PARTITIONED PETLYUK ARRANGEMENTS FOR QUATERNARY SEPARATIONS

Atle C. Christiansen, Sigurd Skogestad[1] and Kristian Lien
Dept. of Chem. Eng., Norwegian University of Science and Technology, N–7034 Trondheim Norway

The task of providing energy efficient separation arrangements have received considerable attention in the literature. The conventional approach to increasing the process efficiency subscribe to integrating conventional distillation arrangements (indirect coupling). Instead, there has recently been a growing interest in the development of new configurations (unit operations) that offer both operational (energy) and capital savings. Among these we find the Petlyuk or dividing wall columns (direct coupling). In this paper we compare the energy consumption in optimized Petlyuk arrangements with that of optimized sequences of regular columns. Our results are based on simulations using a detailed model. We also introduce a novel column arrangement utilizing both direct and indirect coupling, for which we propose to use also a *horizontal* partition in order to avoid remixing of already separated components.

Keywords: Complex distillation arrangements, energy efficiency, optimization.

INTRODUCTION

A schematic of the well known Petlyuk (Petlyuk *et al.* 1965) column, or dividing wall column (Wright 1949), for the separation of ternary mixtures is illustrated in Figure 1. This column has received consid-

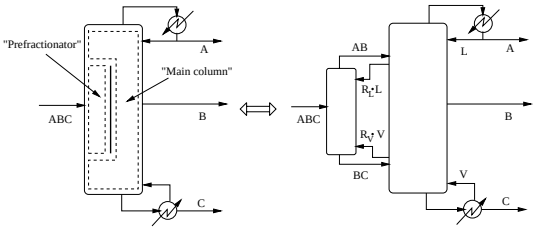


Figure 1: Petlyuk column. Left : Column with vertical partition. Right : Prefractionator arrangement

erable attention in the literature, and it is claimed that savings in capital and energy costs are typically in the order of 30% compared to conventional arrangements with regular columns in sequence (e.g. Smith (1995)), although Glinos and Malone (1988) report that the maximum savings are as large as 50%. In this paper we extend the Petlyuk ideas to also consider quaternary separations, and we compare the energy consumption in optimized Petlyuk arrangements with that of optimized sequences of three regular columns. In order to provide a common basis for our work, we use the following definition:

> *A Petlyuk arrangement is 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 and the separation is thermodynamically feasible).*

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

In another paper (Christiansen *et al.* 1997) we presented a systematic framework for analysis and design of Petlyuk arrangements for separating mixtures of four or more components. We considered two *"superstructures"* previously proposed in the literature and showed how these may be implemented in single shells

[1]Author to whom correspondence should be addressed : Fax : +47 7359-4080, E-mail: skoge@kjemi.unit.no

using vertical partitions. In particular we considered the *sequential arrangement* proposed by Sargent and Gaminibandara (1976) and the *satellite arrangement* by Agrawal (1996), both corresponding to structures with $n-1$ interconnected columns. We also considered a simple structure proposed by Cahn *et al.* (1962) and later by Kaibel (1987). In the present paper we further explore the concept of complex distillation arrangements in single shells with multiple dividing walls or *vertical partitions*, using a single reboiler and a single condenser. We also introduce a novel arrangement, called the $\vdash$ column. The distinguishing feature of this design is that it also uses a *horizontal partition* to facilitate energy integration, so that remixing of already separated components is avoided.

In the literature it is generally agreed that the total vapor consumption (boilup) is the dominant variable when estimating the total cost (operation and capital) of distillation columns (e.g. Glinos and Malone (1988), Tedder and Rudd (1978)). In this paper we thus present numerical results for Petlyuk arrangements optimized with respect to the boilup consumption, and compare the results to optimized schemes of regular columns in sequence. The optimization problem is solved using a gradient projection method embedded in a continuation scheme, which we found to be very efficient. This algorithm is due to the partly unpublished work by Morud (1995), and is also an extension of a method described in a previous work by the authors (Christiansen *et al.* 1996). The principles and further details regarding this method is to be presented in a future work. Our results are based on simulations using a detailed model.

