

## Process Intensification in Multicomponent Distillation

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Process Intensification (PI) is an emerging concept in chemical engineering which describes the design innovations that lead to significant shrinkage in size and dramatic boost in efficiency in a process plant. Distillation, which is one of the most important separation technologies in the chemical industry, is therefore a crucial component in PI. Here, we discuss two aspects of PI in multicomponent distillation: 1) Performing simultaneous heat and mass integration among thermally coupled distillation columns to reduce the number of columns and heat duty requirement; and 2) Conducting any thermally coupled distillation in only a single column shell using a dividing wall column that is fully operable. Through examples, we show that synergistic use of both strategies leads to the design of compact, easy-to-operate, energy efficient and cost effective multicomponent distillation systems.

### 1. Introduction

Distillation is an important separation process that accounts for 90-95% of all liquid separations and consumes >40% of energy in the chemical and refining industries (Humphrey and Siebert, 1992). To separate a multicomponent mixture containing  $n$  components into  $n$  pure products, a sequence of distillation columns known as a distillation configuration is required. The most common class of configurations, known as the regular-column configurations, use exactly  $(n-1)$  columns to separate an  $n$ -component mixture (Shenvi et al., 2012). Regular-column configurations with each column having one reboiler and one condenser are referred to as basic distillation configurations. For a basic configuration, one can derive a sequence of thermally coupled configurations by replacing one or more intermediate heat exchangers with two-way liquid-vapor transfer streams known as thermal couplings. The number of basic and thermally coupled distillation configurations available increases combinatorially as the number of components in the feed increases (Shah and Agrawal, 2010). These configurations, while all carrying out the same separation task, can have very different capital and operating costs. Moreover, due to their structural differences, some configurations are easier to operate and control than others. These concerns naturally raise the following question: which distillation configuration design(s) is/are more attractive to build and operate? For the past decades, a number of ideas and solutions have been proposed for designing energy efficient and cost effective multicomponent distillation configurations. These approaches all fall into the category of "Process Intensification (PI)". The concept of PI started to influence the chemical industry from the 1970s, and has attracted more and more attention ever since, especially since the past decade (Reay et al., 2013). Over the past decades, people have proposed different definitions of PI under various scenarios (Stankiewicz and Moulijn, 2000). However, in the context of multicomponent distillation, we believe that PI stands for innovative process synthesis strategies that minimize the number of equipment pieces, reduce total cost, while boosting energy efficiency of multicomponent distillation systems. The overall goal for PI is to synthesize compact, easy-to-operate, energy efficient and cost effective configurations for a given separation task. Here, we discuss two important strategies for achieving this goal. The first strategy involves incorporating simultaneous heat and mass integration to thermally coupled distillation configurations to achieve maximum energy savings. The second strategy deals with performing any thermally coupled distillation in one column shell taking the form of an easy-to-operate dividing wall column.

## 2. Attractive new configurations synthesized by novel heat and mass integration strategy

Heat and Mass Integration (HMI) to consolidate distillation columns is one of the many ways to reduce the total cost of a configuration. One important aspect of HMI deals with introducing additional column sections. Such a HMI strategy is also referred to as HMI with Additional Sections, or simply HMA (Madenoor Ramapriya et al., 2015). One early example of HMA is shown in Figure 1b (Brugma, 1942). In this paper, pure components are denoted by alphabets A, B, C, and so on, with volatilities decreasing in the same order. Streams with two or more components are called submixtures. Filled and unfilled circles in all figures denote condensers and reboilers, respectively. In the 4-component configuration of Figure 1a, the pure product B produced at the reboiler of column 2 is more volatile than the pure product C produced at the condenser of column 3. Thus, instead of having two individual columns, HMA consolidates column 2 and column 3 into a single column with the introduction of an additional section. It subsequently removes the reboiler and the condenser associated with product B and C, respectively. Products B and C are now withdrawn from column 2-3 as side-draws. Hence, the resulting heat and mass integrated configuration shown in Figure 1b, also known as the Brugma configuration, uses fewer heat exchangers and distillation columns. Also, since the vapor generated at the reboiler of column 2-3 is now utilized for both splits,  $AB \rightarrow A/B$  and  $CD \rightarrow C/D$ , the overall energy consumption reduces. The resulting capital and operating cost reduction makes HMA attractive for many industrial applications. Also, notice that a dividing wall column (DWC) shown in Figure 1c can be derived from the Brugma configuration when thermal couplings are introduced at submixtures AB and CD before merging the two columns into a single shell (Kaibel, 1987).

