**Methods for Synthesis of Multicomponent Distillation and its Optimal Sequencing: A Review**

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**Abstract**

In the early days of chemical engineering, processes were split into different operational units, and the design phase of the process was prioritized above the analytical phase. In contrast, the stage of synthesis has received less attention and thought. On the other hand, modern chemical processes have over time become more extensive and sophisticated, as well as capital and energy intensive. Typically, chemical processes include the separation of multicomponent mixture into multiple products via a sequence of columns, each of which generates two or more products or intermediate streams.

This paper investigates and evaluates the significant advancements in methodologies for selecting the appropriate sequence of distillation column trains for separating multicomponent mixtures into their components. The study investigates different sharp and non-sharp/sloppy separation mechanisms as well as complex distillation column designs. It also identifies prospective research obstacles for the exhaustive synthesis of distillation columns, as well as the possibility of utilizing innovative computational techniques to determine the optimal arrangement.

**Keywords:** [Separation](https://www.annualreviews.org/keyword/Process+Synthesis), [Optimization](https://www.annualreviews.org/keyword/Process+Flowsheets), [Sequencing](https://www.annualreviews.org/keyword/Distillation+Sequences), Energy Integration, [Synthesis](https://www.annualreviews.org/keyword/Superstructures)

**1. Introduction**

In the chemical, biochemical, and petrochemical sectors, separations are ubiquitous, and the majority of separations account for 40-70 percent of the entire plant energy demand (Humphrey and Keller, 1997). The most used separation method in the chemical industry is distillation, which is the process of separating distinct hydrocarbons based on the variations in their relative volatilities or boiling points (Charles, 1997). It is responsible for 90–95% of all separations and utilizes 40–60% of the energy in the chemical and refining industries (Humphrey, 1992; Sholl and Lively, 2016). Many of the world's biggest and most lucrative separations include distillation, including crude oil fractionation, hydrocarbon separation from steam cracking, and natural gas liquids (NGL) separation. Even for gas separations, distillation is the sole option for products with greater throughput and greater purity.

In the process of distillation, separation occurs in a huge column including many trays where hydrocarbon gases and liquids interact. Furthermore, liquid flows down the column while vapour rises (Charles, 1997). Crude oil is primarily a complicated combination of hundreds of hydrocarbons that are fractionated in a petroleum refinery to produce usable crude oil products, such as naphtha, kerosene, diesel, and other gas oils. Light distillate products are the most lucrative; hence, the atmospheric column is designed to maximize the production of these lighter components. One estimate indicates that more than 40,000 distillation columns are in operation in the United States. These column burns the equivalent of 1,200,000 barrels of oil every day (Humphrey and Keller, 1997). It is vital to maximize the efficiency of processes in order to lower the energy consumption at a particular scale of operation within a given budget.

The article examines major trends in the sequencing of conventional and energy-integrated distillation systems, as well as several methods for determining the optimal sequence. The paper examines techniques for sharp and non-sharp/sloppy separation systems as well as various complex distillation column designs.

**2. Different types of distillation**

Distillation has separated several substances based on the relative ease with which they evaporate. The basic component of distillation is the distillation column, which is used to separate liquid mixtures into their components or fractions based on variations in volatilities. There are several varieties of distillation columns, each intended to accomplish certain sorts of separations, and moreover, each design varies in complexity. Small-scale laboratory distillations and large-scale industrial distillations use fractionating columns (Kister,1992).

***2.1. Batch and continuous distillation***

Batch and continuous distillation are the two basic methods of classical distillation, depending on the column feed. In the process of batch distillation, the feed to the column is introduced in batches. In addition, before the distillation process begins, a batch is added to the column. The subsequent batch of feed is then introduced as the required objective is accomplished. Batch distillation accepts one batch (or lot) of feed at a time and separates it into products by eliminating the more volatile fractions over time. Continuous distillation, on the other hand, as its name implies, continually separates a feed into two or more products (Kiss, 2013). They are also capable of handling large through-puts so long as there are no interruptions and no issues with the column or neighbouring process units. In our investigation, we will solely focus on this category of columns.