ONE DIVIDING WALL (THE KAIBEL COLUMN)

We first consider the simplest extension of the Petlyuk column, given in Figure 2, for the separation of a quaternary mixture $ABCD$ into its constituents. The arrangement to the right is due to an early patent by

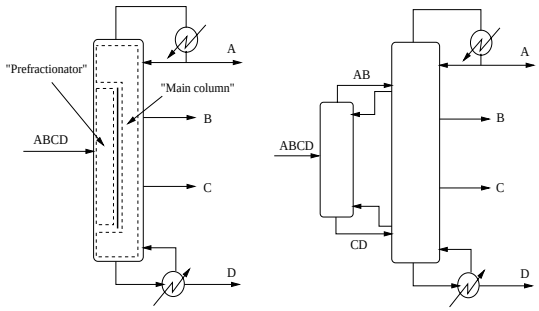


Figure 2: Petlyuk arrangement with one dividing wall for quaternary separations.

Cahn *et al.* (1962) whereas the dividing wall column to the left was later proposed by Kaibel (1987). Since only the latter considered the possibility of implementing columns in a single shell, we in the following denote this arrangement the *Kaibel* column. Compared with the Petlyuk column in Figure 1 for ternary separations, we first acknowledge that the requirement for a potentially *reversible split* is violated. For a reversible split only the components with the highest and the lowest boiling points should be separated at each step (see e.g. Petlyuk *et al.* (1965) or King (1980)), in this case requiring a first split between A and D in the prefractionator. However, for the *Kaibel* column we recognize that to obtain *pure* products, a sharp split between the intermediates B and C is required in the prefractionator. If any B enters the main column from the bottom of the prefractionator, some B necessarily leaves the main column with the sidestream where C is drawn off. Similarly, if any C enters in the vapor over the top, a certain fraction of C is withdrawn in the B–sidestream. The latter argument does however not imply exact *symmetry*, in the sense that for any ideal multicomponent mixture including B and C, the composition of B is by virtue higher in the vapor phase, and the corresponding composition of C higher in the liquid. Hence, if assume that the products are withdrawn as liquid and require equal product purities of B and C, we may allow for a larger fraction of B to enter the main column from the bottom relative to the amount of C entering over the top of the prefractionator.

Furthermore, we note that the section between the sidestreams in the *Kaibel column*, in fact has no des-

ignated separation task, as far as we require that only A and B should enter the main column from the top, and only C and D from the bottom. This leaves only the two binary separations of A/B and C/D for the main column. Ideally, this section should thus act as a *total reflux column* with $L = V$, since we want no net transport between the pure B in the upper sidestream and the pure C in the lower side stream. However, B and C might undergo remixing, which in case represents a source of thermodynamic inefficiency. The impact of this remixing is however counteracted if the intermediate section is operated under total reflux and with a certain minimum number of stages (N_{min}^{BC}), where N_{min}^{BC} may be obtained from the well known Fenske equation which applies to any column or column section;

$$N_{min} = \frac{log\,(S_{LH})}{log\,(\alpha_{LH})}, \;\; S_{LH} = \frac{x_{L,T}/x_{H,T}}{x_{L,B}/x_{H,B}} \tag{1}$$

Here S_{LH} denotes the separation factor, α_{LH} the relative volatility and $x_{L,T}$ and $x_{H,B}$ the purities of the light and heavy keys in the top (T) and bottoms (B) of the column or column section. In practice it may however be very difficult to obtain $L/V = 1$ in the intermediate section, which may prevent high purities of the sidestreams.

INTRODUCING THE ⊢ COLUMN

In order to overcome these operational problems with the *Kaibel* column, we introduce a novel arrangement with a *horizontal* partition between the sidestream outlets for B and C, as illustrated in Figure 3. We stress that the two representations in Figure 3 are identical from a computational point of view, if heat transfer across the partitions is neglected.

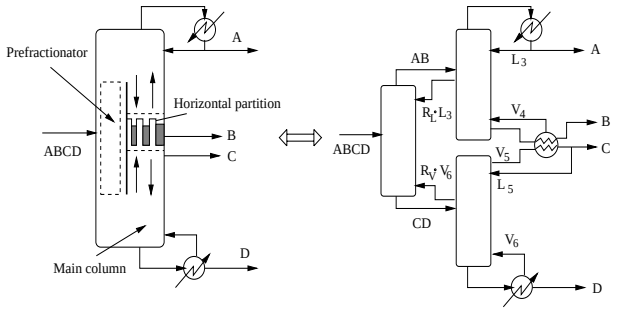


Figure 3: ⊢ column with vertical and horizontal partition for quaternary separations