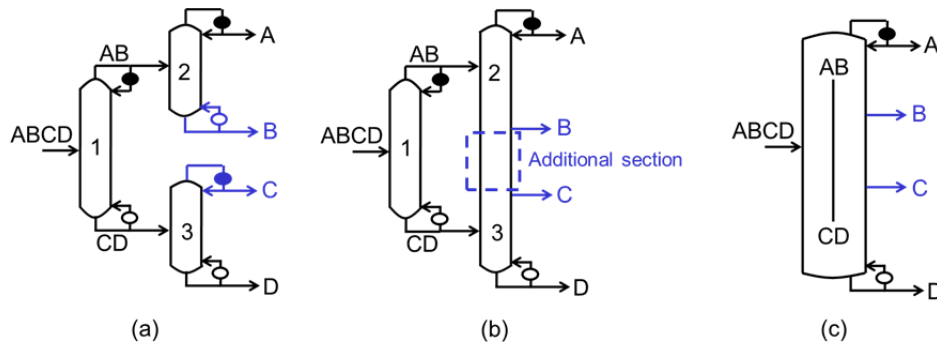


Figure 1: (a) A 4-component basic configuration; (b) Brugma configuration (Brugma, 1942); (c) DWC of Brugma configuration (Kaibel, 1987).

Of course, HMA is not limited to eliminating reboilers and condensers that produce pure product streams. In the 6-component configuration shown in Figure 2a, the submixture BC produced at the reboiler of column 2 is more volatile than the submixture DE produced at the condenser of column 3. Likewise, column 4 produces the bottom product C which is lighter than D, the product produced at the top of column 5. Therefore, a series of new heat and mass integrated configurations can be derived. For example, one may consider consolidating columns 2 and 3 as well as columns 4 and 5 to form two new columns shown in Figure 2b. Two intermediate column sections are added, and submixtures BC and DE as well as pure products C and D are all withdrawn as side-draws from column 2-3 and column 4-5, respectively. Similarly, one may also consider consolidating only columns 2 & 3, or columns 4 & 5, or columns 2 & 5, and so on.

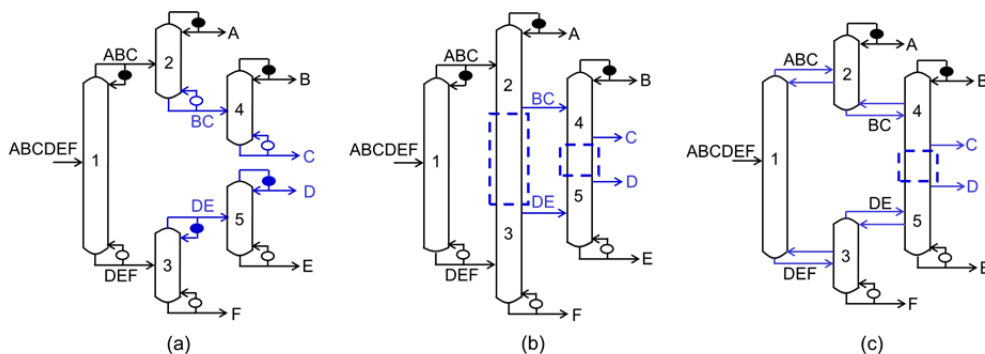


Figure 2: (a) A 6-component basic configuration; (b) Heat and mass integrated configuration using 2 HMAs; (c) The derived HMP configuration from (a).

