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Complex Distillation Arrangements : Extending the Petlyuk Ideas

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Abstract

The task of separating a multicomponent mixture into streams enriched in the respective constituents is commonly carried out in conventional distillation columns arranged in series. However, due to the scrutiny of tighter requirements for energy and cost efficiency, current research aims at alternative column arrangements that offer savings in both operational (energy) and capital costs. Among these are the Petlyuk or dividing wall column, in which three components are separated in a single shell using only one reboiler and one condenser. In this paper we extend the Petlyuk ideas to separations of four components, although extensions to more components is straightforward. We provide a general definition of *Petlyuk arrangements* and discuss alternative structures from the literature. Following this overview we consider the arrangements which allows for implementation in a *single shell* using dividing walls or vertical partitions.

INTRODUCTION

Industrial distillation processes are commonly known to be highly energy–demanding operations. Recent surveys indicate that energy inputs to distillation columns account for roughly 3% of the *total* energy consumption in the U.S. (Ognisty 1995). For this reason there is ample scope for developing more energy efficient separation schemes. In order to reduce energy consumption at least two alternative approaches have been proposed both in the literature and by industrial practicionairs. These approaches subscribe to either integrating conventional distillation arrangements, or to the design of new configurations. The former approach typically involves distillation columns arranged in series with energy integration between columns or other parts of the plant. Among the "new" configurations that offer both energy and capital savings we find the dividing wall column first proposed by Wright (1949). Beloved children are known by many names, and this arrangements is also known as the Petlyuk column, due to a theoretical study of Petlyuk *et al.* (1965), or as a *fully thermally coupled* column (Triantafyllou and Smith 1992). In order to provide a common framework for future work, we define a *Petlyuk arrangement* as follows :

A column arrangement separating three or more components using a single reboiler and a single condenser, in which any degree of separation (purity) can be obtained by increasing the number of stages (provided the reflux is above a certain minimum value).

Use of this definition eliminates for example a conventional sidestream column from being considered as a Petlyuk arrangement, since these require infinite reflux to obtain a pure sidestream product (even with an infinite number of stages).

For separations of ternary mixtures ($n = 3$), the Petlyuk Column is represented by the well known configurations given in figure 1. We emphasize that the two representations are identical from a computational point of view if we neglect heat transfer across the dividing wall.

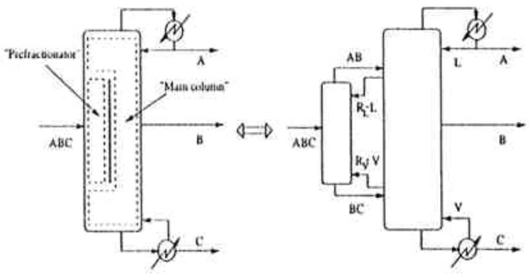

Figure 1: Left : Petlyuk (dividing wall) column
Right : Equivalent prefractionator arrangement.

Although the Petlyuk arrangement shown in figure 1 have been known for almost 50 years (Wright 1949), it has only quite recently gained interest also in industry. The dividing wall column has nevertheless been the subject of several theoretical studies (see e.g. Petlyuk *et al.* (1965), Kaibel (1987), Triantafyllou and Smith (1992) and Wolff and Skogestad (1995)), in which it is reported that for $n = 3$ it requires typically 30% less energy input compared to conventional arrangements using simple columns in sequence. Due to implementation in only one shell, and savings of one reboiler and one condenser, the capital savings are also typically in the order of 30% (Smith 1995). The literature on mixtures with more than three components ($n > 3$) is relatively scarce. Among

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the few contributions are some ideas presented by Kaibel (1987). However, neither detailed analysis nor computational results are presented.

The main contribution from our work lies in providing a systematic framework for analysis and design of Petlyuk arrangements for separations of mixtures with four or more components. Let n here denote the number of components in a mixture to be separated into its pure constituents. For a conventional scheme consisting of a sequence of *simple* columns, it is well known that this requires a minimum of $2(n-1)$ sections with $(n-1)$ reboilers and $(n-1)$ condensers. Here a section denotes a part of the column from which no streams enter or leave. One penalty for using only one reboiler and one condenser is an increase in the required number of sections.