Providing a conceptual interpretation of the ⊢ column, we note that the prefractionator is linked via *direct* (thermal) coupling (Petlyuk idea), whereas there are two indirectly coupled (heat integrated) "simple" columns in which waste heat is utilized. Thus, according to our definition it is not a Petlyuk arrangement in a strict sense, for which only a single reboiler and a single condenser should be used. As illustrated in Figure 3, the heat exchange may occur either within the column shell (left figure) or in an external heat exchanger (right figure). In the former case the heat exchange may take place for example using a dephlagmator, inside which condensation continuously takes place on the hot side and evaporation on the cold side. The choice of heat exchange "unit", either internal or external, is obviously a matter of practicality as well as economy, depending for instance on the required heat transfer area. If the ⊢ column is to be operated without using additional utilities, the vapor flows V_4 and V_5 in the two heat integrated "simple" columns are directly coupled. For instance, in the case of constant molar flows it is required that $V_4 = V_5$. Such a direct coupling may be desirable in a *balanced column*, that is, in a column where the separation difficulty in terms of required boilups V_4 and V_5 are similar for the two columns[2]. However, in general this strong coupling may be undesirable, and one may want to introduce additional heating or even cooling to eliminate the coupling between V_4 and V_5 (this disadvantage also applies to the Kaibel column). The single shell ⊢

[2]Note that both mixtures considered in this work allow for balanced columns since the feed compositions of intermediates are equal

column is in any case a compact and thus cost efficient arrangement, which should be considered whenever there is a sufficient difference in boiling points for the intermediate components. In case of limiting driving forces one might also consider using a heat pump to raise the temperature level of the available heat, or as mentioned use additional utility. The $\vdash$ column should also be relatively easy to operate, and startup may for instance take place simply by running the column under total reflux until there is sufficient buildup of liquid on the cold side.

Tedder and Rudd (1978) and Nikolaides and Malone (1988) have previously considered designs which have *some* of the same features as the $\vdash$ column, but there are at least three important distinctions. Firstly, the previous authors considered only *ternary* mixtures, whereas the $\vdash$ column may also separate quaternary mixtures. Secondly, they only considered heat exchange to take place between columns run at different pressures, which is required for sharp ternary separations since the hot and cold flows have the same composition (i.e. "pure" intermediate component). Last, but not the least, no authors have to our knowledge considered a design with vertical and horizontal partitions in a *single shell*. In an overall (ecological) perspective the simplicity of building a single shell in itself offers energy savings, since production of materials off course requires energy. Before extending the analysis to arrangements with two dividing walls, we give a brief analysis of the number of degrees of freedom (DOFs) available for operation.

DEGREES OF FREEDOM FOR KAIBEL COLUMN

Assuming that the holdups (reboiler and condenser) and the pressure are controlled, the number of DOFs for Petlyuk arrangements with n_S sidestreams and n_D dividing walls are given by the following simple formula (Christiansen *et al.* 1997)

$$\text{DOFs} = 2 + n_S + 2n_D \tag{2}$$

For the Kaibel column we thus have 6 ($n_S = 2, n_D = 1$) potentially manipulated variables available for operation. However, for a sharp split we require that only small amounts of C should appear in the overhead from the prefractionator, and small amounts of B in the bottoms from the prefractionator. Thus we have already implicitly fixed two DOFs (e.g. liquid (R_L) and vapor splits (R_V)). If we want to achieve a certain purity for *all* 4 products, it thus seems as though we have enough DOFs left. However, for energy efficient operation of the Kaibel column, it is as previously noted strongly desirable to balance the column such that total reflux ($L/V = 1$) is achieved in the middle section between the sidestreams. Hence, we need to use one DOF to meet this requirement during operation. We are therefore in fact short of DOFs if we wish to control all purities and at the same time keep the column in the vicinity of the energy minimum. This lack of DOFs is due to the above mentioned direct coupling between V_4 and V_5. One way of compensating for this lack of DOFs is to allow for *overpurification*, i.e. allow for higher purities of one (or both) side product(s). The lack of DOFs also applies to the $\vdash$ column (which has only 5 DOFs), although in this case we at least avoid the additional operational problem of achieving $L/V = 1$ in the intermediate section. Furthermore, it may be easier to use additional heating or cooling in the $\vdash$ column to compensate for the loss of DOFs. In the next section we extend the analysis to Petlyuk arrangements with two dividing walls.