Among the array of heat and mass integrated configurations that can be synthesized from Figure 2a, it turns out that the configuration shown in Figure 2c will always have the lowest heat duty requirement. This configuration is obtained by first replacing all submixture heat exchangers with thermal couplings, followed by introducing HMA between the condenser at D and reboiler at C. To understand why this configuration is energy efficient, notice that the vapor generated at the reboiler of column 3, which is at the highest temperature level, is directed to progressively produce streams DE, D, C, BC, and B which are at continuously decreasing temperature levels. Therefore, by sequentially degrading it to produce submixtures and pure products, this vapor has been utilized to its maximum potential. However, when HMA is directly introduced at submixtures as in Figure 2b, the vapor generated at the reboiler of column 2-3 (reboiler F) is directly degraded to produce streams DE and BC without producing C and D. This implies less separation work accomplished by the vapor and greater irreversibility due to greater temperature difference between BC and DE. Thus, as a simple heuristic, HMA at submixture level should be avoided when vapor can flow freely across different distillation columns through thermal couplings, as in Figure 2c at submixtures BC and DE.

Configurations as the one in Figure 2c can be easily synthesized from the set of basic configurations, by first replacing all submixture heat exchangers with thermal couplings and then performing HMA only between pure components if possible. We refer to this new set of heat and mass integrated configurations as HMP configurations. Recently, Shah and Agrawal (2010) developed a simple-to-use algorithm, referred to as the SA method from hereon, to generate the complete search space of all basic distillation configurations. This powerful synthesis tool enables us to, for the first time, enumerate all HMP configurations. For each configuration generated by the SA method, we can determine if there simultaneously exists any pure product produced at a reboiler that is lighter than any pure product produced at a condenser. If so, this configuration is a candidate configuration for HMP. Table 1 summarizes the enumeration results for up to 6-component separations (Jiang et al., 2018). Note that the last column suggests that there exist multiple possible ways of introducing HMP for some candidate configurations.

Next, through a case study involving 5-component separations, we demonstrate that in many cases, the HMP configurations can often have the same minimum total vapor duty as the Fully Thermally Coupled (FTC) configuration, which is known to always consume the lowest minimum vapor

Table 1: Enumeration of HMP configurations

$n$	Number of basic configurations	Number of candidate configurations for HMP	Number of HMP configurations
4	18	1	1
5	203	15	17
6	4373	282	347

duty requirement among all configurations in the search space (Fidkowski and Królikowski, 1987). This case study involves easy separations for A/B and B/C, and difficult separations for C/D and D/E. The relative volatility between each consecutive component pair is  $\{\alpha_{AB}, \alpha_{BC}, \alpha_{CD}, \alpha_{DE}\} = \{2.5, 2.5, 1.1, 1.1\}$ . For 5-component mixtures, 31 different representative feed compositions can be obtained, depending on whether each component is rich or lean in the feed (Giridhar and Agrawal, 2010). The main feed and pure products are taken to be ideal saturated liquids. The search for minimum vapor duty requirement of any configuration at minimum reflux is formulated as a Nonlinear Programming (NLP) problem where Underwood's method is embedded for vapor duty calculations (Nallasivam et al., 2016). Each NLP problem is solved to global optimality using solvers such as GAMS/BARON (Tawarmalani and Sahinidis, 2005).