***2.2. Binary and multicomponent distillation***

Continuous columns may be further categorized into binary and multicomponent distillation groups. In binary distillation, just two products are separated, and the separation of ethyl alcohol (ethanol) and water is a prime example. As a result of its simplicity, binary distillation is used for the majority of important distillation in education and many theoretical publications. In contrast, multicomponent distillation separates the chemical mixture; petroleum refining is a prime example of this process. In petroleum refining, crude oil is a tremendously complex combination of hydrocarbons containing literally hundreds of distinct molecules, which may be separated using multicomponent distillation. Compared to binary distillation, the theory and practise of multicomponent distillation may be rather complicated, and multicomponent distillation accounts for practically all industrial distillation (Taylor and Krishna, 1993; Charles, 1997). In multicomponent distillation, more than one distillation tower is necessary; for n components, n minus one fractionators are needed. Specification Restrictions Distillation of several components is prevalent in the chemical and industrial industries (Humphrey, 1992). It manages some of the biggest and most lucrative separations in the world, including fractionation of crude oil, hydrocarbon separations from steam cracking, and natural gas liquids (NGL) separations.

A distillation configuration is a succession of distillation columns used to accomplish multicomponent distillation. To distil a non-azeotropic multicomponent mixture with n components into n product streams, each enriched in one of the elements, a distillation arrangement consisting of a series of distillation columns is necessary. To discover a desirable distillation configuration for a particular separation objective, it is necessary to first create a search space that encompasses and contains only all excellent designs.

***2.3. Azeotropic and extractive distillations Distillation***

According to the ways of separating mixtures, distillation may be divided into azeotropic and extractive distillations. Azeotropic distillation is the technique where a third component is employed to separate two close-boiling components (Agrawal, 1996). Typically, the third component added to the combination is water or benzene, since these might aid in enhancing the volatility of a chemical; in this context, volatility refers to the substance's ability to evaporate (Agrawal, 1999). In this technique of distillation, an azeotropic component is produced to enhance the difference in boiling points and assist separation by distillation. In addition, this azeotropic component may be of great assistance in the separation process since it does not modify the components as ordinary distillation would. In this distillation, an entrainer is employed to capture particles in the mixture in order to separate the azeotrope, and a heterogeneous mixture is produced.

In contrast, extractive distillation is a method of distillation in which mixing or miscibility and the measuring of separating a mixture may occur without the formation of an azeotrope component. In this technique, the component may be non-volatile or have a high boiling point, and this approach is often used for mixes with almost identical volatility. In this procedure, the separation solvent interacts with the mixture to aid in the separation of the components. By using the separation solvent, it is possible to quickly separate the mixture without generating an azeotrope, as is usual with conventional separation procedures (Agrawal, 2001; Agrawal, 2003).

**3. Multicomponent distillation Synthesis**

The frequent distillation challenge in multicomponent systems is the separation of a multicomponent feed into distinct streams that are richer in one of the feed stream's components (Agrawal, 2003). Previous scholars calculated the feasible sequence for the separation of n-component mixtures into single-component product streams using an equation (Thompson and King, 1972). When the number of components in the feed stream is larger than three, the number of alternative distillation column configurations to yield n product streams enriched in one of the components is rather enormous (Agrawal, 1996). A significant number of reboilers and condensers were often linked with these configurations, which explains why. Due to the capital cost of reboilers, condensers, and other critical components, it may be cost-effective to construct energy-efficient systems with fewer reboilers and condensers (Agrawal, 2001; Agrawal, 2003). Distillation utilizes the variations in boiling points of components in a mixture to separate them. At the time of reboiler heating, which is the high-temperature heat sink, energy is required. On the other hand, the condenser was the heat source with a lower temperature that required cooling; in these cases, external utilities are necessary. For this reason, energy and cost-saving measures based on thermal integration of the distillation columns, heat sources, and external heat sources have been prioritized in these areas (Agrawal, 2001; Agrawal, 2003).