In order to derive the "optimal" scheme from all possible sequences, various methods have been presented in the literature. The mathematical problem may be formulated as a MINLP–problem to be solved by some optimization–algorithm. However, for a large number of components, one in practice often fails to locate the global optimum due to non–convexities and computational issues. To overcome these limitations, heuristics and evolutionary strategies have been proposed to guide the engineer in choosing from the set of possible arrangements (e.g. (Tedder and Rudd 1978)). Among the most important tasks when seeking to find the optimal column arrangement, is that of deriving a general *superstructure* which incorporates all other configurations as substructures. In this work we consider three different approaches for arriving at such a superstructure for Petlyuk arrangements, based on the previous works of Sargent and Gaminibandara (1976), Agrawal (1996) and Kaibel (1987).

SHARP SPLIT ARRANGEMENTS

It is common practice within theoretical studies on batch, continuous and complex distillation columns to infer the separation of a given mixture in terms of *sharp splits*. For instance, Cerda and Westerberg (1981) use the word *sharp* for the case where the recoveries of light and heavy key are *"close to one"*. However, the sharpness of the splits obviously depends on a number of factors such as the structure of the column, the number of stages, the reflux and the thermodynamic properties (e.g. relative volatility). In this paper we are mainly interested in the structure (arrangement) of the columns and we propose the following definition:

A sharp split arrangement is an arrangement of columns in which any degree of separation (purity) can be obtained by increasing the number of stages (provided the internal refluxes are above certain minimum values and provided the separation is thermodynamically feasible).

A Petlyuk column is then a sharp split arrangement with a single condenser and a single reboiler. To clarify the above definition, we note that a special property of distillation columns is that any degree of separation (purity) can be achieved by increasing the number of stages. In order to illustrate this point, consider first the McCabe–Thiele diagrams in figure 2.

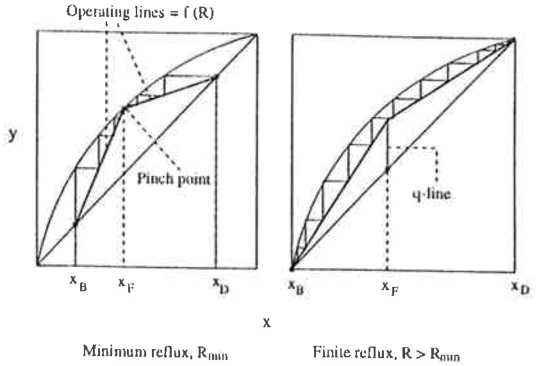

Figure 2: McCabe-Thiele diagrams

In the case of limiting flow conditions (minimum reflux R_{min}), a pinch zone occurs in the vicinity of the feed point, requiring a large number of stages in this section. However, by increasing the number of stages, and allowing for a *finite* increase in R, we may in fact achieve any purity. This relation is also revealed if we consider the approximate expression for the *separation factor* as derived by Skogestad and Morari (1987) for a binary mixture with constant relative volatility α:

$$S \stackrel{\text{def}}{=} \frac{x_T/(1-x_T)}{x_B/(1-x_B)} \approx \alpha^N \frac{(L/V)_T^{N_T}}{(L/V)_B^{N_B}} \qquad (1)$$

where T and B denote the top and bottom respectively. We see clearly that $S \rightarrow \infty$ when $N \rightarrow \infty$, whilst $S \rightarrow \alpha^N$ when $L \rightarrow 1$ (total reflux). The latter is the well known Fenske equation which yields the minimum number of stages N_{min} for a given separation.

SUPERSTRUCTURES FOR PETLYUK ARRANGEMENTS

A simple way to compare the different column arrangements and "superstructures", is provided by the network in figure 3 (e.g. Agrawal (1996)). Such networks yield convenient visualizations of which splits that are actually carried out in the various sections. In such a network, the feed represents a node, whereas each line connecting neighboring nodes represents a column section, i.e. a stripping or rectifying section. The intermediate nodes represent streams that are passed from one two–sectional unit to another. The column configuration corresponding to the network in figure 3 consists of $n(n-1)$ sections (= 12 sections for $n = 4$). It is possible to eliminate some of the intermediate nodes in the network, thus decreasing the number of sections. However, we note that any structure with less than $n(n-1)$ sections, by virtue cannot produce only *"reversible splits"* (Petlyuk *et al.* 1965). For a "reversible split", only the components with the highest and the lowest boiling points should be separated in each section (Petlyuk *et al.* 1965).

