Energy Savings in Complex Distillation Arrangements: Importance of Using the Preferred Separation

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Abstract

Prefractionator arrangements are often preferred from an energy point of view when separating ternary mixtures. The prefractionator performs a separation between the heaviest and lightest component, whereas the intermediate component distributes to both products. The energy usage in the prefractionator itself has a very sharp minimum for a particular distribution, which is the "preferred separation" of Stichlmair (1988). On the other hand, the energy usage in the downstream main column has a minimum when the two parts of the column, above and below the side stream, are "balanced". In the paper we derive simple analytic expression for the total energy usage of the two-column sequence as a function of the separation in the prefractionator. We find that although the preferred separation is optimal, at least for sharp splits in infinite columns, the energy usage is almost the same for any separation between the "preferred" and the "balanced". The same results are shown numerically to hold for columns with finite number of stages and non-sharp separation, as well as when the prefractionator and main column are directly coupled, as in the Petlyuk arrangement. Finally, some implications for the operation and control of such columns are discussed.

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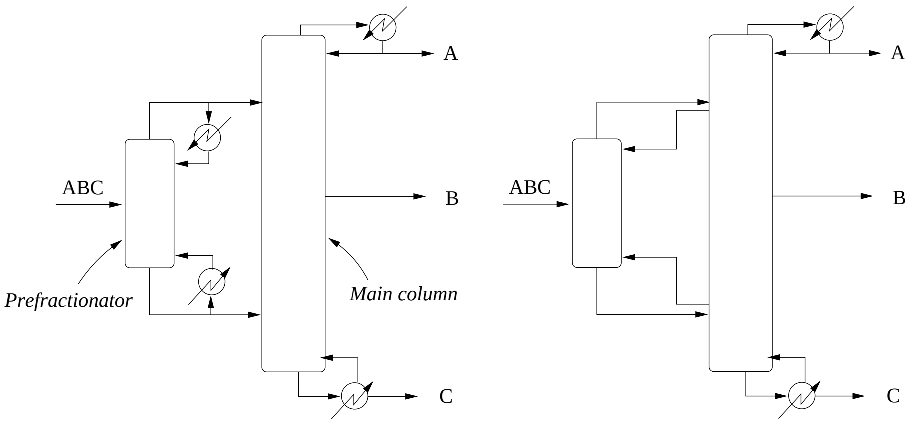
1 Introduction

The task of finding the minimum energy input for multicomponent separations has received considerable interest in the literature. No doubt the most well known methods are those due to Underwood (e.g. Underwood (1948)), for which minimum flows are obtained through an exact (iterative) solution of the material balance equations corresponding to infinite columns. In order to enhance the understanding of minimum energy conditions beyond that of numerical computations, Petlyuk and coworkers dedicated a series of papers (e.g. Petlyuk and Platonov (1964), Petlyuk *et al.* (1965) and Petlyuk *et al.* (1966)) to the task of selecting the *thermodynamically* optimal distillation scheme. Based on the concept of reversible distillation, the authors argue that one of the optimality conditions, is that in each bisectional column only the components with extreme volatilities should be separated. For the ternary case this implies that to ensure reversible mixing of streams, the first split is taken between the light and heavy component, so that the intermediate component distributes between the bottoms and top products. Any other split between adjacent components will inherently introduce additional exergy loss and thus increase the energy usage. However, we strongly emphasize that the concept of reversibility also requires uniform distribution of utility (condensing and boilup) along the column, which is not realized in columns with one reboiler and one condenser. Thus, although arguments based on reversibility may provide expedient guidelines, one needs a more detailed analysis to provide conclusions for real columns.

The issue of minimum energy usage under the presence of distributing components was also examined by King (1971). The author introduced a shortcut (group) method to compute the minimum energy, but until present it remains somewhat unclear as to under which conditions the results of King (1971) apply. Stichlmair (1988) coined the phrase *preferred separation*, to denote the separation in the prefractionator that requires the minimum energy input. This particular split occurs when all components have a pinch at the feed location, and the author demonstrates that the optimality of this particular split is due to colinearity between the distillation and equilibrium lines at the feed point. However, the author does not elaborate on whether carrying out the preferred separation as the first split should give the *overall* minimum energy input for a sequence of columns, although is is stated that it *"usually"* is so.

