI J.*, 1998 (in press).
32. Warren, E. A., B. K. Srinivas, and M. M. El-Halwagi, "Design of Cost-Effective Waste-Reduction Systems for Synthetic Fuel Plants," *J. Env. Eng.*, pp. 742-746 (1995).
33. El-Halwagi, A. M., and M. M. El-Halwagi, "Waste Minimization via Computer-Aided Chemical Process Synthesis — A New Design Philosophy," *Trans. Egypt. Soc. Chem. Eng.*, 18 (2), pp. 155-187 (1992).
34. Stanley, C., and M. M. El-Halwagi, "Synthesis of Mass-Exchange Networks Using Linear Programming Techniques," in "Waste Minimization through Process Design," A. P. Rossiter, ed., McGraw-Hill, New York, pp. 209-224 (1995).
35. Spriggs, H. D., "Design for Pollution Prevention," *AIChE Symp. Ser.*, 90 (303), pp. 1-11 (1995).
36. El-Halwagi, M. M., and B. K. Srinivas, "Synthesis of Reactive Mass-Exchange Networks," *Chem. Eng. Sci.*, 47 (8), pp. 2,113-2,119 (1992).
37. Srinivas, B. K., and M. M. El-Halwagi, "Synthesis of Reactive Mass-Exchange Networks with General Nonlinear Equilibrium Functions," *AIChE J.*, 40 (3), pp. 463-472 (Mar. 1994).
38. Dunn, R. F., and M. M. El-Halwagi, "Optimal Design of Multicomponent VOC Condensation Systems," *J. Haz. Mat.*, 38, pp. 187-206 (1994).
39. Dunn, R. F., M. Zhu, B. K. Srinivas, and M. M. El-Halwagi, "Optimal Design of Energy Induced Separation Networks for VOC Recovery," *AIChE Symp. Ser.*, 90 (303), AIChE, New York, pp. 74-85 (1995).
40. El-Halwagi, M. M., B. K. Srinivas, and R. F. Dunn, "Synthesis of Optimal Heat Induced Separation Networks," *Chem. Eng. Sci.*, 50 (1), pp. 81-97 (1995).
41. Dunn, R. F., and B. K. Srinivas, "Synthesis of Heat-Induced Waste Minimization Networks (HIWAMINs)," *Adv. Env. Res.*, 1 (3), pp. 275-301 (1997).
42. Dye, S. R., D. A. Berry, and K. M. Ng, "Synthesis of Crystallization-Based Separation Schemes," *AIChE Symp. Ser.*, 91 (304), pp. 238-241 (1995).
43. Spriggs, H. D., and W. R. Smith, "Design for Pollution Control: Screening Alternative Technologies," *Env. Progress*, 15 (2), pp. 69-72 (Summer 1996).
44. El-Halwagi, M. M., "Synthesis of Optimal Reverse-Osmosis Networks for Waste Reduction," *AIChE J.*, 38 (8), pp. 1,185-1,198 (Aug. 1992).
45. Srinivas, B. K., and M. M. El-Halwagi, "Optimal Design of Pervaporation Systems for Waste Reduction," *Comp. Chem. Eng.*, 17 (10), pp. 957-970 (1993).
46. Malone, M. F., and M. F. Doherty, "Separation System Synthesis for Nonideal Liquid Mixtures," *AIChE Symp. Ser.*, 91 (304), pp. 9-18 (1995).
47. Wahnschafft, O. M., T. P. Jurian, and A. W. Westerberg, "SPLIT: A Separation Process Designer," *Comp. Chem. Eng.*, 15, pp. 565-581 (1991).
48. Richburg, A., and M. M. El-Halwagi, "A Graphical Approach to the Optimal Design of Heat-Induced Separation Networks for VOC Recovery," *AIChE Symp. Ser.*, 91 (304), pp. 256-259 (1995).
49. Noureldin, M. B., and M. M. El-Halwagi, "A Shortcut Approach to Integrating Process Design and Operation via Mass Integration," presented at AIChE Spring Meeting (New Orleans), AIChE, New York (Mar. 1998).
50. Gopalakrishnan, M., A. Dubbs, A. A. Hamad, and M. M. El-Halwagi, "Process Simplification through Process Integration," presented at AIChE Annual Meeting (Los Angeles), AIChE, New York (Nov. 1997).

ing in the integration concepts and in applications know-how. Thus, a decision will have to be made as to the source of this training and how much of the expertise is brought in-house vs. outsourced. Again, the answer will be company-specific.

