Minimum Energy Consumption in Multicomponent Distillation. 2. Three-Product Petlyuk Arrangements

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We show that the minimum energy requirement for separation of a multicomponent mixture in a three-product Petlyuk arrangement is equal to the minimum energy for the most difficult of the two separations (top/middle or middle/bottom product) in a conventional single column. In the $V_{\min}$ diagram (part 1 of this series), this is simply the highest peak. These results are based on an analytical solution for columns with an infinite number of stages, assuming constant relative volatilities and constant molar flows. The previous analytical results for the Petlyuk column are extended to include nonsharp separations, multicomponent feeds, and any feed quality.

1. Introduction

In this paper, the minimum-energy expressions for the three-product Petlyuk arrangement[1] shown in Figure 1 are generalized to handle any feed quality and nonsharp product splits. We also illustrate by examples that we can easily handle more than three feed components. We use the simplifying assumptions of constant pressure, constant relative volatility (α), and constant molar flow and consider the limiting case with an infinite number of stages.

The ternary feed (F) with components A (light), B (intermediate), and C (heavy) is supplied to the prefractionator (column C1), which performs the "easy" A/C split. The minimum vapor flow in the prefractionator column is obtained for a particular distribution of the intermediate B component, denoted as the *preferred split*.[2] This split also results in a minimum overall energy requirement in the Petlyuk column. Interestingly, this solution is not unique, and several authors, e.g., Fidkowski and Krolikowski[3] and Christiansen and Skogestad,[4] have shown that the optimum can be obtained by operating the prefractionator in the whole region between the preferred split and the so-called "balanced" split where the vapor flow requirements in the bottom of column C21 and in the top of column C22 are equal. This implies that there is a "flat" optimality region and that the minimum vapor flow can be obtained not only at a single operating point but also along a line segment in the space spanned by the 2 degrees of freedom.

An analytical expression for the minimum vapor flow in a Petlyuk arrangement with a ternary feed and liquid side stream was obtained independently by Fidkowski and Krolikowski[3] and Glinos et al.[5] for the case of a saturated liquid feed ($q = 1$) and sharp product splits:

$$V_{\min}^{\text{Petlyuk}} = \max\left(\frac{\alpha_A z_A}{\alpha_A - \theta_A}, \frac{\alpha_A z_A}{\alpha_A - \theta_B} + \frac{\alpha_B z_B}{\alpha_B - \theta_B}\right)F \quad (1)$$

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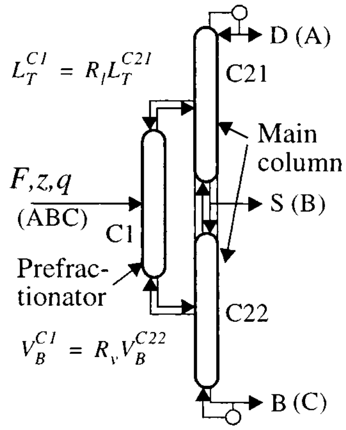


Figure 1. Integrated Petlyuk arrangement for separation of ternary mixtures.

Here, θ_A and θ_B are the two common Underwood roots, obtained from (3) for the prefractionator feed.

Fidkowski and Krolikowski[3] derived (1) by a quite detailed algebraic procedure, via expressions for pinch zone compositions at the connection points as functions of the operating point of the prefractionator. Here we will use another approach, more directly based on the Underwood equations. Such an approach was first presented by Carlberg and Westerberg,[6,7] who also extended the solution to more than one intermediate component.

An important finding in our work is that the minimum energy requirement ($V_{\min}^{\text{Petlyuk}}$) and the detailed vapor flow requirements may be obtained by just a glance at the $V_{\min}$ diagram for a single two-product column. This was presented in part 1 of the series[8,9] and is computed based on Underwood's equations[10−13] for multicomponent distillation in conventional columns. The most important results from part 1 are reviewed in section 2.

In the directly coupled sections of the Petlyuk arrangement, we have recycle flows from the main column into the top and bottom of the prefractionator. This is a new situation compared to the conventional arrangements, and we must really check if Underwood's meth-

10.1021/ie0108649 CCC: $25.00
Published on Web 01/01/2003

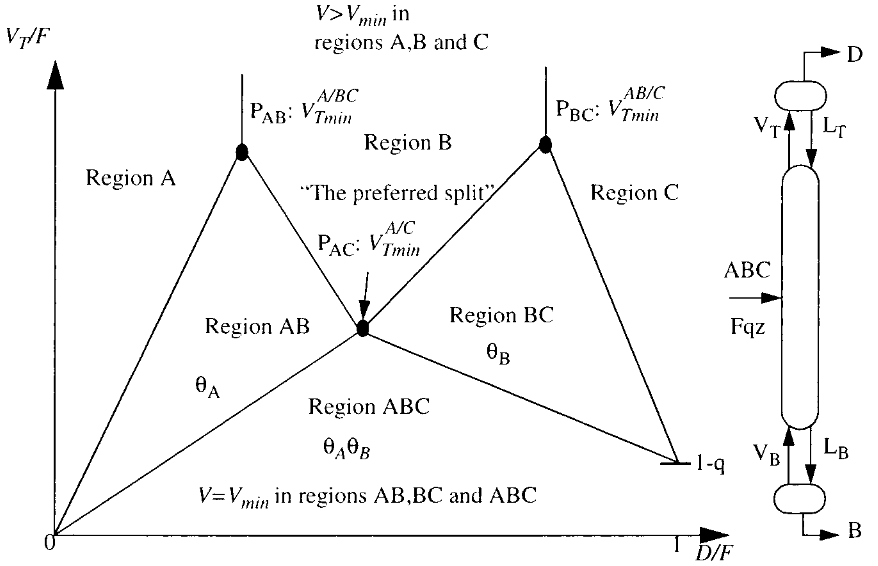


Figure 2. V_{min} diagram for a ternary mixture ABC. The components which are distributed to both ends are indicated in each region with the corresponding active Underwood roots.

ods can be applied. This issue is treated in the thesis by Halvorsen,[8] and it turns out that, with some restrictions on the recycle stream compositions, the directly coupled columns can be treated as ordinary columns. In section 3.2 we present the important result from Carlberg and Westerberg[6,7] on how the Underwood roots carry over to the succeeding directly coupled column. This is the basis for the very simple assessment we can do with a V_{min} diagram.

The main results for a ternary mixture are presented in section 4, and the results are generalized to more than three feed components and nonsharp product splits in section 5.

In section 6, we briefly discuss the results in relation to some other types of column integration.