TWO DIVIDING WALLS

A column arrangement which allows for potentially reversible splits, is represented by the "Sargent column arrangement" in figure 4 (a). This arrangement is due to a superstructure consisting of three interconnected *regular* columns proposed by Sargent and Gaminibandara (1976). We emphasize that the two arrangements in figure 4 (a) are identical from a computational point of view if we neglect heat transfer across the dividing walls. For a more detailed discussion of this and other superstructures for Petlyuk arrangements we refer to another paper by the authors (Christiansen *et al.* 1997). The Petlyuk arrangement consists of $n(n-1) = 12$ sections, which in fact is the *maximum* number for sharp splits of a four component mixture. This is seen more clearly if we consider the network representation given in figure 4 (b). We recognize that since every possible node is present (see e.g. Agrawal (1996)), only the lightest and heaviest components are separated

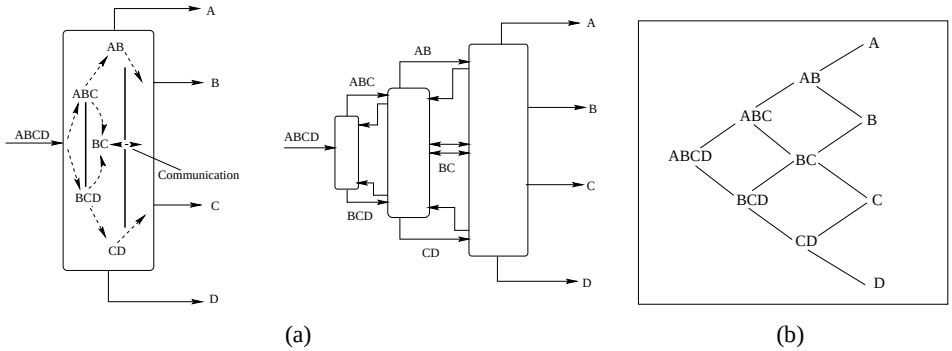


Figure 4: Petlyuk arrangement (a) and network representation (b) for Sargent's superstructure

from the mixture in each section. Hence, due to potentially reversible splits we should expect this design to display better performance in terms of thermodynamic efficiency, thus requiring a lower energy input compared to the Kaibel and the $\vdash$ columns.

By allowing for the possibility of having liquid and vapor transport (communication) between certain stages on both sides of a wall, there are in fact four streams which may be redistributed (liquid and vapor on each side). Thus, we add yet another four degrees of freedom for each such *communication point*. To avoid confusion in the proceeding discussion, we therefore make the somewhat fictive 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 4 (a). The total number of DOFs for a structure with n_C communication points is thus

$$\text{DOFs} = 2 + n_S + 2n_D + 4n_C \tag{3}$$

According to equation 3 we thus have 12 DOFs for the structure in figure 4 (a). However, from physical insight we conjecture that fluid should usually be transported only in the direction towards the final products. If we redistribute liquid and vapor *from* the sidestream side, the internal recycle increases, which for ideal mixtures most probably increases the required energy input. Taking the latter observations into account, 10 potentially manipulated variables remain.

Recently, Agrawal (1996) proposed a methodology from which to derive Petlyuk arrangements with two *satellite* columns in communication with a central distillation column. In the paper Agrawal introduced a superstructure for such satellite arrangements, which in fact is "more general" than the superstructure of Sargent and Gaminibandara (1976) in that the latter is contained as a substructure. We note that both superstructures consists of $n-1$ interlinked columns and require the *maximum* number of sections for a sharp split, i.e. $n(n-1)$. However, Agrawal also provides a procedure from which one may arrive at satellite arrangements having the *"minimum number" of sections*, which he shows to be equal to $4n-6$. However, as we have previously noted (Christiansen *et al.* 1997), this is in fact not the actual minimum for sharp splits in a Petlyuk arrangement, as illustrated by the Kaibel column (7 sections). The schematic in Figure 5 (a) illustrates a satellite arrangement and a corresponding dividing wall implementation similar to Agrawal's superstructure. If we examine the dividing wall arrangement in some more detail, we recognize that further scrutiny is required in order to visualize the feed location and the communication of the intermediate node denoted BC. We first note that the feed may in general enter the column from any of the three parts I, II and III of the dividing wall column in Figure 5 (a). This is perhaps more clearly understood if we consider the column viewed from the top as demonstrated to the left in Figure 5 (b). Furthermore, if there is to be any transfer of the BC node within the dividing wall column, we must enable communication between *any* of the three parts. This is indeed possible if we consider the *triangular* structure to the right in figure 5 (b). With this arrangement one may in principle allow for communication between *any* stages in all parts of the column. Before presenting the numerical results we give some brief comments on the internal distribution of flows.