Increasing demands for energy efficiency and adherence to global environmental rules as priority in industrial operations have shifted the emphasis to production rates and product quality in the contemporary world. The synthesis and analysis phases may be used to classify chemical processes (Caballero and Grossmann, 2004). The synthesis step includes the selection of the process route or configuration, as well as the selection of the fundamental process unit types. In contrast, the analysis phase focuses on establishing the design and optimization of each unit within the framework of a chosen configuration or process scheme. In the early days of chemical engineering, processes were divided down into distinct operational units, with process design taking precedence over the analytical step.

The synthesis process, however, has received less attention and care. The process of identifying the ideal solution relied heavily on the expertise and intuition of the designers. This was exacerbated by the fact that there were no formal processes in place for such choices, and it was seen as more of an art. In spite of this, contemporary chemical processes have been exposed to growing size and complexity, capital and energy demands throughout time. These factors have prompted a greater examination of formal design tools for use in the synthesis phase of the processes. Early distillation was a straightforward physical process. During this era, glassware with long, downward-pointing necks was referred to as retorts. These worked as condensers, cooling the distillate and allowing it to flow downward. These were eventually replaced with alembics made of copper, which, with modifications, became known as pot stills. The operations included one vaporization and one condensation, with further distillation increasing the purity. There have been claims of as many as 500 to 600 distillations in order to achieve a pure chemical. Anthony Perrie possessed Continuous stills were first developed in 1822. (Othmer, 1982).

Distillation entails selective boiling and condensation, resulting in either total separation or partial separation in which variations in the relative volatility of the mixture's components are used to one's advantage. The relative volatilities of components *i* and *j* are defined as follows:

 (1)

where, *yi* = mole fraction of component '*i*' in the vapor

*xi* = mole fraction of component ‘*i*’ in the liquid

**4. Approaches and Methods of Distillation Sequencing**

At the commencement of multicomponent distillation, many parameters are specified, including temperature, pressure, feed composition and rate, distillation pressure, and feed to be put on that tray. Then, we must take into account the number of trays (optimum feed tray position), heat loss (even if assumed to be zero), the number of trays, the reflux ratio (the ratio of vapour generated in the reboiler to residue withdrawn), and the concentration of the component in one product (a maximum of two may be chosen). The multicomponent design must then take into account the ratio of flow of the distillate from one element to the flow of the same element to the residue, or split of the element (a maximum of two may be selected), and the ratio of total distillate to total residue.

***4.1. Heuristics methods***

As previously noted, the number of alternative distillation column configurations may grow rather enormous, and to make the issue more manageable, fewer variants are used. In 1947, Lockhart examined several distillation plant designs based on their design and associated expenses. As the configuration of choice, he found that the configuration in which the components are eliminated has decreasing volatility. The first advantage was the revelation that feedstock may be used as an alternative arrangement. In the early phases of heuristics, a lot of studies have been conducted on the selection of optimum design of multicomponent systems using heuristics like as Heaven (1969) offered a set of heuristics recommendations for a variety of process situations based on systematic design and economic research. King has reported many study results, which are detailed below (King, 1971)

1. When the relative volatility of key components was near to one in the absence of non-important components, separation had to be performed.
2. In these separations, column overheads were selected for eliminating components one by one.
3. Sequences occurred in a more ambiguous separation of the feed into distillate and bottom products.
4. Sequences of very precise recoveries have to be maintained until the final sequence.