2. Review of the Basic Equations for Minimum Energy

2.1. Underwood Equations. Consider a two-product distillation column with a multicomponent feed (F) with liquid fraction q and composition vector z of N components. The defining equations for the Underwood roots (ϕ) in the top and (ψ) in the bottom are

$$\text{Top:}\quad V_T = \sum_{i=1}^{N} \frac{\alpha_i w_{i,T}}{\alpha_i - \phi}$$

$$\text{Bottom:}\quad V_B = \sum_{i=1}^{N} \frac{\alpha_i w_{i,B}}{\alpha_i - \psi} \tag{2}$$

where w_i is the net flow of a component (defined positive upward, also in the bottom). There will be N solutions for each root, and the solution sets from the top and bottom equations are generally different. However, Underwood[10-13] showed that the roots obey LF $\alpha_i \geq \phi_i \geq \psi_{i+1} \geq \alpha_{i+1}$. Furthermore, with an infinite number of stages, at minimum vapor flow, one or more pairs of roots (ϕ_i, ψ_{i+1}) in the top and bottom coincide to a set of common roots (θ_i). The set of $N - 1$ possible common roots are obtained by setting $\phi_i = \psi_{i+1} = \theta_i$ and subtracting the two defining equations above. This gives the feed equation where the set of common roots depends only on the feed properties α, z, and q:

$$V_T - V_B = \sum_{i=1}^{N} \frac{\alpha_i (w_{i,T} - w_{i,B})}{\alpha_i - \theta} = \sum_{i=1}^{N} \frac{\alpha_i z_i F}{\alpha_i - \theta} = (1 - q)F \tag{3}$$

However, it is not obvious when we may apply the common roots (θ) solved from (3) back into the defining equations (2), in particular for more than two components. The rule is that we may apply the common roots in the range of volatilities for the components distributed to both ends (including components exactly at the limit of being distributed). We denote these *active roots*. When we have any active roots then, $V = V_{min}$. The minimum vapor flow and component distribution can then be found by solving the equation set obtained by applying all of the *active* roots in (2). A total of 2 degrees of freedom (e.g., two key component recoveries) must be specified. If there are no active roots, $V > V_{min}$.

2.2. V_{min} Diagram for a Single Conventional Column. We here review the results from part 1 of this series.[8,9] Because a two-product column operated at constant pressure has only 2 degrees of freedom, we may visualize all possible operating points in the $D-V$ plane. This is illustrated in the V_{min} diagram, which is shown for a ternary feed (with components ABC) in Figure 2.

Each peak or knot in this diagram (P_{ij}) is the operating point for minimum vapor flow and sharp split between the component pair ij ($V_{min}^{i/j}$). The straight lines between the peaks and knots are distribution boundaries. At a boundary, a component is at the limit of appearing or disappearing in one of the product streams. We denote the distribution regions by the components being distributed to both products when operating in that region. For example, in region AB components A and B are distributing to both products, whereas component C only appears in the bottom product. In region ABC all three components distribute to both products. At point P_{AC}, the preferred split,[2] only the intermediate component B distributes. The light A appears only in the top and the heavy C only in the bottom, and both A and C are exactly at the limit of

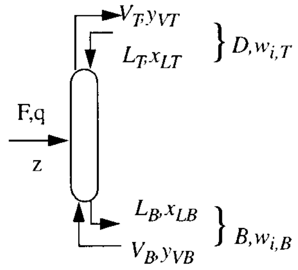


Figure 3. Prefractionator of a Petlyuk arrangement.

being distributed. In regions A, B, and C we have nonoptimal operation with $V > V_{\min}$. All of the possible minimum-energy solutions ($V = V_{\min}$) are found below the "mountain", in regions AB, ABC, or BC. There is a unique minimum-energy solution for each feasible pair of a key component specification in the top and in the bottom. Note that the active common Underwood roots are those in the range between the volatilities of the distributing components and that no roots are active in the nonoptimal regions above the "mountain".

In the following we show how to use the $V_{\min}$ diagram for directly coupled columns such as the Petlyuk arrangement.

3. Underwood Equations Applied to Directly Coupled Sections

3.1. Petlyuk Column Prefractionator. In the prefractionator of a Petlyuk column, we can still use the net component flow (w) to describe the separation carried out in the column. From the material balance at any cross section in the column:

$$w_{i,n} = V_n y_{i,n} - L_{n+1} x_{i+1,n} \tag{4}$$

Thus, for the column in Figure 3 the composition in the flow leaving the column top is dependent on the composition of the incoming flow through the material balance:

$$y_{i,\mathrm{VT}} = \frac{w_{i,\mathrm{T}} + L_\mathrm{T} x_{i,\mathrm{LT}}}{V_\mathrm{T}} \tag{5}$$

For a conventional column with total condenser, we have $x_{i,\mathrm{LT}} = y_{i,\mathrm{VT}}$ and $y_{i,\mathrm{LT}} = w_{i,T}/D$, where $D = V_\mathrm{T} - L_\mathrm{T}$, but this does not apply here. However, even if there are external streams entering into the top and bottom, the compositions in these streams normally do not affect the distribution of the feed components (w_i) to the top and bottom in the column, and the following rule[8] can be used:

The V_{min} diagram for a conventional column can also be applied to the Petlyuk prefractionator, provided that a component, that would have been removed from one end in the conventional column, does not appear in the end "feeds" of the Petlyuk prefractionator.

3.2. "Carryover" of Underwood Roots in Directly Coupled Columns. The first part of this section is mainly based on work by Carlberg and Westerberg,[6,7] who pointed out that Underwood roots "carry over" from the top of the first columns to the second column in the directly or fully thermally coupled columns as shown in Figure 4.

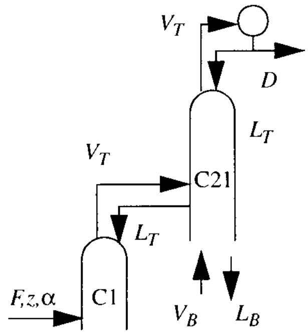


Figure 4. Directly coupled columns (fully thermally coupled).

The vapor flow in the top of the prefractionator is given by the Underwood defining equation:

$$V_\mathrm{T}^\mathrm{C1} = \sum_i \frac{\alpha_i w_{i,\mathrm{T}}^\mathrm{C1}}{\alpha_i - \phi^\mathrm{C1}} \tag{6}$$

Note that we generally have to apply the actual Underwood roots (ϕ). The common roots (θ) only apply for minimum-energy operation.

The top and bottom defining equations (2) for column C21 become

$$V_\mathrm{T}^\mathrm{C21} = \sum_i \frac{\alpha_i w_{i,\mathrm{T}}^\mathrm{C21}}{\alpha_i - \phi^\mathrm{C21}} \quad \text{and} \quad V_\mathrm{B}^\mathrm{C21} = \sum_i \frac{\alpha_i w_{i,\mathrm{B}}^\mathrm{C21}}{\alpha_i - \psi^\mathrm{C21}} \tag{7}$$

The material balance at the connection point gives

$$V_\mathrm{T}^\mathrm{C21} - V_\mathrm{B}^\mathrm{C21} = V_\mathrm{T}^\mathrm{C1} \quad \text{and} \quad w_{i,\mathrm{T}}^\mathrm{C21} - w_{i,\mathrm{B}}^\mathrm{C21} = w_{i,\mathrm{T}}^\mathrm{C1} \tag{8}$$

The combination of these gives the feed equation (3) for column C21 where the common roots (θ^C21) appear

$$V_\mathrm{T}^\mathrm{C21} - V_\mathrm{B}^\mathrm{C21} = \sum_i \frac{\alpha_i (w_{i,\mathrm{T}}^\mathrm{C21} - w_{i,\mathrm{B}}^\mathrm{C21})}{\alpha_i - \theta^\mathrm{C21}} = \sum_i \frac{\alpha_i w_{i,\mathrm{T}}^\mathrm{C1}}{\alpha_i - \theta^\mathrm{C21}} = V_\mathrm{T}^\mathrm{C1} \tag{9}$$