We first consider the superstructure proposed by Sargent and Gaminibandara (1976) consisting of $n(n-1)$ sections as shown in figure 4. As indicated, the authors also incorporate the option of additional heating and cooling in each column section. As the authors note, the number $n(n-1)$ actually represents the maximum number of sections in sharp split arrangements for an n–component separation, as all nodes in figure 3 are included.

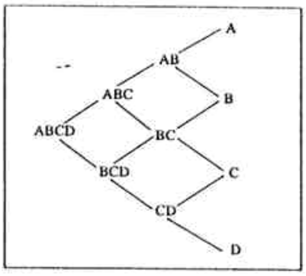

Figure 3: Network representation of possible separations involved in separating 4–component mixtures.

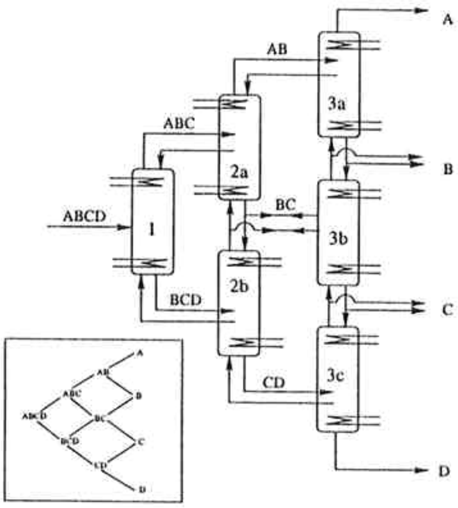

Figure 4: Sequence proposed by Sargent and Gaminibandara with $n(n-1) = 12$ sections for $n = 4$ (gives Petlyuk arrangement by deleting intermediate heaters and coolers)

According to the authors, this superstructure contained all *functionally possible* column arrangements as substructures. However, as we show below, the proposed superstructure actually fails to do so (see e.g. figure 6 and 7). This means that some potentially interesting column arrangements cannot be obtained by removing either column sections or flows from the superstructure.

In a recent article, Agrawal (1996) proposes an alternative superstructure for a certain subclass of Petlyuk arrangements. By considering arrangements with $n-2$ *satellite columns* in communication with a central distillation column, he arrives at the superstructure shown in figure 5. Agrawal claims that this superstructure includes as substructures all previously proposed configurations giving "sharp splits", which in fact is not quite true as we will illustrate by a structure proposed by Cahn *et al.* (1962) and later by Kaibel (1987). In any case, Agrawal's superstructure is more general than Sargent's arrangement, in the sense that fluid transfer may take place between *any* of the interconnected columns, and it includes Sargent's superstructure and also Kaibel's and Cahn's arrangements as substructures. We furthermore note that in Sargent's sequential structure, there is no direct fluid flow between the first and the last columns. This conceptual difference between the "superstructures' owes to the fact that Sargent considers $n-1$ interconnected distillation columns *in sequence*, whereas Agrawal's superstructure consists of $n-2$ *satellite columns* arranged around a central distillation column.

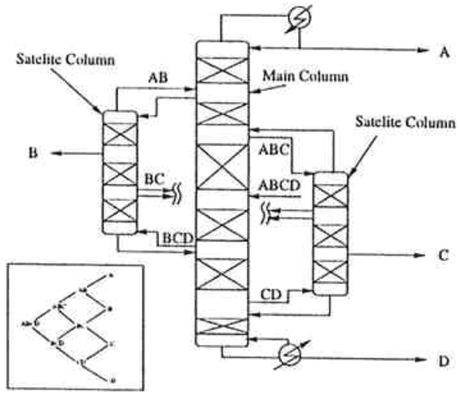

Figure 5: Satellite column arrangement proposed by Agrawal with $n(n-1) = 12$ sections for $n = 4$