Previous studies have proposed heuristics and evolutionary techniques based tactics (Seader and Westerberg, 1977). Prior to then, at least fourteen algorithms for synthesizing multi-column separation sequences have been developed by other researchers (Lockhart, 1976, Seader and Westerberg, 1977, Stephanopoulos and Westerberg, 1976). Previous researchers described the viewpoint of evolutionary synthesis as adopting these four fundamental concepts (Seader and Westerberg, 1977). The first two concepts were an initial flow sheet and rules for making minor, systematic adjustments to the flow sheet, resulting in what is known as a neighbouring flow sheet (Seader and Westerberg, 1977). The other two concepts were an efficient method for applying these rules and a method for comparing the original flowchart to its neighbour (Seader and Westerberg, 1977). Previous scholars have investigated the use of the set of six heuristics numbered 1 through 6 to the synthesis of multicomponent distillation, as briefly explained below (Seader and Westerberg, 1977).

They designated Heuristic 1 as forbidding splits and conducted a preliminary screening of separation techniques based on an analysis of a number of factors in an effort to lessen the complexity of the combinatorial issue. The first heuristic started with an examination of the technical feasibility of conventional distillation, which was applicable to the full area of coexisting vapour and liquid phases. It supplied species that were thermally stable at the employed circumstances, and this area stretched from the temperature of crystallization to the convergence pressure. If refrigeration was necessary for the overhead condenser, alternatives to conventional distillation, such as absorption and reboiled absorption, may be explored. On the other end of the spectrum, liquid-liquid extraction with different solvents may be considered if the vacuum operation of standard distillation is required.

In the Heuristic 2 technique, an ordered list of components based on volatility and another separation index was constructed. Then, they assessed the relative volatility for each adjacent pair of elements, such as I and j, and evaluated whether the nearby relative volatility Ki/Kj was more than 1. According to King, this heuristic 2 was connected to the heuristic where separations and the relative volatility of the crucial components were near to unity and should be carried out in the absence of non-critical parts (1971). In the third heuristic, the sequence of splits to eliminate components was determined in order of decreasing molar percentage in the process feed when the molar percentage fluctuated significantly but the relative volatility did not (Lockhart, 1947).

In the Heuristic 4 rule for removing components sequentially as overhead products, neither relative volatility nor molar proportion in the feed fluctuated much, and the resultant structure, which was the direct sequence, was regularly utilized in practise. In this construction, the pressure tended to be maximum in the first separator and decreased with each succeeding division when conventional distillation was used as the separating technique (Hudd, Powers, and Siirola, 1973). In the instance of Heuristic 5, when a mass separating agent was utilized and withdrawn from the separator immediately after the one into which it was entered, it was determined that the agent was effective (Hendry and Hughes, 1972). On the other hand, in Heuristic 6, when multi-component products were defined, sequences that directly created those products with little blending were favoured. The nearby relative volatilities were much lower than they would have been in a configuration requiring extra separators and included (Thompson and King, 1972).

***4.2 Evolutionary Strategies***

Following Heuristic approaches, evolutionary tactics were used for ways of sequence distillation (Gadkari and Govind, 1988). Evolutionary approaches were those that synthesized new sequences by modifying previously created sequences, and it included improvements to the discovery process. These techniques led to the maximization of benefits, which was followed by an evaluation of the new process. Some researchers devised evolved algorithms for Heuristic search that seemed successful and were backed by a lower-level heuristic that was violated to get the present structure (Nath and Motard, 1981). They devised a rating mechanism to quantify the choices of the next separation task, with which they qualitatively agreed. An "evolutionary" modification was then made if an alternate option at each selection stage had a value within a few percent of the optimal solution and the structures arising from this alternative decision were also searched. They created far fewer shapes and could ensure optimum structures. In their study, Umeda et al. (1979) presented an evolutionary approach technique for heat integrated system design based on the available energy idea for the distillation process.