Here we observe that the feed equation of column C21 (9) is identical to the top section defining equation for column C1 in (6). Thus, the possible common roots in column C21 are equal to the actual roots from the defining equation in the top of column C1:[6,7]

$$\theta^\mathrm{C21} = \phi^\mathrm{C1} \tag{10}$$

Assume that we recover all of the light A in the top of column C21. Then $w_{\mathrm{A,T}}^\mathrm{C21} = z_\mathrm{A} F$ and $w_{\mathrm{B,T}}^\mathrm{C21} = w_{\mathrm{C,T}}^\mathrm{C21} = 0$. The minimum vapor flow in column C21 for any given operation of C1 is when the common root ($\theta_\mathrm{A}^\mathrm{C21}$) is active, which implies $\phi_\mathrm{A}^\mathrm{C21} = \theta_\mathrm{A}^\mathrm{C21} = \phi_\mathrm{A}^\mathrm{C1}$, and from (2), we have

$$\frac{V_\mathrm{T,min}^\mathrm{C21}}{F} = \frac{\alpha_\mathrm{A} z_\mathrm{A}}{\alpha_\mathrm{A} - \phi_\mathrm{A}^\mathrm{C1}} \tag{11}$$

where $F = F^\mathrm{C1}$ and z is the feed composition to column C1. Because $\theta_\mathrm{A}^\mathrm{C1} \le \phi_\mathrm{A}^\mathrm{C1}$, the absolute minimum solution is found when $\phi_\mathrm{A}^\mathrm{C1} = \theta_\mathrm{A}^\mathrm{C1}$. Then the common root of C1 becomes active in both columns C1 and C21 at the same time ($\phi_A^{C21} = \theta_A^{C21} = \phi_A^{C1} = \theta_A^{C1}$) and

$$\min_{C1}\left(\frac{V_{T,min}^{C21}}{F}\right) = \frac{\alpha_A z_A}{\alpha_A - \theta_A^{C1}} \quad (12)$$

As usual the notation "V_{min}" represents the minimum vapor flow for a single column for a given feed. The outer "min( )" represents the effect of the operation of column C1 to the feed composition and the effective feed quality for column C21. The common roots (θ^{C1}) are given by the feed equation (3) for the main feed to column C1.

We may generalize this expression to any number of components and feasible recoveries of components from the main feed in the top of column C21 with the following equation set (one equation for each active Underwood root in column C1 $\theta_k \in [\theta_1, ..., \theta_{N_{dT}^{C21}-1}]$ given by the N_{dT}^{C21} components distributed to the top of C21):

$$\min_{C1}\left(\frac{V_{B,min}^{C22}}{F}\right) = \frac{-\alpha_C z_C}{\alpha_C - \theta_B^{C1}} = \frac{\alpha_A z_A}{\alpha_A - \theta_B^{C1}} + \frac{\alpha_B z_B}{\alpha_B - \theta_B^{C1}} - (1-q) \quad (13)$$

For column C22 connected to the bottom of column C1, we have equivalent results. For the ternary feed case, with full recovery of the heavy component C in the bottom of column C22 and zero recovery of the middle and light components, the equivalent to eq 12 is

$$\min_{C1}\left(\frac{V_{B,min}^{C22}}{F}\right) = \frac{-\alpha_C z_C}{\alpha_C - \theta_B^{C1}} = \frac{\alpha_A z_A}{\alpha_A - \theta_B^{C1}} + \frac{\alpha_B z_B}{\alpha_B - \theta_B^{C1}} - (1-q) \quad (14)$$

Note that we have not considered the actual compositions in the junction streams. However, we know from section 3.1[8] that the composition in the return flow into the top of C1 has no influence on the product split in C1 unless a component which would have been removed in a conventional prefractionator was to be introduced in that return flow. This implies that, for nonsharp operation of C1 (where all components distribute and all common roots are active), the return-flow composition has no influence at all. For preferred split operation, this is also true when we ensure that there is no heavy (C) component in the return flow from C21 to C1.

In normal operation regimes of C1 and C21, the conditions are trivially fulfilled.

4. Minimum Energy for Separating a Ternary Feed in a Petlyuk Arrangement

We here consider the separation of a ternary feed mixture (components A, B, and C) in the three-product Petlyuk arrangement in Figure 1. In the following, all Underwood roots (θ, ϕ, and ψ) without superscripts are related to column C1 or an equivalent two-product column with the same feed.

4.1. Coupling Column C22 with Columns C21 and C1. For a sharp A/C split in column C1 and a sharp A/B split in column C21, the minimum vapor flow requirement in the top of C21 is given by (11):

$$V_T^{C1} \geq V_{T,min}^{C21} = \frac{\alpha_A z_A}{\alpha_A - \phi_A}F \quad (15)$$

We can also find the equivalent for the bottom flow in C22 for a sharp B/C split from (14):

$$V_B^{C22} \geq V_{B,min}^{C22} = \frac{-\alpha_C z_C}{\alpha_C - \psi_C}F \quad (16)$$

Because of the direct coupling, we know that the absolute minimum vapor flow in C21 is found when we operate column C1 in a region where $\phi_A = \theta_A$. Similarly, the absolute minimum for vapor flow in C22 is found when C1 is operated in a region where $\psi_C = \theta_B$. For sharp product splits, the preferred split is the only point of operation where *both* common roots carry over to C21 and C22 at the same time. (Any other solution will give a larger value for the minimum vapor flow in at least one of C21 or C22.)

The Petlyuk arrangement has a single reboiler, and the flow there must exceed the demands from both columns C21 and C22. Thus, we have

$$V_{B,min}^{Petlyuk} = \max\left[\min_{C1}(V_{T,min}^{C21}) - (1-q)F, \min_{C1}(V_{B,min}^{C22})\right] \quad (17)$$

For sharp product splits, we can express this as

$$V_{B,min}^{Petlyuk} = \max\left(\frac{\alpha_A z_A}{\alpha_A - \theta_A} - (1-q), \frac{-\alpha_C z_C}{\alpha_C - \theta_B}\right)F \quad (18)$$

or equivalently for the top of the Petlyuk arrangement

$$V_{T,min}^{Petlyuk} = V_{B,min}^{Petlyuk} + (1-q)F = \max\left(\frac{\alpha_A z_A}{\alpha_A - \theta_A}, \frac{\alpha_A z_A}{\alpha_A - \theta_B} + \frac{\alpha_B z_B}{\alpha_B - \theta_B}\right)F \quad (19)$$

This expression (19) is identical to (1) of Fidkowski and Krolikowski,[3] but (19) is more general in that it is also valid for an arbitrary feed quality (q). Note from (3) that q affects the solution for the common Underwood roots (θ_A, θ_B) and not only the term $(1-q)F$.

At this minimum solution either C21 or C22 may get a vapor flow larger than its minimum. However, this only affects the local behavior of that column and not the product composition and the operation of the prefractionator and the other column (refer to section 3.1).