Agrawal derives by simple arguments that the *minimum* number of rectifying and stripping sections required for sharp splits using such *satellite arrangements*, is equal to $4n - 6$ (10 sections for $n = 4$). These may be obtained by deleting the BC, ABC or BCD node from the network in figure 3. By deleting for example the BC node we obtain such a structure with 10 sections as shown in figure 6. For $n \geq 4$ this is considerably less than $n(n-1)$ as suggested by Sargent and Gaminibandara (1976). However, we also note that $4n-6$ in fact *is not* the minimum number for sharp split arrangements, as illustrated by considering the Kaibel column in figure 7.

We ask the reader to note that in figure 11 we will demonstrate how the arrangements in figures 6 and 4 may be implemented in a single shell with two vertical partitions.

In the work of Kaibel (1987), columns consisting of *vertical partitions* are considered, based on the *dividing wall* column previously described by Wright (1949). Although Kaibel analyzes in detail only the case of $n = 3$, he also indicates interesting arrangements for $n \geq 4$. The dividing wall column proposed by Kaibel for $n = 4$ is given in figure 7. In order to understand how the *Kaibel column* may result, and how pure products can be obtained in the sidestreams, we draw attention to the schematic in figure 8.

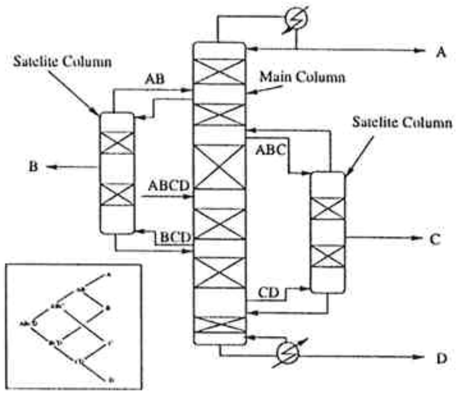

Figure 6: Agrawal's substructure with *"minimum"* number of sections, $4n-6 = 10$ sections for $n = 4$

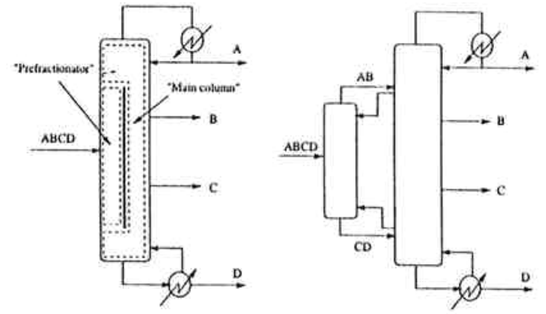

Figure 7: Kaibel's dividing wall column for $n = 4$

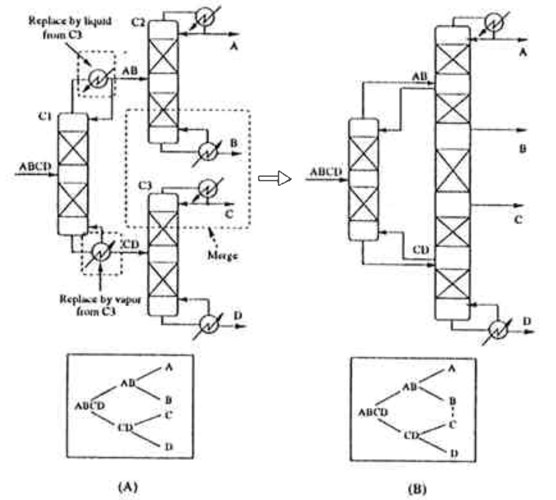

Figure 8: Steps towards the Kaibel column.
(A) : conventional arrangement. (B) : arrangement after (1) merging columns $C2$ and $C3$ (2) adding an intermediate section and (3) taking boilup and reflux for $C1$ from $C2$ and $C3$.