In this paper we consider separating ternary mixtures in the prefractionator arrangements shown in Figure 1. This includes a "conventional" prefractionator (a) as well as the Petlyuk column (b), where the prefractionator and main columns are directly coupled so that the prefractionator has no heater or cooler. Both of these arrangements are interesting alterna-



(a) Prefractionator (b) Petlyuk column

Figure 1: Prefractionator arrangements for separation of ternary mixtures

tives for industrial implementations. The task for the prefractionator is to split the heavy and light components, whereas the intermediate component distributes to both products. The downstream main column is a side-stream column where the three components may be recovered as pure products.

Several authors have considered methods to obtain the minimum energy usage for the Petlyuk column (see e.g. Cerda and Wersterberg (1981), Fidkowski and Krolikowski (1986), Nikolaides and Malone (1988), Glinos and Malone (1988) and Carlberg and Westerberg (1989)). Without going into detailed discussions of these previous works, we put emphasis on the important finding by Fidkowski and Krolikowski (1986). Using a comprehensive analysis of the Underwood equations, the authors showed that that there is a *region* for the recovery of the intermediate component in which the minimum energy usage in fact is *constant.* According to Glinos and Malone (1988) the formulas presented by Fidkowski and Krolikowski (1986) was previously derived in a thesis of Stupin (1970). In this work we give results from numerical simulations which demonstrate that the results of Fidkowski and Krolikowski (1986) carry over also to columns with a finite number of stages. The results are obtained from numerical simulations of columns with a sufficiently large number of stages, assuming constant molar flows and constant relative volatilities. In this paper we provide results also for the prefractionator arrangement in Figure 1 (a). In particular, whereas the previous works consider exact solutions using Underwood's method, we in this work propose approximate *explicit* expressions that proves useful in the analysis of the prefractionator arrangement. By first considering the prefractionator arrangement, we also obtain physical insight related to the regions of constant energy usage (Fidkowski and Krolikowski 1986) for the Petlyuk column.

For the prefractionator arrangement, we demonstrate that there is a similar "flat" region where the energy usage remains relatively constant. We show analytically and numerically that this region is characterized by recoveries of the intermediate component corresponding to the *preferred* separation and a *balanced* main column. We then elaborate on an important issue that has not been given appropriate attention by the previous authors. This refers to the importance that this "flat" or constant region has for practical operation. In terms of practical operation, we find for both column arrangements that one may control the composition only at one end in the prefractionator and "overpurify" the other column end ("one–point control") without significant increases in the energy usage. Which end to control depends on whether the preferred or a balanced separation requires the largest recovery of intermediate component in the prefractionator.

We also consider briefly the issue of *non–sharp* separations, for which we present results from numerical simulations where the product purity of the intermediate component is decreased relative to a pure product. The results show that the minimum energy usage moves away from the preferred separation for *sharp splits*, which in itself is hardly surprising. We give an account for the results by considering the distribution of the light and heavy components in the prefractionator for non-sharp separations.

Finally we stress that for ideal mixtures it is always optimal in terms of boilup to use a vapor feed when possible. This is also discussed in the forthcomong thesis (Christiansen 1997), where we find that taking this into account has a strong impact on previous results in the literature (e.g. the optimality regions for the direct and indirect split schemes presented by Glinos and Malone (1988)). Hence we consider prefractionator arrangements in which *partial condensers* are used for upstream columns, providing vapor feeds to downstream columns. In order to analyze such prefractionator arrangements we "decompose" the task, so that we first consider the prefractionator column and then the main column. Knowledge from these findings are then combined to understand the behavior when we consider the column *sequence.* In total we hope to provide a lucid and comprehensive overview of the literature, that may clear up some apparent misunderstandings.

2 Degrees of Freedom Analysis

One of the important aspects to be addressed in this paper, is how one should utilize the degrees of freedom (DOFs) for the prefractionator arrangements in Figure 1 in a "optimal" manner. For both arrangements we have after stabilization of levels and pressure five DOFs available for operation. Three of these are then consumed if we specify one purity in each product. The objective of this paper is then partly to indicate how one should treat the remaining two DOFs. In a sense, they are both related to the prefractionator column, even though the DOFs in a strict sense apply to the column *arrangement*.