4.2. Visualization in the V_{min} Diagram. By a closer inspection of the vapor flow rates for the Petlyuk arrangement, we observe that all of the important information can be found in the V_{min} diagram for the feed to the prefractionator (C1). Figure 5 illustrates this for a ternary example. The expressions for the peaks P_{AC} and P_{BC} (and also the preferred split P_{AC}) for a ternary feed are given by Underwood's equations (part 1 in this series[8]). In P_{AB} we recover all of the light A component ($w_{A,T} = z_A F$) and θ_A is active. In P_{BC} we recover all of both A and B and θ_B is active; thus, we

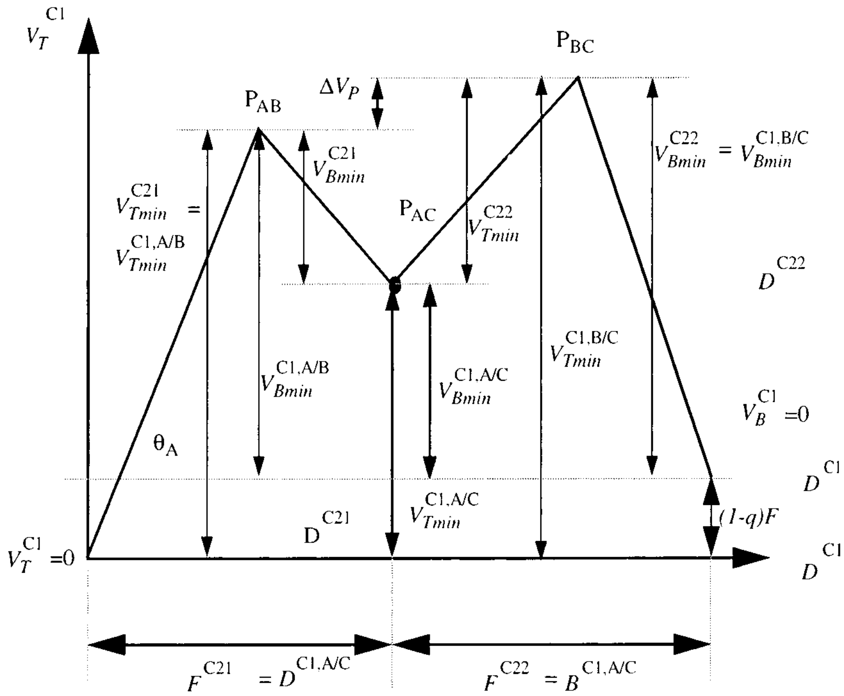


Figure 5. Use of the V_{min} diagram for assessment of a Petlyuk arrangement.

get from (2) and (3)

$$P_{AB}:\quad \frac{V_{T,min}^{C1,A/B}}{F} = \frac{\alpha_A z_A}{\alpha_A - \theta_A} \tag{20}$$

$$P_{BC}:\quad \frac{V_{T,min}^{C1,B/C}}{F} = \frac{\alpha_A z_A}{\alpha_A - \theta_B} + \frac{\alpha_B z_B}{\alpha_B - \theta_B} \tag{21}$$

These are exactly the same terms as the expression for $V_{T,min}^{Petlyuk}$ in (19) (the notation *i*/*j* in the superscript denotes a sharp *i*/*j* split in a two-product column), that is

$$V_{T,min}^{Petlyuk} = \max\,(V_{T,min}^{C1,A/B}, V_{T,min}^{C1,B/C}) \tag{22}$$

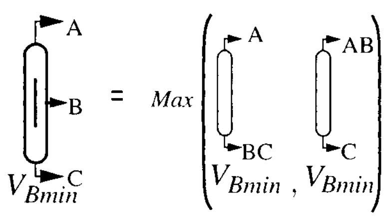
Similarly we find for the vapor flow requirement into the bottom of the Petlyuk column:

$$V_{B,min}^{Petlyuk} = \max\,(V_{B,min}^{C1,A/B}, V_{B,min}^{C1,B/C}) \tag{23}$$

This leads to the following important conclusion for pure product specifications:

The minimum vapor flow rate requirement in the Petlyuk column with three pure products is the same as the minimum vapor flow for the most difficult of the two sharp component splits A/B or B/C in a single conventional distillation column.

This is illustrated in the following equation, where we use the column drawings as superscripts (the Petlyuk column is shown as a dividing wall column):



(24)

In the V_{min} diagram this conclusion is the same as the following:

The minimum energy of a Petlyuk arrangement is characterized as the highest peak in the V_{min} diagram.

Thus, for the case shown in Figure 5, we observe by a glance at the diagram that P_{BC} is the highest peak and thereby $V_{B,min}^{Petlyuk} = V_{B,min}^{B/C}$.

We may also read the required minimum vapor flows in all sections of the Petlyuk arrangement directly from the V_{min} diagram for the prefractionator feed as shown in Figure 5. The relations are trivial to derive from the material balance at the junctions.

4.3. Flat Optimality Region. When we consider the preferred split operation, we have, in general, three different solution cases, characterized by the requirement for minimum vapor flow from columns C21 and C22 in the main column:

1. C22 controls: $V_{B,min}^{C22} > V_{T,min}^{C21} - (1 - q)F$ or $V_{B,min}^{C1,A/B} < V_{Bmin}^{C1,B/C}$

2. Balanced: $V_{B,min}^{C22} = V_{T,min}^{C21} - (1 - q)F$ or $V_{B,min}^{C1,A/B} = V_{Bmin}^{C1,B/C}$

3. C21 controls: $V_{B,minn}^{C22} < V_{T,min}^{C21} - (1 - q)F$ or $V_{B,min}^{C1,A/B} > V_{B,min}^{C1,B/C}$

In cases 1 and 3, there are different vapor flow requirements in the bottom of C21 and the top of C22. The difference is given directly as the difference between the height of the peaks in Figure 5. For a balanced main column (case 2), the peaks are equal. The highest peak always sets the overall requirement.

When we implement the vapor flow in the reboiler, we simply use

$$V_B^{C22} = V_{B,min}^{Petlyuk} \tag{25}$$

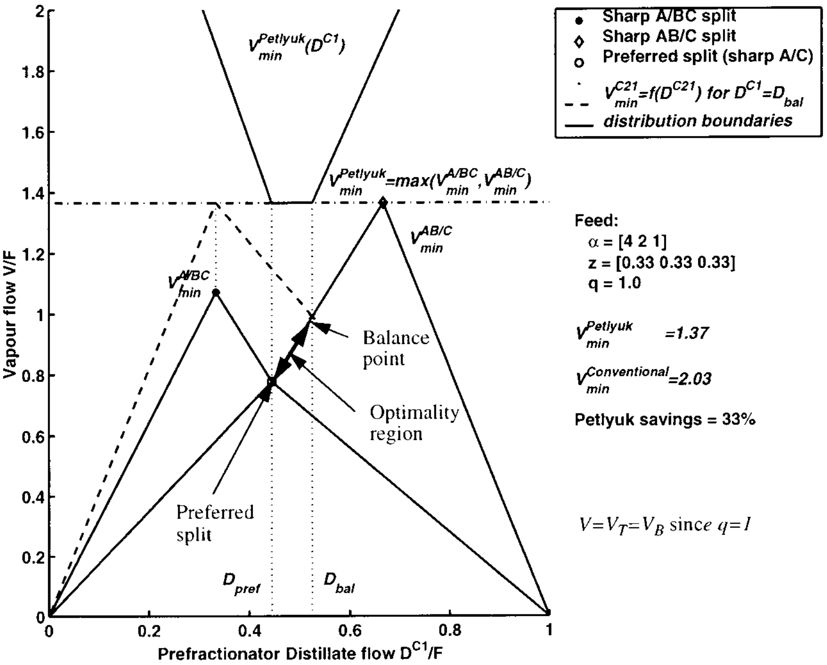


Figure 6. $V_{\min}$ for the prefractionator (C1) and the overall Petlyuk column (with a flat optimality region) as a function of D/F for the prefractionator.