The column consists of 7 sections, which is considerable less than Agrawal's *"minimum"* number of 10. The reason for this "inconsistency" is that the Kaibel arrangement consists of only $n - 2 = 2$ columns, whereas Agrawal only considers satellite arrangements with $n-1$ columns. Comparing the network representations given in the bottom of figure 8 with the network in figure 3, we see that Kaibel's structure corresponds to eliminating the ABC, BCD and BC nodes, whereas one section is added between the B and C nodes. It is also easily derived from both Agrawal's and Sargent's superstructures. Ideally, this latter section should act as a *total reflux* column ($L/V = 1$), in which "heat integration" of the two columns $C2$ and $C3$ in figure 8 is facilitated. We thus recognize that it in fact has no designated separation task. Its task is simply to transfer heat while avoiding remixing of the already separated components (B and C). Thus, total reflux is needed to avoid net transport of components between the two sidestreams. Further, since we require that only A and B should enter the main column from the top, and only C and D from the bottom, we see that the Kaibel column violates the requirement for a potentially "reversible split" for which only the lightest and heaviest component should be removed at each stage (e.g. (Petlyuk *et al.* 1965) or (King 1980)). Thus, one should bear in mind that *any* arrangement with less than $n(n-1)$ sections introduces additional irreversibilities, which most likely increases the required energy input. In another paper (Christiansen *et al.* 1996) we show that there are additional difficulties with respect to the operation of the Kaibel column.

Having considered some "superstructures" for Petlyuk arrangements, we now focus on how these may be implemented in single shells with dividing walls.

DIVIDING WALL COLUMNS

A benefit of a Petlyuk arrangement with a single reboiler and a single condenser is that it may be realized in a single shell with dividing walls, which possibly yields capital savings in addition. In this work we consider arrangements with only *vertical* paritions, in which we also allow for *communication points* between neighboring sections. In order to cope with some inadequacies of the conventional *dividing wall* columns, we also introduce some novel geometrical wall structures such as the *triangular* wall column. From these superstructures we also demonstrate how one may derive arrangements with the "minimum" number of $4n - 6$ sections. Before going into detailed discussion on these particular designs, we elaborate some on the large number of degrees of freedom offered by Petlyuk arrangements.

Degrees of freedom (DOF) analysis. When analyzing the degrees of freedom for a given process, one should in the general case distinguish between degrees of freedom (DOF) for *design* and the DOFs for *control* (operation). In this paper we consider only the latter, hence we restrict ourselves to columns with fixed number of stages, feed location(s) and feed condition(s). These variables off course must be taken into account for optimization purposes, i.e. optimal design.

Assuming that the holdups and the pressure are controlled, conventional binary columns yield two potentially manipulated variables, e.g. reflux (L) and boilup (V). For columns with vertical partitions, we gain in general one DOF for each sidestream and two for each dividing wall (the vapor and liquid split). Hence, the following formula yields the number of operation DOFs for a column with n_S sidestreams and n_D dividing walls

$$DOF = 2 + n_S + 2 * n_D \qquad (2)$$

For Petlyuk columns with one dividing wall (figure 1), this gives 5 DOFs at steady state, and with two walls 8 DOFs result.

If we also allow for the possibility of having liquid and vapor transport (communication) between certain stages on both sides of a wall, there are in fact <u>four</u> streams which may be redistributed (liquid and vapor on each side). Thus, we add yet another four degrees of freedom for each *communication point.* In Figure 9 we give an illustration of the additional liquid and vapor splits due to fluid transfer through the communication point.

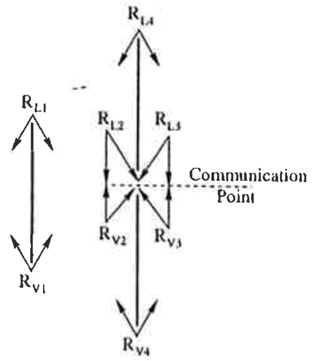

Figure 9: Additional DOFs due to communication between partitions

To avoid confusion in the proceeding discussion of columns with such communication points, we make the somewhat fictious distinction between *dividing walls* for the overall structure and *vertical partitions*. Hence, we may have a column with two dividing walls and three partitions as illustrated in figure 10. The total number of DOFs for a structure with n_C communication points is thus