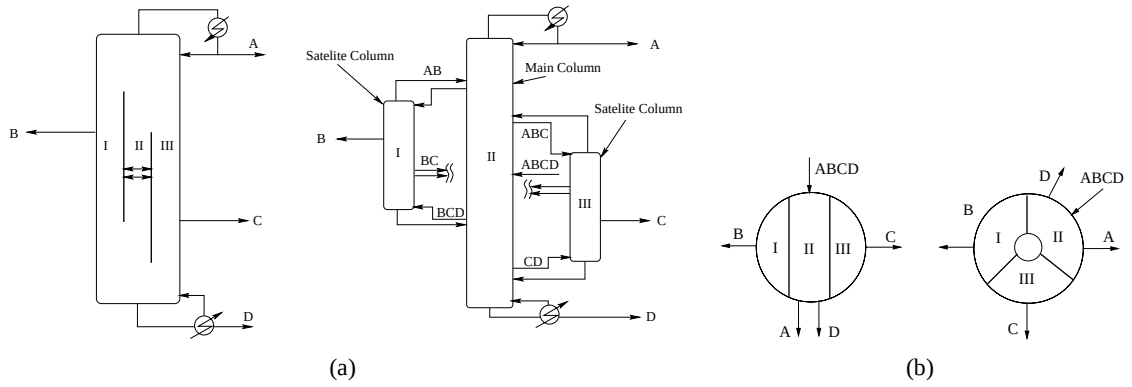


Figure 5: Petlyuk arrangement (a) and top view (b) of dividing wall implementation for Agrawal's superstructure

(i) Fluid transfer between communicating sections. For the Petlyuk arrangement in Figure 5, we note that B is withdrawn above the communication point whereas C is withdrawn below. Thus, arguing in terms of the physics of the separation, we conjecture that the optimum should be to "lift" B up towards the outlet and conversely "push" C downwards. We thus expect that the communication of fluid between the side–sections should take place so that vapor is transferred *primarily* in the direction *from* the sidesection where C is drawn off (part III) and *to* the section where B is withdrawn (part I). Conversely, one would expect that liquid should be transferred primarily in the direction from part I towards part III. However, we stress that the task of determining the optimal distribution liquid and vapor is still left for optimization. The same argument applies in the general case also for the Petlyuk arrangement in Figure 4. In the latter case this guideline suggests that fluid should (only) be transferred in the direction towards the product withdrawals.

(ii) Fluid distribution within sections. If we consider the sections from which the intermediates B and C are withdrawn, we argue that one should in general avoid operating regimes where there is a net flux of intermediates across the side outlets. The reason is that such a net flux inevitably leads to an internal recycle within the column, which most likely increases the energy requirements. Hence, above the side outlet there should be a larger fraction of intermediates in the liquid, and conversely a larger fraction in the vapor below the outlet. If we denote the liquid and vapor flows of intermediates above the side-outlet by L_i^a, V_i^a and below by L_i^b, V_i^b, we pose the following *heuristic guideline* for the distribution of internal flows within the sidestream sections

$$L_i^a > V_i^a \quad L_i^b < V_i^b \tag{4}$$

Condition (4) thus ensures that there is no net transport of intermediates across the outlets. We however stress that the optimum distribution of internal flows is still left for optimization.