For the Petlyuk column in Figure 1 (b), one might imagine that one of the remaining DOFs could be used to control one of the impurities in the sidestream product. The last DOF is then used to minimize the energy usage. However, due to the coupling between the upper and lower parts of the main column, it is in practice not possible to control two purities in the sidestream. Wolff and Skogestad (1995) showed that "holes" may appear in certain operating regions in this case. A detailed explanation for this behavior was later given by Morud (1995). The conclusion is that one should control only one composition in each product for the Petlyuk column. In this work we show that for "optimal" operation one in practice needs to use one DOF to stay within a certain operating region where the energy usage in fact remains relatively constant.

3 The Prefractionator Column

In this section we present expressions for the minimum energy usage V_{min} for ternary separations in a prefractionator column, using a group method previously introduced by King (1971). The feed is assumed to be saturated liquid (in Appendix A we also give the formulas that apply for *saturated vapor feeds*). Note that we in the following use subscript *min* to denote a column with an infinite number of stages, and that we use lowercase letters to denote the distillate (d) and bottoms (b) flows to avoid confusion with the components (B) and the superscripts for the direct split scheme (e.g. V^D).

3.1 V_{min} and the preferred separation

Our starting point is the "binary equation" for the minimum reflux in a column with an infinite number of stages and a *saturated liquid feed* (King 1971)

$$\left(\frac{L}{F}\right)_{min} = \frac{\phi_L^d - \alpha_{LH}\phi_H^d}{\alpha_{LH} - 1} \tag{1}$$

Here L denotes the reflux, F the feed, ϕ_i^d the fractional recoveries of light and heavy components in the distillate d and α_{LH} the relative volatility between the two components. More precisely, the fractional recoveries are given by

$$\phi_i^d = \frac{d x_i^d}{F z_i} \tag{2}$$

where z_i denotes the feed composition of component i. Actually, equation (1) applies to any two components in a multicomponent mixture if we assume that <u>all</u> components pinch at the feed stage, and King (1971) states that (1) applies if all *non-key* components distribute. In practice this means that the non-key components must be intermediate relative to the two (key) components.

If we then consider the separation of a ternary mixture ABC, for which we want to obtain a top product depleted in the high boiler (C) and a bottoms product depleted in the low boiler (A), equation (1) is valid if B has a pinch at the feed location. This is the *"preferred"* separation of Stichlmair (1988), and the corresponding preferred recovery of B is denoted ϕ^{pref}. For the separation between components A and C equation (1) gives

$$\left(\frac{L}{F}\right)_{min}^{pref} = \frac{\phi_A^d - \alpha_{AC}\phi_C^d}{\alpha_{AC} - 1} \tag{3}$$

where we use the superscript $pref$ to make clear that it only applies to the case of the preferred separation where all components have a pinch at the feed stage. The corresponding minimum boilup is

$$\left(\frac{V}{F}\right)_{min}^{pref} = \left(\frac{L}{F}\right)_{min}^{pref} + \left(\frac{d}{F}\right)^{pref} \tag{4}$$

In this case equation (1) applies also to the (non–sharp) separation between components A and B, i.e.

$$\left(\frac{L}{F}\right)_{min} = \frac{\phi_A^d - \alpha_{AB}\phi_B^d}{\alpha_{AB} - 1} \tag{5}$$

By equating (3) and (5) for given values of ϕ_A^d and ϕ_C^d, and solving with respect to ϕ_B^d, we obtain the "preferred" recovery of B in the distillate

$$\phi^{pref} \equiv \left(\phi_B^d\right)^{pref} = \frac{\phi_A^d\left(\alpha_{BC} - 1\right) + \alpha_{BC}\phi_C^d\left(\alpha_{AB} - 1\right)}{\alpha_{AC} - 1} \tag{6}$$

The distillate flow is then

$$\frac{d^{pref}}{F} = \phi_A^d z_A + \phi^{pref} z_B + \phi_C^d z_C \tag{7}$$

and we derive the desired expression for V_{min}

$$\left(\frac{V}{F}\right)_{min}^{pref} = \frac{\phi_A^d - \alpha_{AC}\phi_C^d}{\alpha_{AC} - 1} + \phi_A^d z_A + \phi^{pref} z_B + \phi_C^d z_C \tag{8}$$