and in the top we have

$$V_T^{C21} = V_{B,\min}^{\text{Petlyuk}} + (1 - q)F \tag{26}$$

Let us now assume that we have the situation in case 1. It is obvious that, because $V_T^{C21} > \min(V_{T,\min}^{C21})$, the root θ_A cannot be active in C21. The amount of distillate product is the total amount of A from the feed, and we have the following defining equation with this specification, from which we can solve for the root.

$$V_T^{C21} = \frac{\alpha_A z_A F}{\alpha_A - \phi_{A,\text{bal}}^{C21}} = V_{B,\min}^{\text{Petlyuk}} + (1 - q)F \tag{27}$$

We have two limiting cases. The first is when we operate the prefractionator at the preferred split. Then θ_A is active in C1, and because it will carry over to the feed equation in C21, we clearly waste vapor flow in C21. The other limiting case is when we move the operation point of C1 along the boundary BC/B until $\phi_A^{C1} = \phi_{A,\text{bal}}^{C21}$. In this case the vapor flow in C21 is a local $V_{\min}$ solution in C21; thus, $V_T^{C22} = V_{T,\min}^{C21} > \min(V_{T,\min}^{C21})$. Now the main column is balanced

$$V_T^{C22} = V_{T,\min}^{C22}(D^{C1},V^{C1}) = V_{B,\min}^{C21}(D^{C1},V^{C1}) = V_B^{C21} \tag{28}$$

Outside this flat optimality region, the overall vapor flow requirement increases rapidly. Figure 6 gives an example where we have plotted the balance point and also shows how the overall minimum vapor flow for the Petlyuk column depends on the prefractionator net product flow (D).

In this example, we may find the real root ($\phi_{A,\text{bal}}$) in the top of C1 (which carries over to C21), related to the balance point from

$$\frac{V_{T,\min}^{\text{Petlyuk}}}{F} = \frac{\alpha_A z_A}{\alpha_A - \theta_B} + \frac{\alpha_B z_B}{\alpha_B - \theta_B} = \frac{\alpha_A z_A}{\alpha_A - \phi_{A,\text{bal}}} \tag{29}$$

Knowing ϕ_A and $\phi_B = \theta_B$ in the balance point, we find the actual D and V for the prefractionator directly from the defining equations for the Underwood roots. The $V_{\min}$ diagram for C21 when $\phi_A = \phi_{A,\text{bal}}$ is shown dashed in Figure 6.

If the peak P_{AB} were the highest, we would have a case 3 situation, with the optimality region to the left of the preferred split. We may summarize as follows:

The flat optimality region is found from the preferred split and on the V-shaped minimum-energy boundary for sharp A/C split toward the highest peak. The extent of the optimality region depends on the difference of the height of the peaks or, in other words, the difference in how difficult it is to separate A/B or B/C in a single column.

4.4. Nonsharp Product Specifications. For nonsharp specifications the minimum vapor flow in the top section of column C21 is given from (13) when the net component flows in the top product is known. In the ternary case where both A and B may appear in the top and both B and C in the bottom, the generalization of (15) and (16) becomes

$$V_{T,\min}^{C21} = \frac{\alpha_A w_{A,T}^{C21}}{\alpha_A - \theta_A} + \frac{\alpha_B w_{B,T}^{C21}}{\alpha_B - \theta_A} \tag{30}$$

$$V_{B,\min}^{C22} = \frac{\alpha_B w_{B,B}^{C22}}{\alpha_B - \theta_B} + \frac{\alpha_C w_{C,B}^{C22}}{\alpha_C - \theta_B} \tag{31}$$

where the net flows are obtained from the product specifications. These results represent the minimum energy in a single column for the splits of the top/middle and middle/bottom products, respectively, just as in the sharp split case. The overall requirement is determined by the one giving the highest reboiler requirement according to (17).

Nonsharp product specifications for the ternary feed case have been treated in full detail in Chapter 9 of Halvorsen,[8] where we show that the nonsharp side-

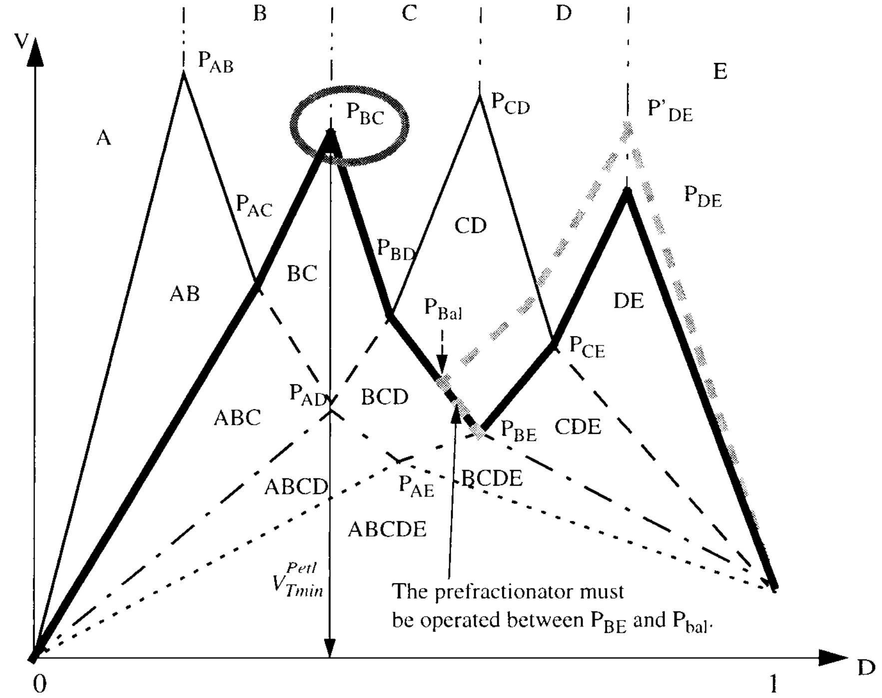


Figure 7. V_{min} diagram for a five-component feed used to find minimum vapor flow requirements in a three-product Petlyuk arrangement for sharp product splits AB/CD/E.

stream impurity specification actually extends the optimality region from a line segment to a quadrangle in the plane spanned by the 2 selected degrees of freedom.

5. Multicomponent Feed

We here extend the results from the previous section to more than three feed components. The minimum energy is still given by the largest minimum energy requirement from either column C21 or C22 as in (17).

First note that the Underwood roots carry over from the prefractionator to columns C21 and C22 in the same way for any number of components in the feed. This implies that if we operate the prefractionator at its preferred split, all of the common Underwood roots carry over. The general expression in (13) covers both multicomponent feed and nonsharp separations. However, this implies that the V_{min} diagrams for columns C21 and C22 will overlap the diagram for column C1 also in the multicomponent case. Note that the V_{min} diagram is based solely on the properties of the feed to column C1 and characterize distribution regions in an ordinary two-product column. The fact that we can use the same diagram for the whole Petlyuk arrangement is very important and gives us a powerful and simple tool for assessment of any given separation task in a Petlyuk arrangement.