NUMERICAL RESULTS

The numerical results presented here are obtained from optimizations of relatively simple, yet rigorous models assuming constant molar flows and constant relative volatility. The data for the mixtures 1 and 2, and the product specifications are given in table 1

Table 1: Process data for case studies

Feed mixture 1	$z_A = z_B = z_C = z_D = 0.25$
Feed mixture 2	$z_A = z_D = 0.4$, $z_B = z_C = 0.1$
Distillate purity	$x_A \geq 0.99$
Upper sidestream purity	$x_B \geq 0.95$
Lower sidestream purity	$x_C \geq 0.95$
Bottom purity	$x_D \geq 0.99$
Relative volatility	$8:4:2:1$

In order to compare the performance of Petlyuk arrangements with conventional designs it might be disputed how one should select the number of stages. For the optimizations given in this work we have chosen to compare the arrangements on basis of the *total* number of stages according to the model formulation, i.e. adding every stage in all sections. We denote this total number of stages by N_T, whereas we use the term N_H (where H denotes Height) for the number of stages in the *main column* counting from the reboiler (bottom) to the condenser (top). If we consider Agrawal's and Sargent's superstructures with 12 sections and 10 stages in each section, we thus have $N_T = 120$ and $N_H = 60$. In general, a Petlyuk arrangement generally requires more stages (N_T) for a finite reflux ratio. However, as noted also by Wolff and Skogestad (1995), using N_T may be conservative when comparing the Petlyuk arrangements with conventional arrangements. To justify using N_H instead of N_T, one could also argue that the number of stages in sections on either side of a partition comes *for free* with respect to column height and diameter. This follows if we take into account that the internal flows, which more or less determine the size of the column internals, are distributed between the partitioned sections within the single shell. However, for the results we indicate both N_H and N_T for the different arrangements.

1. Minimum energy input for arrangements with one dividing wall - finite number of stages.
To compare the performance for the $\vdash$ and Kaibel columns, we need to specify the number of stages in the middle section between the sidestream outlets in the Kaibel column. As discussed above, this section should ideally act as a total reflux column for which it requires a certain minimum number of stages to avoid remixing of intermediates B and C (the $\vdash$ and Kaibel columns are almost identical if total reflux $L/V = 1$ is achieved in the intermediate section with $N = N_{min}^{BC}$). With the given purity requirements, we may compute a "lower bound" for N_{min}^{BC} if we consider a separation in which only intermediates are present in the sidestreams. In this case equation (1) yields $N_{min}^{BC} = 8.5$. A more reasonable situation is to assume an even distribution of impurities as discussed above, hence equal amounts of A and C in the B–sidestream and conversely equal amounts of B and D in the C–sidestream. In the latter case we have a pseudo-binary mixture of B and C for which $x_{L,T} = x_{H,B} = 0.95$ and $x_{L,B} = x_{H,T} = 0.025$. Using this we obtain $N_{min}^{BC} = 10.5$ from equation (1). Hence, if we use 11 stages in the middle section, we should obtain a purity slightly higher than 95% for the side streams.

To address the impact of the number of stages on the minimum energy input, we considered arrangements with $N_T = 60$ and $N_T = 90$. However, we emphasize that when comparing the $\vdash$ and the Kaibel columns, the difference (in cost) owes to either an additional (external) heat exchanger or an intermediate section (dephlagmator) within the column shell. For the simulations we have thus chosen to *exclude* the "extra" stages in the intermediate section for the Kaibel column, corresponding to the required N_{min}. In order to compare the performance with conventional designs we considered a conventional "prefractionator" arrangement which closely resembles the $\vdash$ and the Kaibel arrangement as shown in Figure 6 (a). The main reason for choosing this particular design is that it yields the same values for both N_T and N_H. Results from

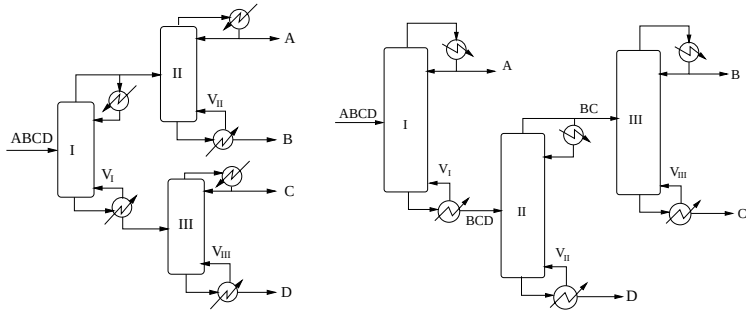


Figure 6: Conventional arrangements : (a) "Prefractionator" and (b) "Direct-indirect"

the optimizations are given in table 2, where ΔV is the fractional savings relative to the "prefractionator" arrangement.