For the special case of a sharp split between A and C ($\phi_A^d = 1$, $\phi_C^d = 0$) we get

$$\phi^{pref} = \frac{\alpha_{BC} - 1}{\alpha_{AC} - 1}, \quad \frac{d^{pref}}{F} = z_A + \tfrac{\alpha_{BC}-1}{\alpha_{AC}-1} z_B \tag{9}$$

and for the boilup

$$\left(\frac{V}{F}\right)_{min}^{pref} = \frac{1}{\alpha_{AC} - 1} + z_A + \frac{\alpha_{BC} - 1}{\alpha_{AC} - 1} z_B \tag{10}$$

This is the same expression as was previously presented by Stichlmair (1988) for the preferred separation.

However, the question remains as to how V_{min} changes if ϕ_B^d differs from the particular value ϕ^{pref}, and what is the additional cost? This is the central question to be addressed in the next section.

3.2 V_{min} for splits other than the preferred separation

In the following we want to derive an expression for V_{min} for sharp splits between A and C that applies to *any* recovery of the intermediate component, i.e. for all $\phi_B^d \in [0, 1]$.

Introductory example.

Table 1: Data for ternary separation in an "infinite" column

Number of stages	$N = 100$ $(N_F = 50)$
Feed compositions	$z_F = [1/3, 1/3, 1/3]$
Molar feed	$F = 1$ [kmol/min]
Relative volatility	$\alpha = 4 : 2 : 1$
Impurity spec.'s	$x_A^b \le \epsilon$
	$y_C^d \le \epsilon$

In order to address this issue, we first consider an introductory example for the separation of an equimolar saturated liquid feed with data given in Table 1. For the simulations we specify that the composition of light component A in the bottom, and the composition of heavy C in the top, should be equal to or less than a given upper bound, i.e. $x_A^b \le \epsilon$ and $y_C^d \le \epsilon$. Note that for a sharp split $\epsilon \to 0$, but in the numerical calculations we mostly use $\epsilon = 10^{-4}$ (we should also mention that we used a finite number of stages $N = 100$, but exact calculation with the Underwood equations for infinite columns give almost identical results for V_{min}). The purpose of this numerical example is to compute V_{min} as a function of the distillate flow d with $\phi_B^d \in [0, 1]$.

By specifying d we fix one degree of freedom, and since we have only two degrees of freedom for a single column at steady state, we will find that only one of the impurity specifications (i.e. $x_A^b \le \epsilon$ or $y_C^d \le \epsilon$) will be active as an equality. Numerically we thus obtained two solution curves where we selected $x_A^b = \epsilon = 10^{-4}$ and $y_C^d = \epsilon = 10^{-4}$. The curves were computed using the continuation scheme presented in (Christiansen 1997), and are shown in Figure 2. As seen from the Figure we find that there is a sharp minimum located

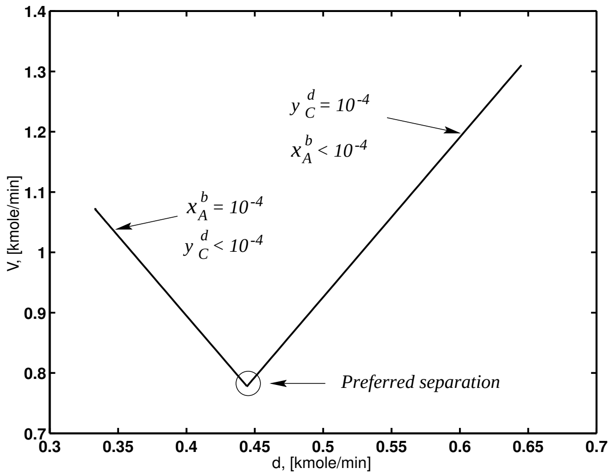


Figure 2: V_{min} as a function of distillate flow d for sharp A/C split, i.e. $\epsilon = 10^{-4}$.

at the intersection, which is the point corresponding to the preferred separation. Along the curves extending from this point we have that both purity specifications are satisfied, one as an equality the other as an inequality (see Figure 4). The only point where both appear as equalities ($x_A^b = y_C^d = \epsilon$) is at the intersection, which as mentioned is at the preferred separation.