$$DOF = 2 + n_S + 2 * n_D + 4 * n_C \qquad (3)$$

For the structure in figure 10 we thus have, according to equation (3), a total of $2 + 2 + 2 * 2 + 4 = 12$ DOFs for operation. However, from physical insight we conjecture that fluid should only be transported in the direction towards the final products, i.e. with $R_{L3} = R_{V3} = 0$ in figure 9. The liquid and vapor splits in the middle partition (R_{L2} and R_{V2}) then constitutes an intermediate feed *to* the sidestream side. The *"feed condition"* thus depends on the relative amounts of R_{L2} and R_{V2}. The optimal condition of this feed could be optimized, but for practical purposes it is much easier to transport only liquid across the partition (i.e. $R_{V2} = 0$). Taking the latter observations into account, 9 potential DOFs remain. As a comparison, the structure proposed by Kaibel yields only 6 DOFs.

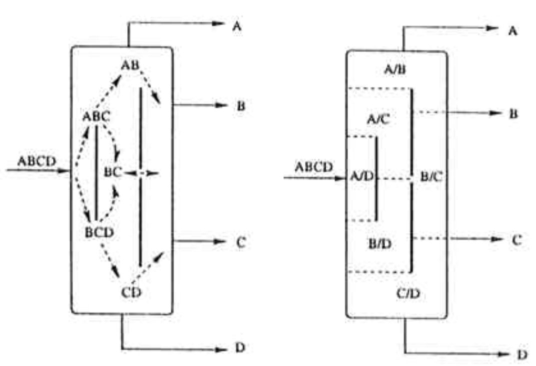

Figure 10: Distribution of components for dividing wall column given in figure 4

Dividing wall implementations for $n = 4$. The arrangement illustrated in figure 10, which is a special case of Sargent's superstructure, consists of $n(n-1) = 12$ sections. This is as previously noted the "maximum" number of sections required for a sharp split arrangement. An illustration of the corresponding sequence of binary splits is given in the right schematic. We have assigned all splits corresponding to the different two–sectional "columns", and also indicated the "direction" of flows for the various subgroups. In figure 11 we show dividing wall implementations of the Kaibel, Sargent and Agrawal arrangements.

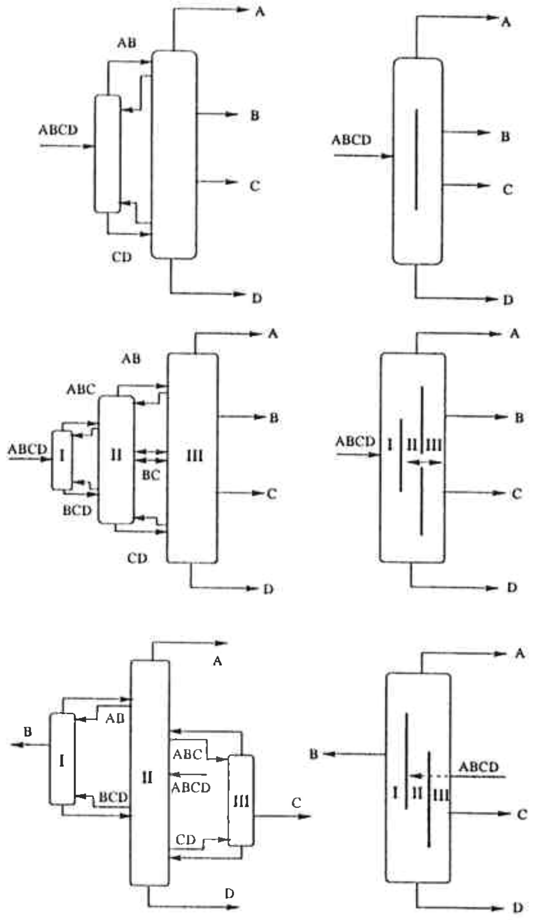

Figure 11: Dividing wall implementations of the Kaibel (top), Sargent (middle) and Agrawal (bottom) arrangements

For the Agrawal arrangement we recognize that the feed should enter the dividing wall column from the middle partition. This is perhaps more clearly understood if we consider the column viewed from the top as demonstrated in figure 12.