Table 2: Minimum energy inputs for arrangements with one vertical partition

	"Prefractionator"	$\vdash$ and Kaibel[3]	Savings
$N_H = 40$, $N_T = 60$	$V_1 = 2.90$	$V_1 = 1.85$	$\Delta V_1 = 36\%$
	$V_2 = 2.39$	$V_2 = 1.40$	$\Delta V_2 = 41\%$
$N_H = 60$, $N_T = 90$	$V_1 = 2.31$	$V_1 = 1.55$	$\Delta V_1 = 33\%$
	$V_2 = 1.96$	$V_2 = 1.17$	$\Delta V_2 = 40\%$

From the results we find that the $\vdash$ and Kaibel columns offer considerable energy savings in the order of 40% compared to the conventional "prefractionator" arrangement. However, one may argue that it is reasonable to compare the $\vdash$ column with a conventional arrangement in which the reboiler for column II and the condenser for column III are heat integrated so that V_{II} should be neglected. Taking this into account we still find that the energy savings amount to 18%. We also note that among the other conventional arrangements (5 possible sequences for 4 components), results showed that the performance of the indirect split sequence (with vapor feed to subsequent columns) was similar to the "prefractionator" arrangement whereas the direct split required a larger V for mixtures 1 and 2.

2. Minimum energy input for arrangements with two dividing walls - finite number of stages.
A (conventional) column arrangement which resembles the Petlyuk arrangement in Figure 5, denoted the "direct–indirect" scheme, is given in Figure 6 (b). Referring to the discussion in the previous section, we mention that the main reason for choosing this particular arrangement for comparative purposes, is the fact that it may yield the same values for both N_T and N_H as the Petlyuk arrangements. In table 3 we give results in which we compare Agrawal's (satellite) and Sargent's (sequential) arrangements with the "Direct–indirect" scheme in Figure 6 (b). ΔV denotes the fractional savings of the Agrawal arrangement relative to the "direct–indirect" scheme.

Table 3: Minimum energy inputs for arrangements with two vertical partitions

	"Direct–indirect"	Sargent	Agrawal	Maximum savings
$N_H = 40$, $N_T = 80$	$V_1 = 2.45$	$V_1 = 2.27$	$V_1 = 2.11$	$\Delta V_1 = 14\%$
	$V_2 = 1.90$	$V_2 = 1.69$	$V_2 = 1.47$	$\Delta V_2 = 23\%$
$N_H = 60$, $N_T = 120$	$V_1 = 2.27$	$V_1 = 1.32$	$V_1 = 1.31$	$\Delta V_1 = 42\%$
	$V_2 = 1.75$	$V_2 = 1.07$	$V_2 = 1.06$	$\Delta V_2 = 39\%$

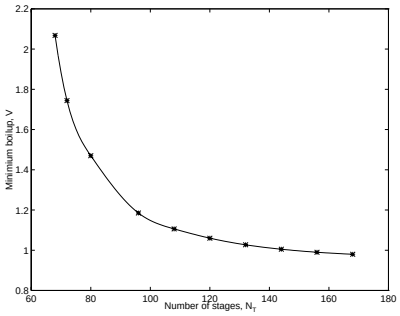
In order to compare the results for Petlyuk arrangements with one and two dividing walls given in tables 1 and 2, we stress that the Petlyuk arrangements are not strictly comparable in terms of N_T. This is firstly due to the $\vdash$ column not being a Petlyuk arrangement in a strict sense, since both direct and indirect thermal coupling takes place. Secondly, we previously pointed out that the Kaibel column in fact requires $N_T + N_{min}^{BC}$ and that the $\vdash$ requires either an extra heat exchanger or an internal region to provide the required heat transfer area. Secondly, there is a difference between the conventional schemes in Figure 6 used for comparisons, since we aimed for arrangements which may yield the same N_T *and* N_H. If we use the same N_H, we find that the the "direct-indirect" arrangement in Figure 6 (b) yields a larger N_T than the "prefractionator" arrangement in Figure 6) (a). Finally, we add that other *conventional* schemes such as the direct split scheme required a considerably larger energy input than the "direct-indirect" scheme, whereas the indirect split (vapor feed to subsequent columns) with the same N_T was slightly better, i.e. in the order of 1 % lower V. However, we again stress that the direct and indirect splits with the same N_T requires a structure with larger N_H.