Figure 3 which gives V_{min} for other values of ϵ, illustrates that selecting $\epsilon = 10^{-4}$ indeed gives the limiting value of V_{min} corresponding to a sharp separation. For example, the curves for purities of $\epsilon = 10^{-3}$ and $\epsilon = 10^{-4}$ are as shown in Figure 3 practically indistinguishable.

One may also note that there is a well defined minimum also for non-sharp separations, an

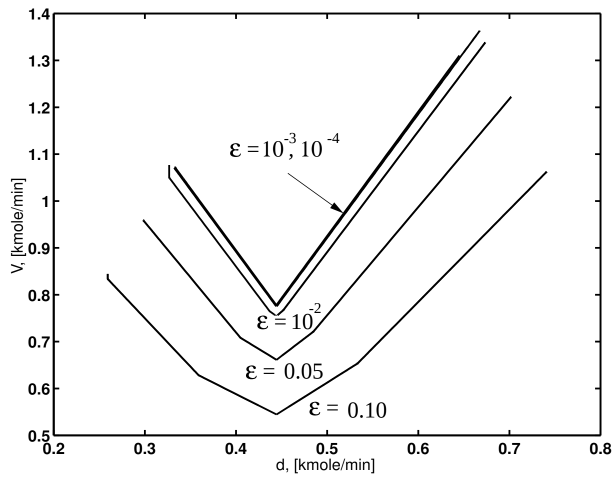


Figure 3: V_{min} as a function of distillate flow d for non-sharp separations with $\epsilon \in [10^{-4}, 10^{-1}]$.

issue to be discussed later. In Figure 4 we have plotted on a semi-log scale the corresponding impurities at the column ends which is purer than required. We see that the impurities are

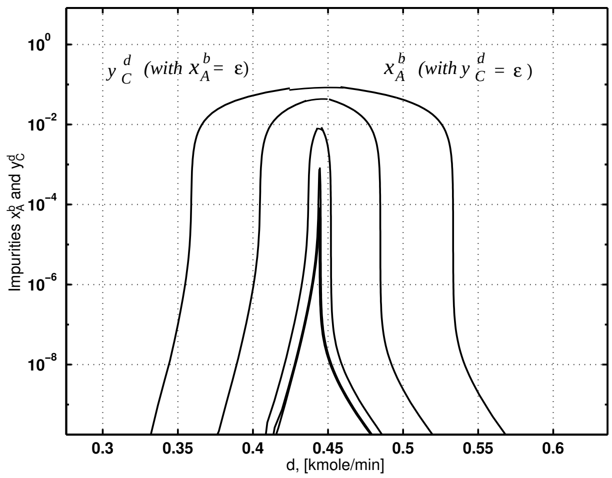


Figure 4: Impurities at column end which is purer than required for $\epsilon = 0.1, 0.05, 10^{-2}, 10^{-3}, 10^{-4}$. The left branches give $y_C^d < \epsilon$ when $x_A^b = \epsilon$ is kept constant, and the right branches give $x_A^b < \epsilon$ for constant $y_C^d = \epsilon$.

satisfied as inequalities for all values of d, except for the preferred separation where they are both satisfied as equalities. Similar observations were also made by Carlberg and Westerberg (1989), in a detailed analysis of the Underwood equations for a simple non-sharp column. The authors showed that minimum reflux behavior (infinite column) divides into four distinct regions depending on the recovery of intermediate. Each region is then characterized by the recoveries of light and heavy in the distillate being either at their lower bound, upper bound or intermediate. Note that we have instead assigned bounds on the *mole fractions* of heavy key in the top and light key in the bottom, whereas Carlberg and Westerberg (1989) consider the recoveries of light and and heavy in the top. Using either mole fractions or recoveries is however somewhat complementary from a mathematical point of view, since the recoveries depend linearly on the mole fractions. In practice, the mole fractions will however have more of a physical significance. We will later demonstrate that these observations are of great importance for practical operation of prefractionator arrangements and the Petlyuk column.