5.1. General Rule. We extend the rule given in section 4.2 for a ternary feed and sharp component splits to a general multicomponent feed and three composite and possible nonsharp product specifications:

The minimum vapor flow requirement in the Petlyuk column with three products is the same as the minimum vapor flow for the most difficult of the two possible product splits (top/middle or middle/bottom products) in a single conventional distillation column.

This is characterized as the highest peak in the resulting V_{min} diagram for the products.

We simply replaced the term "component" from section 4.2 with "product".

To obtain analytical solutions for minimum vapor flow and product splits, we apply the computational tools based on the Underwood equations presented in part 1 of this series.[8,9]

Two examples, with $N = 5$ components (ABCDE) in the feed, are now used to illustrate how simple it is to use the V_{min} diagram to find the minimum-energy solution and flow requirements in the Petlyuk arrangement. We do not give any particular feed properties; thus, the diagrams should be interpreted qualitatively.

5.2. Example: Sharp Separations. First consider a case where we want AB in the top product, CD in the side stream, and pure E in the bottom. A V_{min} diagram is shown in Figure 7.

In the prefractionator we have to remove AB from the bottom and E from the top. This is obtained along the "V"- shaped boundary $P_{BC}-P_{BD}-P_{BE}-P_{CE}-P_{DE}$ (solid bold). The "preferred" solution for the prefractionator is to operate at P_{BE}. In column C21 it is known that the diagram for C21 overlaps the diagram for C1 to the left of the preferred split when column C1 is operated at the preferred split. Column C21 shall perform a sharp AB/C separation and the minimum-energy solution then simply found at P_{BC}. Similarly, in column C22 the peak P_{DE} gives the corresponding minimum vapor flow for a sharp split between CD/E. Thus, the Petlyuk arrangement requirement is simply given by the highest peak P_{BC} or P_{DE}, which is the encircled P_{BC} in the figure.

In this case we will also have a flat optimality region. It is shown qualitatively that if we move the operation of column C1 to the left of the preferred split, along the boundary BCD/CD, the peak P_{DE} will start to increase. At P'_{DE}, it becomes equal to P_{BC} and the main column is balanced, and the prefractionator (C1) is operated at P_{bal}. Thus, minimum vapor flow for the Petlyuk column

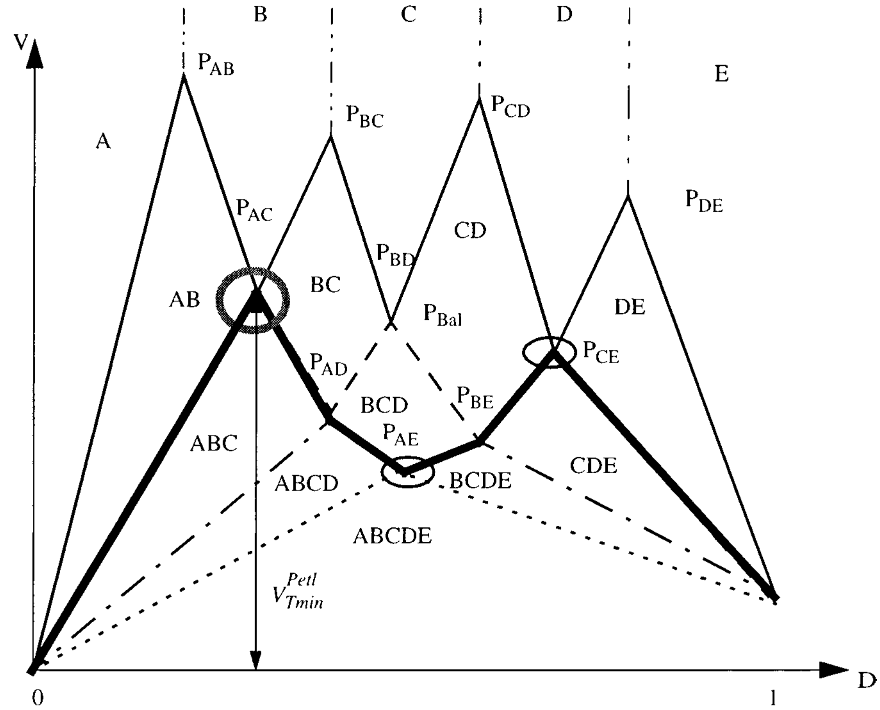
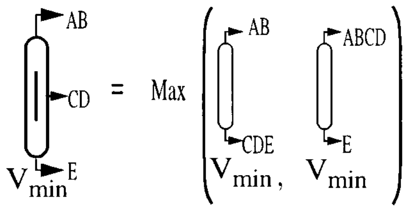


Figure 8. V_{min} diagram for a five-component feed used to find minimum vapor flow requirements in a three-product Petlyuk arrangement specification with nonsharp product splits AB/BCD/DE.

can be obtained only when the prefractionator is operated along the line between P_{BE} and P_{bal}.

Note that a peak in the V_{min} diagram is simply the vapor flow requirement for a particular sharp split in an ordinary two-product column. Thus, the minimum vapor flow requirement for the Petlyuk arrangement is given by most difficult split between two of our specified product groups, if the separation was to be carried out in a conventional two-product column.

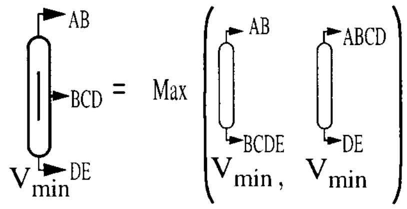
This is illustrated in "equation" (32). In this example P_{CD} is a higher peak than P_{BC} or P_{DE}, but this does not matter because we do not attempt to split the D and C components into separate products (subscript T,B is not used because we may consider either tops or bottoms).



(32)

5.3. Example: Nonsharp Separations. In the next example, as shown in Figure 8, we use the same feed and V_{min} diagram, but we change the product specifications so that all of the light A component is recovered in the top, all of the C component in the side stream, and all of the heavy E in the bottom. However, in this case we allow B to appear in both top and side-stream products and D to appear in both the side-stream and bottom products.

The solution is still quite simple to obtain from the V_{min} diagram. In the prefractionator, we need to remove A from the bottom and E from the top, and the minimum vapor flow in the prefractionator is found at the preferred split P_{AE}. This time all common roots carry over, and C21 and C22 become columns with four-component feeds. However, the interesting point of operation in the column is the sharp split between A and C. Because both θ_A and θ_B carry over from C1, the minimum vapor flow in the top of C21 is trivially found at P_{AC}. Similarly, P_{CE} will give the requirement in C22. Again, the separation is found to be exactly the same as the most difficult product split when we compare one and one such split in an ordinary two-product distillation column as shown in "equation" (33).



(33)

Note that, in both of these examples, the bold lines represent a minimum-energy solution for a sharp split between a pair of specified (composite) products in either the top or bottom of an ordinary two-product column.