Even more important to success, however, is something we call "integration mindset." This is an attitude toward problem solving that runs counter to managers' and engineers'

experience, training, and instincts. An integration mindset mandates that the big picture be addressed first using fundamental principles and that details be reserved for later. This "science of the big picture" approach allows the correct structural decisions to be made at each stage of problem solving without slipping into unnecessary detail. This is the power of the integrated approach.

But, it is difficult to implement and to make this mindset stick. There is a strong tendency to slip back into traditional problem solving, which is more intuitive and much more focused on building a solution from the details up rather than from the big picture down. This, though, results in suboptimal solutions that take a longer time to generate, because much of the effort is spent on the (unnecessary) details.

The integration mindset causes engineers to see the forest first and worry about individual trees later.

Tools. The final element of change is a suite of integration tools that complements existing process simulators and design programs. Because design will be performed differently, existing tools will assume different roles than they historically have.

For example, process simulators currently are used for what we term "design by case study." The existing process is first simulated in detail and, then, design changes (different cases) are tested to determine their impact on performance. This is repeated until no other ideas are apparent or until time expires. One of the resulting solutions then is implemented. This approach generates workable but suboptimal designs.

Design using integration principles makes far less use of process simulators in the early stages. The major structural decisions can be made with simpler models, together with visualization techniques that put engineers in a stronger decision-making role. This generates better designs in a shorter amount of time.

The integration tools can be constructed internally or acquired externally. Each company must decide the

source of the tools and how much external support is needed in using them.

An optimal approach

As this article has emphasized, there is now a "science of the big picture." It adopts a different problem-solving philosophy — addressing the big picture first using fundamental principles, then dealing with details only after the major structural decisions have been made — and can deliver immediate, substantial, and far ranging benefits.

This approach is bolstered by powerful tools that treat industrial processes and sites as integrated systems. Proven and ready to use, these tools are highly effective for the full range of a company's processing objectives, including site planning, process development, process design, resource conservation, regulatory compliance, and operations planning and control.

This combination provides companies with a unique opportunity to align technology with corporate strategy, and to create, defend, or extend their competitive advantage.

Nomenclature

- b_j = intercept of equilibrium line
 - c_j = operating cost related to the j th MSA
 - c_j^f = operating cost of the j th MSA per kg of removed species
 - i = index for rich streams
 - j = index for lean streams
 - m_j = slope of equilibrium line
 - MR = quantity of mass transferred from rich stream
 - N_R = number of rich streams
 - N_S = number of lean streams
 - P^{total} = total pressure of gas
 - P^o = vapor pressure of an individual species
 - R = rich stream
 - r = reaction rate
 - S = mass separating agent
 - T = temperature
 - T^d = dew point
 - T^* = target temperature
 - Q = amount of heat released
 - Q^c = minimum cooling duty
 - u = index for processing units
 - V_v = flow rate of v th gas stream
 - W_w = flow rate of w th liquid stream
 - x_j = composition of a species in an MSA
 - x_j^* = maximum theoretically attainable composition of a species in an MSA
 - y_i^* = rich-phase equilibrium composition of targeted species
 - y_v = composition of species in v th gas stream
 - z_w = composition of species in w th liquid stream
- superscripts:**
- R = rich phase
 - r = removed species
 - S = lean phase
 - s = starting (or supply) value
 - t = target value
- Greek letters:**
- ϵ_j = minimum allowable composition difference
 - ν = index for gaseous source
 - ϕ_ν = load of species in ν th gas stream
 - ψ_w = load of species in w th liquid stream
 - ω = index for liquid source

M. M. EL-HALWAGI is a professor in the Chemical Engineering Dept. of Auburn University, Auburn, AL ((334) 844-2064; Fax: (334) 844-2063; E-mail: mahmoud@eng.auburn.edu), and cofounder of Matrix Process Integration, L.C. He has pioneered many developments in mass integration, and has served as a consultant to companies throughout the CPI. Recipient of several awards including the National Science Foundation's National Young Investigator Award, the Birdsong Merit Teaching Award, the Fred H. Pumphrey Award, and an Alumni Professorship from Auburn Univ., he has a BS and MS from Cairo University, and a PhD from the University of California, Los Angeles, all in chemical engineering.

H. D. SPRIGGS is cofounder and President of Matrix Process Integration, L.C., Leesburg, VA (703) 779-0199; Fax: (703) 771-2146; E-mail: hdennis@mindspring.com), a company that specializes in developing integrated process designs and operating strategies, and aligning technology with corporate strategy. A pioneer in the development and application of methods for systematic process design, he has over 30 years of industrial and academic experience, including at Allied Chemical and Union Carbide, and has operated his own business since 1985. He received a BS from West Virginia Inst. of Technology and a PhD from the Univ. of Virginia, both in chemical engineering. He is a Fellow of AIChE.