To further verify the numerical results, we compute from equation (10) the minimum vapor flow for a sharp split (with data from Table 1)

$$V_{min}^{pref} = 0.778 \tag{11}$$

and the corresponding distillate flow using equation (7) is

$$d^{pref} = 0.444 \tag{12}$$

which agrees with our numerical simulations.

3.3 Analytical Results

In this section we will derive approximate analytical expressions for V_{min} as a function of the distillate flow d for the prefractionator arrangement. The reader may note that we here choose to use d as the independent variable, since it represents a variable of grater physical significance than for instance the recovery ϕ_B^d. From King (1971) we have the following exact expression for the minimum boilup

$$\left(\frac{V}{F}\right)_{min} = \frac{\alpha_{AC}\phi_A^d}{\alpha_{AC}-\theta_i} + \frac{\alpha_{AB}\phi_B^d}{\alpha_{BC}-\theta_i} + \frac{\phi_C^d}{1-\theta_i} \tag{13}$$

where θ_i is the appropriate solution of the Underwood following 2nd order equation

$$\frac{\alpha_{AC}z_A}{\alpha_{AC}-\theta_i} + \frac{\alpha_{BC}z_B}{\alpha_{BC}-\theta_i} + \frac{z_C}{1-\theta_i} - F(1-q) = 0 \tag{14}$$

Here q is the feed enthalpy and $q = 1$ for saturated liquid feeds. For a sharp split between A and C ($\phi_A^d = 1$, $\phi_C^d = 0$) we get

$$\left(\frac{V}{F}\right)_{min} = \frac{\alpha_{AC}}{\alpha_{AC}-\theta} + \frac{\alpha_{AB}\phi_B^d}{\alpha_{BC}-\theta} \tag{15}$$

This proves that the relationship between V and ϕ_B^d, and thus between V and $d = Fz_A + F\phi^{pref}z_B$, is given by straight lines. This was already observed from the numerical results in Figures 2 and 3. The break in the straight lines at the preferred separation corresponds to a switch of Underwood root θ_i. As illustrated in Figure 2, the straight lines extend from the preferred separation to the two end points. The left end point is where all intermediate B leaves in the bottom, i.e. the direct A/BC split. Here $d = d^D$ and $V_{min} = V_{min}^D$. The right end point is where all B is taken over the top, i.e. the indirect AB/C split. Here $d = d^I$ and $V_{min} = V_{min}^I$. This is further illustrated graphically in Figure 5.

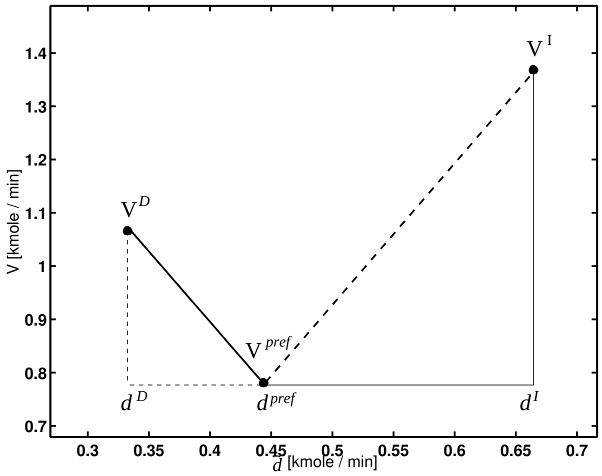
Since the relation between V_{min} and d yields a straight line for sharp splits and infinite columns, we thus find from Figure 5 the desired relationships

$$V_{min}^{C1} = V^{pref} + \frac{V_{min}^D - V^{pref}}{d^{pref}-d^D}\left(d^{pref}-d\right), \quad d < d^{pref} \tag{16}$$

$$V_{min}^{C1} = V^{pref} + \frac{V_{min}^I - V^{pref}}{d^I-d^{pref}}\left(d-d^{pref}\right), \quad d > d^{pref} \tag{17}$$