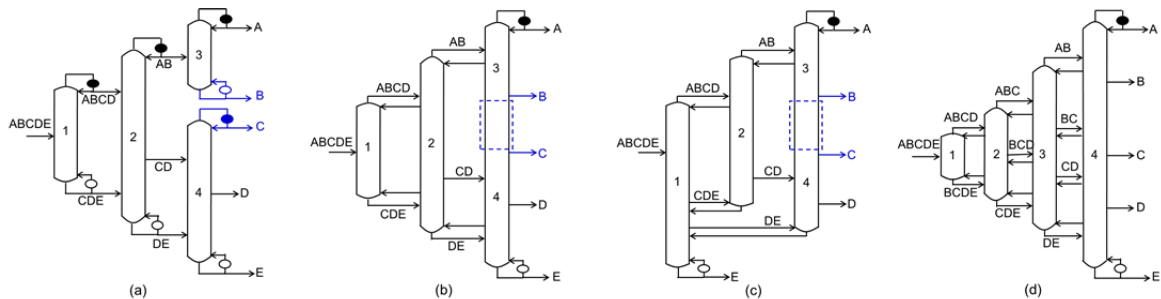


Figure 3: (a) Candidate basic configuration for HMP; (b) The resulting HMP configuration; (c) An operable version of (b) by moving the bottom sections of columns 2 and 3-4 under column 1; (d) The FTC configuration.

The HMP configuration of interest, which is shown in Figure 3b, is derived from the basic configuration of Figure 3a (Jiang et al., 2018). Under 18 of all 31 feed cases, this HMP configuration requires the same minimum total vapor duty as the FTC configuration of Figure 3d, but uses only 3 distillation columns instead of

4. Also, under 7 other feed cases, the minimum vapor duty for the HMP configuration is only less than 10% higher than the FTC configuration. Further, this HMP configuration uses only 13 column sections and 5 intercolumn transfers of submixtures, as opposed to 20 sections and 9 submixture transfers for the FTC configuration. With much fewer column sections and submixture transfers, the HMP configuration of Figure 3b is expected to be easier to control than the FTC configuration. Furthermore, exhaustive calculations for all basic and thermally coupled configurations in the entire search space generated by the SA method indicate that no configuration with less than 14 column sections can give the same minimum total vapor duty requirement as the FTC configuration. Therefore, under these feed conditions, the HMP configuration of Figure 3b is very attractive to build.

Last but not least, due to presence of thermal couplings, HMP configurations can also be made fully operable by rearranging the column sections that are connected by thermal couplings (Agrawal and Fidkowski, 1998). An operable version of the HMP configuration of Figure 3b is drawn in Figure 3c. The derived operable versions are thermodynamically equivalent to the original HMP configurations, i.e. they have the same minimum total vapor duty requirement as the HMP configurations. This potential operational benefit associated with HMP configurations offers industrial practitioners with great opportunities for PI.

### **3. New systematic methodology to synthesize all dividing wall columns for any thermally coupled configuration**

Although the concept of DWC was first proposed as early as 1940s (Wright, 1949), the first industrial implementation of DWC did not happen until the late 1980s (Kaibel, 1987). Since then, the use of DWC has gained increasing prominence among researchers and industrial practitioners. DWC is considered as an exciting path for PI in multicomponent distillation, since it reduces the capital cost by as much as 30% over conventional distillation configurations (Dejanovic et al., 2010). Moreover, for a specific separation task, it allows process engineers to design an array of configurations that uses all the way from 1 to  $(n-2)$  columns, as opposed to conventional configurations which generally use  $(n-1)$  columns (Madenoor Ramapriya et al., 2018a). Despite these potential benefits, the number of DWCs currently in implementation is minuscule compared to the number of conventional distillation columns in operation. Also, almost all industrial implementations of DWC have been for separating a feed into three product streams or a 3-component separation.

The major reason why DWCs have not been implemented in large scale is because conventional DWCs face difficulties in achieving the desired vapor split at the bottom of the vertical partition (Madenoor Ramapriya et al., 2014). Consider a 3-component DWC shown in Figure 4b. This DWC is derived from the FTC configuration of Figure 4a. Clearly, the external independent control on the bottom vapor split is missing. As a result, the vapor flows in the two parallel zones on either side of the vertical partition are left unregulated in this DWC. Therefore, it is difficult to operate the DWC at the desired  $L/V$  ratio in either parallel zone that corresponds to optimal operating condition. This leads to inferior performance and hence much higher energy consumption in practice. It also explains why higher-component DWCs have not been implemented in industry extensively, as there exists more than one vertical partitions for such distillations and thus more than one unregulated vapor split.