6. Discussion

6.1. Improved Second Law Results in Petlyuk Arrangements. Several authors, e.g., Carlberg and Westerberg,[7] Agrawal and Fidkowski,[14] and Annakou and Mizsey,[15] mention that a typical Petlyuk column, where all of the heat input is done at the highest temperature level and all of the heat removal is done at the lowest temperature level, has a drawback compared to conventional arrangements where some heat is added and removed at intermediate levels. Even if the overall vapor flow rate, which can be regarded as a first law (of thermodynamics) effect, is always less than that in a conventional arrangement,[16] the temperature range between heat input and removal is always the

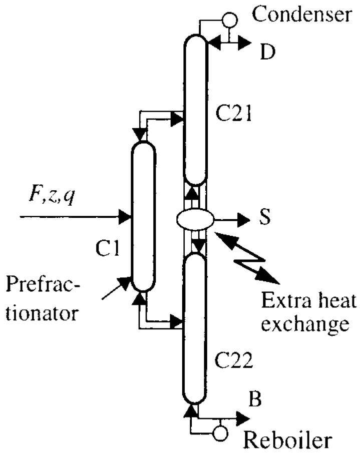


Figure 9. Petlyuk arrangement with an extra heat exchanger at the side-stream stage.

largest boiling point difference, which gives a low performance in terms of the second law effect. Thus, to recommend a Petlyuk arrangement, the first law effect must dominate over the second law effect with respect to the utility requirement.

However, when the peaks in the V_{min} diagram are of different height, this implies that a change in vapor flow could be allowed at the side-stream stage. In the case when the vapor flow requirement in the lower end is larger, this may easily be realized by extracting some of the side-stream product as vapor. This may be done either directly or by withdrawing all of the liquid from C21 and returning it slightly cooled, exactly sufficient to condense the required change in vapor at the return stage. In cases where the vapor flow in C21 is higher, some of the heat can be supplied at the side-stream stage. The maximum flow rate is still given by the highest peak, but not all of it has to be supplied or removed at the most extreme temperatures.

A heat exchanger at the side-stream stage as illustrated in Figure 9 can ensure that both C21 and C22 are operated at minimum energy at the same time. The actual change in vapor flow can easily be found from the V_{min} diagram as the difference height of the two peaks P_{AB} and P_{BC} (see ΔV_P in Figure 5). The prefractionator now has to be operated exactly at its preferred split.

The cases where the second law effect cannot be improved is for a balanced main column. Then the vapor flow requirements are the same in the top and bottom, and this is also the case where we obtain the largest vapor flow rate savings, compared to the best of conventional direct or indirect split configurations (refer to Chapter 8 of work by Halvorsen[8]). In these cases the first law effect is most likely to dominate over the second law effect.

We have not done a detailed comparative study with other types of columns and heat integration, taking a heat exchanger at the side-stream stage into consideration, but it is clear that some results in other studies[14,15] would have been more favorable for a Petlyuk arrangement if this extra heat-exchange ability had been included.

6.2. Composition Profiles. An operational and computational advantage with the directly connected prefractionator is that we may decouple the feed split, expressed by the net flow of each component ($w_{i,T}$) from the composition in the flow leaving the column. In Figure 10 the profiles for the preferred split are shown for a Petlyuk column prefractionator (a) and a conventional column (b). The end-feed compositions have been set equal to the pinch zone compositions in each end of the Petlyuk prefractionator. This implies that the vapor and liquid compositions in each end are at equilibrium, and these will also be the feed pinch compositions of the succeeding columns when the Petlyuk arrangement is operated at minimum energy.

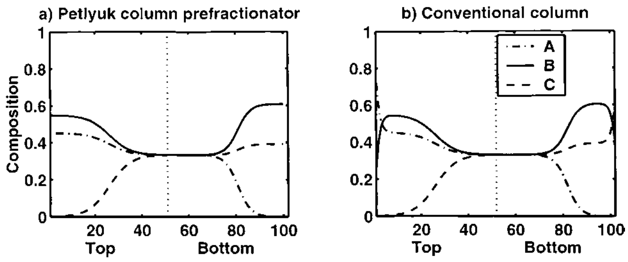


Figure 10. Composition profiles as the preferred split. Feed data $z = [0.33, 0.33, 0.33]$, $\alpha = [4, 2, 1]$, and $q = 1$. End feeds in part a are set equal to pinch zone compositions.

The differences between the conventional and directly coupled column are observed toward the ends. In the conventional columns, remixing occurs caused by recycling of the condenser and reboiler products.

6.3. Nonoptimal Operation. In the case of operation of the prefractionator outside the flat optimality region of the Petlyuk arrangement, the energy requirement increases rapidly.[17] In some cases we may get recycling of net flow of the intermediate component from either column C21 or column C22 back into the prefractionator column. This violates Underwood's assumption about positive net flow of components from the feed to each of the product ends as discussed in section 3.1. However, with some constraints on the composition in the liquid entering at the top or the vapor into the bottom, we may still use Underwood's equations to compute the minimum vapor flow solutions for all parts of the arrangement. This issue is treated in more detail in the thesis by Halvorsen[8] also for nonsharp product specifications.

6.4. Extra Condenser or Reboiler in the Prefractionator. Several authors, e.g., Agrawal and Fidkowski,[14] have pointed out that, in some cases, the overall minimum vapor flow rate may be unaffected if a condenser is used at the prefractionator top as shown in Figure 11. This is very simple to assess by the V_{min} diagram. The effect of introducing a prefractionator condenser is that the vapor requirement in column C21 increases. To obtain the same minimum boilup requirement, first the peak P_{BC} has to be the highest peak, and second the difference between the peaks has to be larger than the additional vapor requirement in column C21 imposed by a (possibly partial) condenser on the top of column C1.

Similarly, a combined arrangement with a direct coupling between columns C1 and C21 and a reboiler at the bottom of C1 and conventional feed to C22 may require the same total minimum vapor flow as a Petlyuk arrangement only if the peak P_{AB} is significantly higher than P_{BC}.

6.5. Use of a Conventional Prefractionator Column. A configuration with a conventional prefractionator column with its own reboiler and condenser as shown in Figure 12 was studied by Christiansen.[18] This approach may in some cases come close to the Petlyuk arrangement in terms of overall vapor flow but will

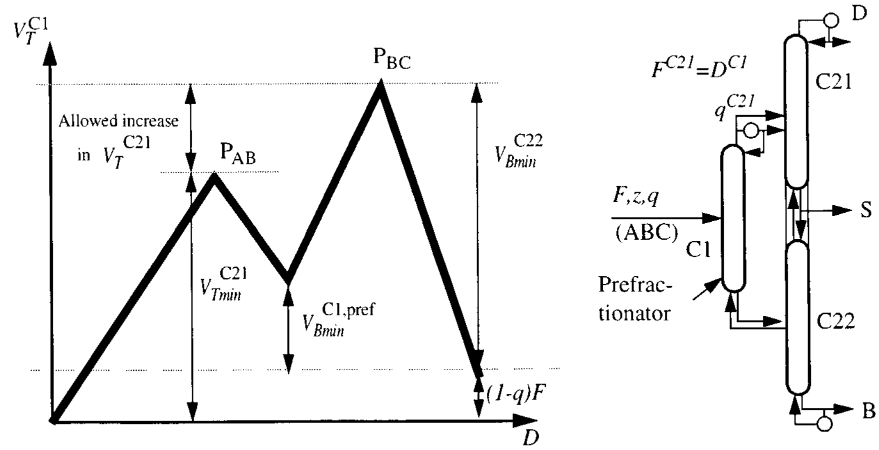


Figure 11. V_{min} diagram for the three-component feed (ABC) giving directly the allowed increase in the vapor flow in column C21. The modified Petlyuk arrangement may then obtain the same minimum reboiler flow as the full Petlyuk arrangement, given by P_{BC}.