(We write V_{min}^{C1} with superscript $C1$ to make clear that these relationships apply only to the prefractionator column $C1$.) For sharp splits we have

$$\frac{d^D}{F} = z_A, \quad \frac{d^{pref}}{F} = z_A + \frac{\alpha_{BC}-1}{\alpha_{AC}-1}z_B, \quad \frac{d^I}{F} = z_A + z_B \tag{18}$$



(a) $\alpha = 4:2:1$

Figure 5: Boilup V for sharp A/C split as a function of the distillate flow d. The results are shown for an equimolar feed mixture with $\alpha_{AB} = \alpha_{BC} = 2$

Furthermore, Glinos and Malone (1984) derived reasonably accurate analytical expressions for V^D_{min} and V^I_{min} for sharp splits

$$\left(\frac{V^I}{F}\right)_{min} = \left(\frac{z_B + z_C}{\alpha_{BC} - 1} + \frac{z_A}{\alpha_{AC} - 1}\right) \frac{1}{1 + z_A z_C} + z_A + z_B \tag{19}$$

and

$$\left(\frac{V^D}{F}\right)_{min} = \frac{z_A + z_B}{f\left(\alpha_{AB} - 1\right)} + \frac{z_C}{f\left(\alpha_{AC} - 1\right)} + z_A \tag{20}$$

where $f = 1 + z_B/100$. The authors claim that these equations yield average errors in the order of 4% compared to exact methods (i.e. Underwood) and thus serve our purpose well. From (8) and (18)–(20) we derive the following expressions for the slopes in Figure 5

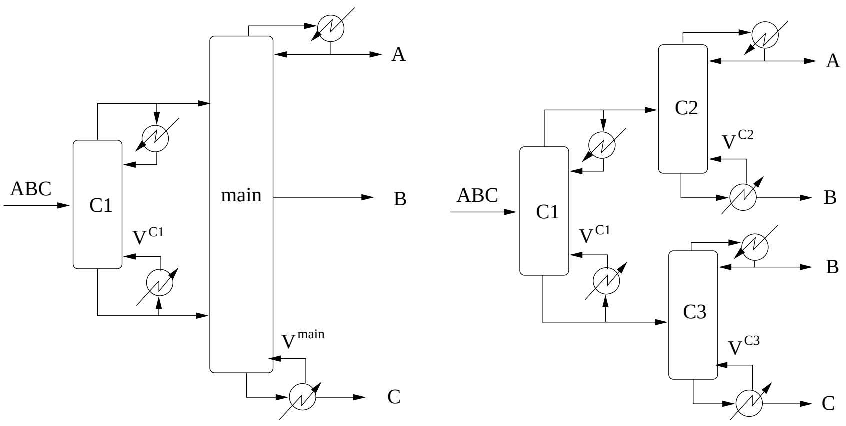
$$\frac{V^D_{min} - V^{pref}}{d^{pref} - d^D} = \frac{\left(\alpha_{AC} - 1\right)\left(z_A + z_B\right)}{f z_B \left(\alpha_{AB} - 1\right)\left(\alpha_{BC} - 1\right)} + \frac{z_C - 1}{f z_B \left(\alpha_{BC} - 1\right)} - 1 \tag{21}$$

$$\begin{aligned} \frac{V^I_{min} - V^{pref}}{d^I - d^{pref}} &= \frac{z_A - \left(1 + z_A z_C\right)}{z_B \left(\alpha_{AC} - \alpha_{BC}\right)\left(1 + z_A z_C\right)} \\ &\quad + \frac{\left(z_B + z_C\right)\left(\alpha_{AC} - 1\right)}{z_B \left(\alpha_{AC} - \alpha_{BC}\right)\left(1 + z_A z_C\right)\left(\alpha_{BC} - 1\right)} + 1 \end{aligned} \tag{22}$$

As one will expect we find that the slopes of the curves extending from the minimum (preferred separation) towards the direct and indirect splits, depend on the difficulty of the separation, i.e. on the ratio α_{AB}/α_{BC}. In the next section we use expressions (17)–(21) to determine (analytically) whether it is always optimal to produce the preferred separation as the first split for the Prefractionator arrangement in Figure 1 (a).