To overcome this operational difficulty associated with the DWC of Figure 4b, Madenoor Ramapriya et al. (2014) realized that the solution lay in the original FTC configuration. The authors proposed a versatile methodology of “converting thermal couplings to liquid-only transfer” for the FTC configuration to derive thermodynamically equivalent, but operable configurations, from which operable DWCs can be systematically drawn. Conversion of a thermal coupling to liquid-only transfer stream was first proposed by Agrawal (2000). This strategy enables one to draw an array of new configurations that consume the same total vapor duty as the original thermally coupled configuration, but eliminate the use of thermal couplings which can cause controllability issues. For a detailed discussion about how the conversion works and a rigorous mathematical proof of thermodynamic equivalence, the reader is referred to Madenoor Ramapriya et al. (2014).

Consider the example of converting thermal coupling AB in the FTC configuration of Figure 4a to liquid-only transfer. This is done by adding an additional column section to the top of the prefractionator column, followed by adding a condenser to the additional section to withdraw some pure product A. In this way, the thermal coupling at AB, which is a two-way liquid-vapor transfer, is now made as a liquid-only stream. The resulting configuration is shown in Figure 4c. Similarly, one may also convert the thermal coupling BC in the FTC configuration of Figure 4a to liquid-only transfer. And Figure 4e shows the resulting configuration when both thermal couplings AB and BC are converted to liquid-only transfers. Compared with the original FTC configuration of Figure 4a, these new configurations of Figure 4c and 4e have the same total vapor duty requirement, but are more operable because the desired vapor split between the two distillation columns can now be easily achieved by regulating the flow rates of the liquid-only transfer streams using valves as well as

manipulating heating and/or cooling duties associated with the added reboilers and/or condensers. The strategy of “converting thermal couplings to liquid-only transfer” allows one to systematically enumerate all DWCs for any  $n$ -component FTC configuration, as well as to identify the easy-to-operate ones (Madenoor Ramapriya et al., 2016). Some of the easy-to-operate 3-component FTC DWCs are shown in Figure 4 (Madenoor Ramapriya et al., 2014). Instead of knowing only a few DWCs which are not operable, industrial practitioners can now generate hundreds to thousands of easy-to-operate FTC DWCs!

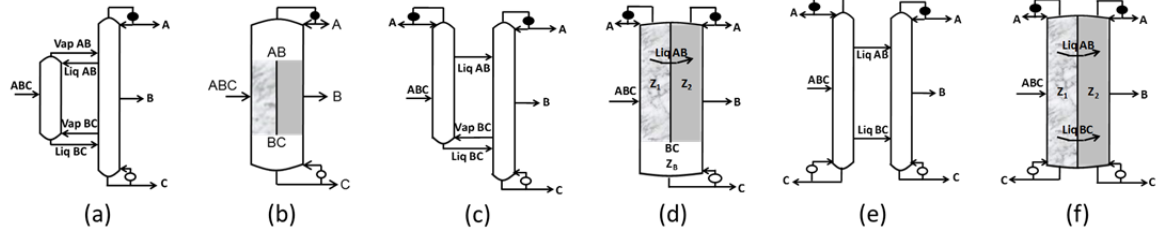


Figure 4: (a) 3-component FTC; (b) Conventional DWC; (c) Configuration derived from (a) by converting thermal coupling AB into liquid-only transfer; (d) Resulting DWC from (c); (e) Configuration derived from (a) by converting thermal couplings AB and BC into liquid-only transfers; (f) Resulting DWC from (e).