For the objective of enhancing the energy performance of a process, previous researchers developed a remarkable and relatively flexible evolutionary strategy. Afterwards, they refined charting and managing the integrated heating and cooling curves utilized by prior researchers to determine the lowest utility needs for a process (Hohmann, 1981). They also advised recognizing where these curves constricted additional heat integration, and then adapting the process to "open" the pinch by modifying process operating parameters, such as raising column pressure (Umeda, 1979). In addition, they demonstrated the concepts by designing a better heat-integration technique for a five-component distillation issue as part of their study. However, it might improve heat recovery around distillation systems using heat pumps and multi-effect boilers and coolers in addition to intermediate boilers (Umeda, 1979).

Previous scholars have also discussed the evolutionary methodologies for multi-component distillation to synthesize the optimal separation configuration by often modifying configurations that have been previously developed (Boozarjomehry et al. 2009). In addition, they underlined the necessity to consider three essential activities for this goal. (1) Development of an initial separation sequence; (2) identification of the evolutionary rules that can generate all valid structural changes in a separation sequence; and (3) determination of the evolutionary rules that will specify how the system evolves to a series of increasingly superior configurations. (Muraki and Hayakawa) (1987). In their study, the first step was to identify the ideal separation sequence, and the second was to improve the distillation separation process by integrating energy into the existing separation sequence. In addition, these two phases were repeated until the optimum energy integrated distillation separation technique was formulated. Some scholars, on the other hand, established a systematic approach by viewing column pressure as a continuous variable in a mixed-integer nonlinear formulation (Floudos and Paules, 1988). It was built on a superstructure that included all heat integration choices as well as the various distillation sequences, while the majority of earlier synthesis methods relied on energy savings, which was dependent on utility costs.

On the other hand, researchers have created a thermodynamic branch-and-bound approach for the distillation system that results in the least amount of heat availability loss. However, neither a search for the ideal separation sequence nor an economic assessment was included in their methodology (Naka et al., 1982). Other researchers considered the heat integration of distillation columns into the whole process and devised the concept of the pinch point as if their study focused only on heat integration for a constant material balance (Linnhoff et al., 1983).

***4.3 Conceptual Approaches***

The construction of chemical process flow-sheets required a substantial amount of conceptual knowledge as a consequence of the engineers' lifetime expertise with the process (Shah and Kokossis, 2001). In order to evaluate tradeoffs and make design choices, ideas were merged with thermodynamic insights, physical insights, and an expanding number of modelling tools. Prior studies recognised the significance of engineering expertise for synthesis, design, and optimization methodologies focused mostly on mathematical programming (Raman & Grossmann, 1991; Daichendt & Grossmann, 1994). For the process of multi-component distillation, the separation of multi-component mixtures using distillation columns may result in a considerable reduction in energy consumption compared to simple columns (Doukas and Luyben, 1978; Glinos and Malone, 1988; Petlyuk et al., 1965; Tedder and Rudd, 1978). For this reason, the selection of several complicated configurations is highly dependent on the separation challenge, including the feed's composition and temperature state. In addition, it depends on the characteristics of the products and the relative volatilities of the components to create combinations that behave optimally. As a result, these techniques take the shape of general-purpose programme with low conceptual meaning, which complicate the synthesis process.

For the distillation process, it was known that heuristics would often overlap and that a systematic method would be difficult to cover. In addition, the difficulty and biases of the mix impact the interpretation of superstructures towards traditional designs. Although ideas and models were kept distinct, the application of technical knowledge sometimes imposed more limits than it eliminated. The disintegration structures were often advantageous because they did not account for the significant relationships between the different subsystems. Combining sophisticated separation systems necessitated a number of obstacles and problems, such as a huge number of design possibilities, complicated design models, and various economic ramifications.