Based on the results in table 2, in which we computed savings to be larger than $40\$$, we clearly see that the number of stages have a considerable impact on the fractional savings. We found that Agrawal's arrangement required the lowest boilup for $N_T = 80$, whereas Sargent's arrangement were almost equal as N_T was increased. The fact that the savings increase with N_T demonstrates that for $N_T = 80$ the minimum energy input for the conventional arrangement lies much closer to the "absolute" minimum than those of the Petlyuk arrangements. We therefore examine more closely the relation between the number of stages and minimum energy input.

[3]For the Kaibel column we must add N_{min}^{BC} stages in the intermediate section

3. Minimum energy input - infinite number of stages. Here we are interested in finding the minimum energy input required for a quaternary separation in a Petlyuk arrangement with two dividing walls. Since there are no analytical or shortcut methods available for complex columns with two dividing walls, we here resort to rigorous numerical computations. We pose the task of finding reliable shortcut methods for estimating the minimum energy input as an interesting and important problem for future work.

For conventional distillation columns we know that the relation between the number of stages N and the energy input V takes the shape of a hyperbolic function, for which there are two distinguished asymptotes. One gives N_{min} for the limiting case of infinite internal flows $(V/F = \infty)$, whereas minimum boilup V_{min} is obtained for $N = \infty$. The relation between N_T and V for the mixture in case 2 is given in figure 7. Note here that each point of the curve represents the minimized V for the given N_T. As demonstrated in the Figure we find that with a relative volatility of $\alpha_{ij} = 2$, the hyperbolic curve approaches $V^2_{min} \approx 0.98$ as $N > 80$. From Figure 7 (a) we find that V^2_{min} is in fact less than one[4]. This is surprisingly low, since



(a) $\alpha_{ij} = 2$

Figure 7: Trade–off between N_T and V for Agrawal's arrangement and feed mixture 2

even the binary separation between A and B requires a V_{min} of about 0.85. Thus, to illustrate the efficiency of the Petlyuk arrangement, we find that by adding approximately 10–15% to the minimum boilup for the *binary split*, it is possible to separate the quaternary mixture into its constituents even with a finite number of stages.

DISCUSSION

In this paper we have considered using Petlyuk arrangements for the separation of quaternary mixtures, and demonstrated how such arrangements may be implemented in a single shell using dividing walls. Numerical results for two feed mixtures indicated that the fractional savings compared to conventional schemes are more than 40%, provided there is a sufficient number of stages. However, we emphasize that the results are only tentative, in the sense that savings may be even larger (or smaller) for other mixtures. Our results also demonstrate that Petlyuk arrangements require a larger number of stages compared to conventional designs to achieve the minimum boilup. However, we also emphasize that it is arguable how one should count the total number of stages in a Petlyuk arrangement.

Since Petlyuk arrangements give a large number of DOFs at steady state and commonly display highly nonlinear behavior, the task of computing the optimal solution is indeed a very difficult one. In fact, even finding a steady state solution requires robust and reliable numerical algorithms. In this work we used numerical integration of a *dynamic* model to obtain an initial steady state solution. The optimized solutions was then computed from a *steady state* model by continuation in the subspace spanned by the DOFs. Furthermore, due to the size and complexity of the problems (typically $3N_T$ number of equations and up to 12 DOFs), there is no way of guaranteeing a global optimum. However, based on a comprehensive analysis of the numerical results, in which we studied characteristics such as the composition profiles, the internal

[4]If the volatility is increased to $\alpha_{ij} = 3$, $V^2_{min} \approx 0.73$ is approached as $N > 60$.

distribution of components, the presence internal recycles etc., it seems likely that the results are at least close to the global optimum.

Furthermore, we stress that the Petlyuk arrangements have only been optimized with respect to the degrees of freedom available for *operation*. For a column with a given structure (i.e number of sections and total number of stages), we stress that additional savings are possible if one also takes into account discrete *design* parameters such as the distribution of stages between the different sections and the feed– and side–stream locations. Still, the main contribution from our work is to demonstrate that there is a large potential for extending the Petlyuk ideas to separations of four or more components. Future work is needed to investigate for what mixtures Petlyuk arrangements yield the largest potential savings.

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