Of course, this strategy can, not only be applied to draw all FTC DWCs, but also to draw DWCs for any thermally coupled configuration, including the HMP configuration discussed earlier. Recently, Madenoor Ramapriya et al. (2018a) developed an intuitive six-step method to draw the DWC for any given thermally coupled configuration. Shortly after, they were able to extend the same concept of “thermal coupling to liquid-only transfer” to systematically synthesize thermodynamically equivalent but easy-to-operate DWCs for every thermally coupled configuration (Madenoor Ramapriya et al., 2018b). For example, the easy-to-operate DWC shown in Figure 5b is derived from the HMP configuration of Figure 3b by first converting all bottom thermal couplings at DE and CDE to liquid-only transfers as shown in Figure 5a (Jiang et al., 2018). The operable DWC of Figure 5b derived from the HMP configuration uses only 2 vertical partitions and 5 submixture transfers, as opposed to three vertical partitions and 9 submixtures in the FTC DWC of Figure 5c. As a result, synergistic use of both PI strategies discussed in this paper can lead to many new attractive configurations. Finally, it is worth noting that, while Figure 5b shows the version using only one distillation column, versions that use two distillation columns can also be drawn by combining only columns 1 and 2 or 2 and 3-4 of Figure 5a through the use of DWC.

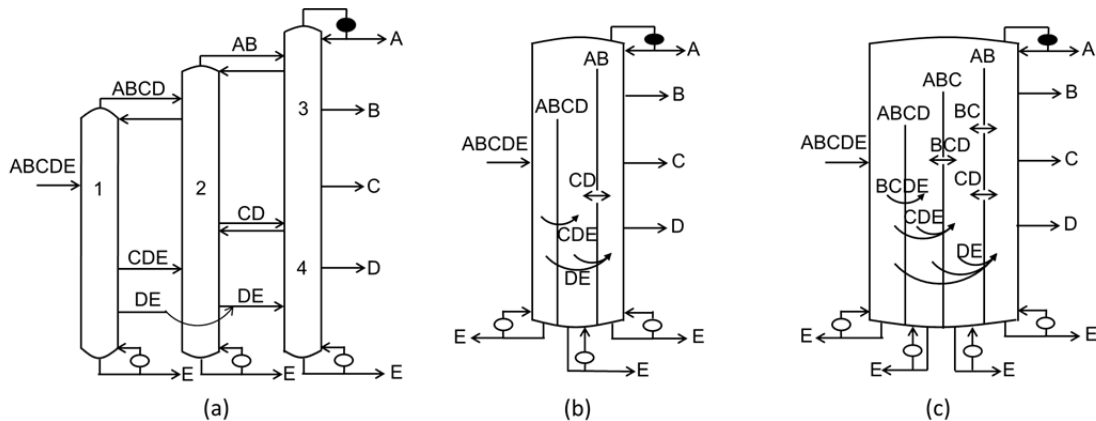


Figure 5: (a) Thermodynamically equivalent version of HMP configuration of Figure 3b; (b) The resulting easy-to-operate DWC version; (c) An operable DWC version of the FTC configuration.

#### 4. Conclusions

Process Intensification has gained great attention in the chemical industry. In this paper, two PI strategies are introduced that allow industrial practitioners to build compact, easy-to-operate, energy efficient and cost effective multicomponent distillation configurations. In the first strategy, combining thermal coupling and simultaneous HMA leads to a new set of HMP configurations which not only reduces the number of columns and heat exchangers, and the associated pipes and fittings, but at the same time, reduces the energy consumption significantly relative to the original basic configuration. For several feed conditions, the HMP

configurations require the same minimum total vapor duty as the FTC configuration, making them very attractive to build and operate in practice. In the second strategy, a systematic approach is introduced to synthesize all DWCs for any thermally coupled configurations, including those that are easy-to-operate ones. The strategy of converting thermal coupling to liquid-only transfer streams removes the longstanding challenge related to the operation of DWCs involving the control of vapor splits, which has hindered implementations of DWC on large scale. These easy-to-operate DWCs consume the same energy, yet they can save significant capital investment relative to the original thermally coupled configuration. Both strategies, especially when used synergistically, present great opportunities for PI in multicomponent distillation. Additional design methodologies, such as the use of operable double- or multi-effect distillation to significantly reduce the heat duty required for a given separation, will be considered and included in the future to further enrich the concept of PI in multicomponent distillation.

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