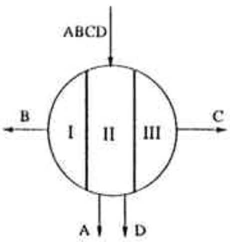

Figure 12: Top view of dividing wall column

Agrawal (1996) argues that there are 3 different satellite arrangements corresponding to the "minimum" of 10 sections for a 4–component separation. If we consider the dividing wall implementation in figure 12, we find that it allows for only one possible arrangement with 10 sections, because no communication is allowed between parts *I* and *III*. However, the *triangular structure* in figure 13 allows for interconnections between any two neighboring *parts* of the column. Hence, we may in principle allow for communication between any two stages in the column arrangement.

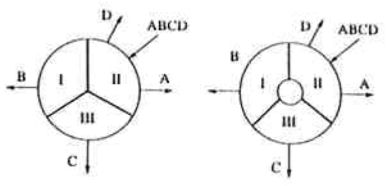

Figure 13: Top view of triangular wall structure

At right in figure 13 we have indicated that one may implement a tube along the center section in order to allow for fluid transport between various sections. In practice this is achieved by withdrawing fluid from one stage and passing it to the appropriate stage in the corresponding section.

NUMERICAL RESULTS

In order to compare the energy efficiency for the different Petlyuk arrangements proposed in the previous sections, we give here some preliminary numerical results for a case where the relative volatility between the four components is 8 : 4 : 2 : 1. For all columns we use 40 stages counting from the bottom to the top of each column. The results are computed from optimization of relatively simple models, for which we assume constant molar flows and constant relative volatility. For more details we refer to Christiansen *et al.* (1996). In Table 1 we give the minimum energy inputs (boilup to feed ratio V/F), the number of sections and the number of DOFs used for optimization for two mixtures of different feed composition. $(V/F)_A$ corresponds to an equimolar feed, i.e. all feed compositions are $z_i = 0.25$, whereas $(V/F)_B$ corresponds to $z_A = z_D = 0.4$ and $z_B = z_C = 0.1$. In all cases the purity specifications for the products are 99% in the top and bottom and 95% for the side products. The Agrawal arrangement with 10 sections (figure 6) is seen to be the better for this study. However, even though the Kaibel arrangement requires the largest heat input, it still constitutes an interesting alternative taking into account its simplicity. Initial results (not shown) for Agrawal's superstructure with 12 sections (figure 5) yields even lower values, typically in the order of $(V/F)_B = 1.60$.

Table 1 : Minimum energy inputs

	Sections	DOFs	$(V/F)_A$	$(V/F)_B$
Kaibel	7	6	2.58	1.99
Sargent	12	12	2.27	1.76
Agrawal	10	8	**2.11**	**1.65**

CONCLUSIONS

In this paper we have extended the Petlyuk ideas to dividing wall columns that permit multicomponent separations within a single shell. In particular we have addressed different *superstructures* proposed in the literature for arrangements with $n-1$ interconnected columns, and demonstrated how such arrangements may be implemented in a single shell with vertical partitions. We have briefly discussed the extent to which such arrangements allow for potentially reversible splits, which strongly influences the required exergy input.

In order to provide direction and a consistent framework for future work, we have also proposed definitions of what is to be referred to as *Petlyuk arrangements* and *sharp split arrangements*. A discussion on how to utilize the large number of degrees of freedom for such column arrangements is also given in some detail. In this respect we suggested simple formulas for computing the number of DOFs for Petlyuk arrangements. For design and optimization purposes, we find that the number of DOFs become excessive if all variables may be set arbitrarily. An issue of great importance for future work thus rests in providing guidelines which implicitly reduce the set of DOFs. In particular we will aim at finding adequate relations between the corresponding liquid and vapor splits to be used for operation.

Finally we introduced a novel geometrical column arrangement, the *triangular wall*, to overcome some limitations of conventional dividing wall columns with respect to flexibility.

Acknowledgment. *Valuable comments from John Morud are gratefully acknowledged*

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