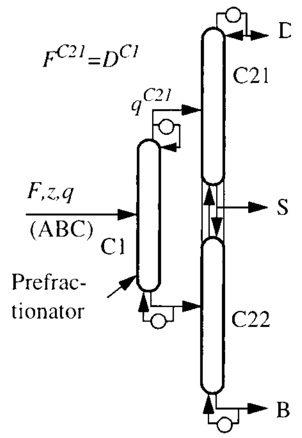


Figure 12. Conventional prefractionator arrangement.

never be better. In other cases, the minimum vapor flow will be higher than that with the conventional configurations. Halvorsen[8] showed that the optimum is always found when the prefractionator is operated exactly at the preferred split. We will also have an operating point where the main column is balanced, but in the this case there is no completely flat optimality region because the total vapor flow with a balanced main column will always be slightly above the requirement at preferred split operation.

6.6. Real Mixtures. As shown in part 1 of this series, a V_{min} diagram can be made for real (zeotropic) mixtures. This implies that we may use the V_{min} diagram for assessment of the separation of real mixtures in the Petlyuk arrangement too. However, unlike in the ideal case where the V_{min} diagram is a visualization of the exact analytical solution for minimum energy, we have to treat it as a tool that gives us approximate estimates, and the accuracy will, of course, be best for close to ideal mixtures. For more accurate computations, we must adjust for changes in molar flows and other properties along the column sections.

The main characteristic of the minimum-energy solution is still (with reservations for some very nonideal cases) that the prefractionator should be operated at its preferred split. This gives us the feed distribution in column C1, and thereby the feed stage conditions and the minimum energy requirements for the succeeding columns may easily be calculated numerically, for example, in a rigorous process simulator.

7. Conclusion

The minimum-energy solution for a three-product Petlyuk arrangement has been analyzed. The solution is given by the highest peak in the V_{min} diagram for the feed, and this is equivalent to the following rule:

The minimum total vapor flow requirement in a Petlyuk arrangement is the same as the required vapor flow for the most difficult split between two of the specified products if that separation was to be carried out in a single conventional two-product column.

The V_{min} diagram is based on feed data only, and in addition to the overall vapor flow requirement, we find the individual vapor flow requirement for each column section, directly from the same diagram as shown in Figure 5.

The plain Petlyuk arrangement will probably be most attractive when the peaks in the V_{min} diagram are of similar height. Otherwise, combined arrangements may give a similar performance in terms of minimum vapor flow and an even better performance in terms of separation work.

The minimum-energy expression in itself is not a new result. However, the simple way to "see" the answer as "the most difficult binary split", the generalization to multicomponent feed and nonsharp product specifications, and assessment by the V_{min} diagram are hopefully useful contributions to the distillation literature. In this paper we have limited the analysis to three-product Petlyuk columns, and we left an open question if a similar approach can be used for more than three products. We reveal that this is possible indeed, and the generalization to extended multiproduct Petlyuk arrangements is the subject in part 3 of this series.[19]

Literature Cited

(1) Petlyuk, F. B.; Platonov, V. M.; Slavinskii, D. M Thermodynamically optimal method for separating multicomponent mixtures. *Int. Chem. Eng.* **1965**, *5* (No. 3), 555−561.

(2) Stichlmair, J. Distillation and Rectification. *Ullmann's Encyclopedia of Industrial Chemistry*; VCH: Weinheim, Germany, 1988; Vol. B3, pp 4-1−4-94.

(3) Fidkowski, Z.; Krolikowski, L. Thermally Coupled System of Distillation Columns: Optimization Procedure. *AIChE J.* **1986**, *32* (No. 4), 537−546.

(4) Christiansen, A. C.; Skogestad, S. Energy Savings in Integrated Petlyuk Distillation Arrangements. Importance of Using the Preferred Separation. AIChE Annual Meeting, Los Angeles, Nov 1997; Paper 199d (updated version is found in work by Christiansen[18]).

(5) Glinos, K. N.; Nikolaides, I. P.; Malone, M. F. New complex column arrangements for ideal distillation. *Ind. Eng. Chem. Process Des. Dev.* **1986**, *25* (No. 3), 694−699.

(6) Carlberg, N. A.; Westerberg, A. W. Temperature−Heat Diagrams for Complex Columns. 3. Underwood's Method for the Petlyuk Configuration. *Ind. Eng. Chem. Res.* **1989**, *28*, 1386−1397.

(7) Carlberg, N. A.; Westerberg, A. W. Temperature−Heat Diagrams for Complex Columns. 2. Underwood's Method for Side-strippers and Enrichers. *Ind. Eng. Chem. Res.* **1989**, *28*, 1379−1386.

(8) Halvorsen, I. J. Minimum Energy Requirements in Complex Distillation Arrangements. Dr. ing. Thesis, Norwegian University of Science and Technology (NTNU), 2001. Available from the web page of Sigurd Skogestad, Department of Chemical Engineering at NTNU (May 2002: http://www.chemeng.ntnu.no/∼skoge/publications/thesis/2001/halvorsen/).

(9) Halvorsen, I. J.; Skogestad, S. Minimum Energy Consumption in Multicomponent Distillation. 1. V_{min} Diagram for a two-product column. *Ind. Eng. Chem. Res.* **2003**, *42*, 596−604.

(10) Underwood, A. J. V.; et al. Fractional Distillation of Ternary Mixtures. Part II. *J. Inst. Pet.* **1946**, *32*, 598−613.

(11) Underwood, A. J. V. Fractional Distillation of Multi-Component Mixtures—Calculation of Minimum Reflux Ratio. *J. Inst. Pet.* **1946**, *32*, 614−626.

(12) Underwood, A. J. V Fractional Distillation of Multi-Component Mixtures. *Chem. Eng. Prog.* **1948**, *44* (No. 8).

(13) Underwood, A. J. V.; et al. Fractional Distillation of Ternary Mixtures. Part I. *J. Inst. Pet.* **1945**, *31*, 111−118.

(14) Agrawal, R.; Fidkowski, Z. Are Thermally Coupled Distillation Columns Always Thermodynamically More Efficient for Ternary Distillations. *Ind. Eng. Chem. Res.* **1998**, *37* (No. 8), 3444−3454.

(15) Annakou, O.; Mizsey, P. Rigorous Comparative Study of Energy-Integrated Distillation Schemes. *Ind. Eng. Chem. Res.* **1999**, *35* (No. 6), 1877−1855.

(16) Fidkowski, Z.; Krolikowski, L. Minimum Energy Requirements of Thermally Coupled Distillation Systems. *AIChE J.* **1987**, *33* (No. 4), 643−653.

(17) Halvorsen, I. J.; Skogestad, S. Optimal Operation of Petlyuk Distillation: Steady-state Behaviour. *J. Process Control* **1999**, *9*, 407−424.

(18) Christiansen, A. C. Studies on optimal design and operation of integrated distillation arrangements. Ph.D. Thesis, Norwegian University of Science and Technology (NTNU), 1997 http://www.chemeng.ntnu.no/∼skoge/publications/thesis/1998/christiansen/.

(19) Halvorsen, I. J.; Skogestad, S. Minimum Energy Consumption in Multicomponent Distillation. 3. Generalized Petlyuk Arrangements with More Than Three Products. *Ind. Eng. Chem. Res.* **2003**, *42*, 616−629.

Received for review October 19, 2001
Revised manuscript received June 3, 2002
Accepted June 24, 2002

IE0108649