For Conceptual Approaches, the researchers used conceptual models with a rigorous optimization agenda to monitor and evaluate all design alternatives using essential process data prior to comprehensive simulation (Shah and Kokossis, 2001). They described the conceptually dense presenting models that facilitate the concurrent application of engineering insights and mathematical programming. In addition, the separation synthesis issue was addressed using a distinct technique that presupposes discrete representation, and the discrete model was used to cover prior research (Hendry and Hughes, 1972). In addition, they accommodated complicated column configurations by developing hybrid jobs and transformations, and they created performance models that capitalize on the impacts of distillation (Shah and Kokossis, 1997). In their study, they extended the presentation of 'conceptual losses' for complicated distillation systems and were able to create theoretical models in advance of simulation and optimization. As feed conditions and requirements fluctuate throughout the distillation system, these abstract approaches are capable of addressing mixed-integer linear programming problems and are easy to modify.

***4.4. Algorithmic Methods***

For the design and pricing of distillation columns, algorithmic approaches use dynamic programming tools, thorough analytical procedures, and optimization strategies. There are two subcategories of algorithmic approaches, including parametric studies and mathematical programming methods.

***4.4.1. Parametric studies***

In parametric studies, researchers attempted to find essential factors and analyzed their impact on the optimal separation configuration for multicomponent distillation. Numerous variables impacted the cost of separation configuration, and these parametric analyses had limited utility. Their primary outcomes are a collection of heuristic rules for rapid and straightforward estimation..

Genetic Algorithm (GA) is a stochastic global optimization method that mimics the natural selection and evolution processes (Holland, 1975; Goldberg, 1989). This approach produces new search space points by applying operators to existing points and statistically progressing toward more optimum portions of the search space (Haupt and Haupt, 1998). The GA has the intrinsic capacity to encode very complex structures using a basic representation and applies simple transformations to enhance such structures during the optimization of highly complex objective functions. GA has been frequently used to combinatorial optimization issues because to this property and its simplicity of implementation.

***4.4.2. Mathematical programming methods***

Mathematical programming approaches, on the other hand, utilize a variety of mathematically-based optimization strategies to generate the optimal separation arrangement. The primary benefit of these procedures is their rigour and the fact that only they can ensure an optimal outcome. However, the primary disadvantage of these strategies is that they can only be used to issues that are simplified.

**5. Conclusions**

Much work and time has been invested on research into the creation of systematic techniques to planning distillation sequences, as these may account for up to 60-70 percent of a chemical plant's capital output. As an ever-evolving process, synthesizing multicomponent separation has a significant place. When it comes to running costs, utility consumption in distillation sequences accounts for a sizable share. This has paved the way for additional research into heat integration across different columns in a distillation cycle. The effect is unanimously acknowledged to be far more economically advantageous. As a result, the chemical industry is eager to work on improving energy efficiency in order to save energy and so make it more cost effective.

A major critique of optimization approaches has been their inefficiency in the face of synthesis issues and an inability to explain how they physically work. Heuristics and thermodynamic objectives have helped to solve various synthesis challenges, particularly in the heat recovery industry. They have, however, been unable to fulfill demands from varied concerns in synthesis from the standpoint of a uniform framework. Furthermore, attaining optimization is not a certainty. One significant limitation is their inability to explain the causes for relationships in the synthesis process in various portions of a complete processing system (Papoulias, 1983).

Sharp separators have been the subject of several studies on the improving energy efficiency of separation sequences. It has shown that additional work on heat recovery for separation sequences allowing component distribution across the top and bottom products is necessary. It has been showed that, despite their potential, present techniques are incapable of correctly describing every physical fact and engineering practice. On the other hand, it is not feasible to do all the extensive time-consuming computations for all process possibilities at the preliminary design stage. As a result, heuristics and the engineers' experience were employed in the early stages before moving on to more complex procedures. The number of design possibilities for distillations increases with the number of components in the mixture to be separated increases. According to reports, there are approximately 500,000 permutations possible for a six component mixture separation. To complicate matters further, each of these arrangements differs from the others and consumes different amounts of energy under different conditions. As newer techniques, artificial neuron networks and fuzzy logic techniques are being used in diverse formulations. In this situation, it is timely to systematize all of these approaches and conduct investigations in a more comprehensive manner.

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