4 The Main Sidestream Column

We here consider the energy usage (V_{min}) in the main column with sharp separations between components A, B and C. To derive the desired expression we first consider the 3–column arrangement in Figure 6 (b), where the prefractionator is denoted $C1$ whereas the downstream columns are $C2$ and $C3$. Note that a partial condenser is used for the prefractionator column $C1$, since using a *total* condenser increases the energy usage in $C2$ by a term proportional to the feed of light key (Fz_A). For minimum reflux calculations we can represent the main



(a) 3–column arrangement (b) Prefractionator

Figure 6: Prefractionator arrangements for separation of ternary mixtures

column of the Prefractionator in Figure 6 (a) as a special case of Figure 6 (b), and we then refer to columns $C2$ and $C3$ in order to distinguish between cases when the *upper* or *lower* feed controls for the main column. The required energy usage in the main column is thus

$$V_{min}^{main} = max\left(V_{min}^{C2}, V_{min}^{C3}\right) \tag{23}$$

depending on whether the lower feed (V^{C3}) or upper feed V^{C2} controls, i.e. is the larger. To compute V_{min}^{C2} and V_{min}^{C3} we make use of the following exact expressions for a sharp separation of a binary mixture:

The feed to column $C3$ is *saturated liquid*, and for a sharp split between A and B we have from (1)

$$V_{min}^{C3} = \frac{F^{C3}}{\alpha_{BC} - 1} + d^{C3} \tag{24}$$

where $d^{C3} = F\left(1 - \phi_B^d\right) z_B$ is the distillate flow and $F^{C3} = F\left(1 - \phi_B^d\right) z_B + F z_C$ is the feed to column $C3$. F is as before the overall feed to the prefractionator $C1$, and ϕ_B^d is the fractional recovery of the intermediate component B in the distillate of the prefractionator. We then get for column $C3$

$$\left(\frac{V}{F}\right)_{min}^{C3} = \frac{\left(1 - \phi_B^d\right) z_B + z_C}{\alpha_{BC} - 1} + \left(1 - \phi_B^d\right) z_B \tag{25}$$

The feed to column $C2$ is a saturated vapor, and a similar derivation for a sharp split between B and C yields

$$\left(\frac{V}{F}\right)_{min}^{C2} = \frac{F^{C2}}{\alpha_{AB} - 1} = \frac{z_A + \phi_B^d z_B}{\alpha_{BC} - 1} + \left(1 - \phi_B^d\right) z_B \tag{26}$$

To compute V_{min}^{main} need to determine when the lower and upper feed controls respectively, for different mixtures and different values of ϕ_B^d. The "switch–over" value for ϕ_B^d occur when the main column is "balanced (i.e. $V_{min}^{C3} = V_{min}^{C2}$), and by equating (25) and (26) we find

$$\phi^{bal} = \frac{\left(1 - z_A\right)\left(\alpha_{AB} - 1\right) + z_B\left(\alpha_{AB} - 1\right)\left(\alpha_{BC} - 1\right) - z_A\left(\alpha_{BC} - 1\right)}{z_B\left(\alpha_{BC} - 1\right) + z_B\left(\alpha_{AB} - 1\right) + z_B\left(\alpha_{AB} - 1\right)\left(\alpha_{BC} - 1\right)} \tag{27}$$

We thus have the following three operating regimes for the main sidestream column

(i)	$\phi_B^d < \phi^{bal}$	Lower feed controls,	i.e. $V_{min}^{main} = V_{min}^{C3}$
(ii)	$\phi_B^d = \phi^{bal}$	Balanced column,	i.e. $V_{min}^{main} = V_{min}^{C2} = V_{min}^{C3}$
(iii)	$\phi_B^d > \phi^{bal}$	Upper feed controls,	i.e. $V_{min}^{main} = V_{min}^{C2}$

Since V_{min}^{C3} decreases as we increase ϕ_B^d, and since V_{min}^{C2} increases as we increase ϕ_B^d, we find for the main (sidestream) column that

$$\min_{\phi_B^d} V_{min}^{main} \tag{28}$$

is obtained for $\phi_B^d = \phi^{bal}$. Thus when we consider only the energy consumption in the main column, then the best choice is to operate the prefractionator such that $\phi_B^d = \phi^{bal}$. This is illustrated in Figure 7. Before proceeding we mention that if we instead use a total condenser

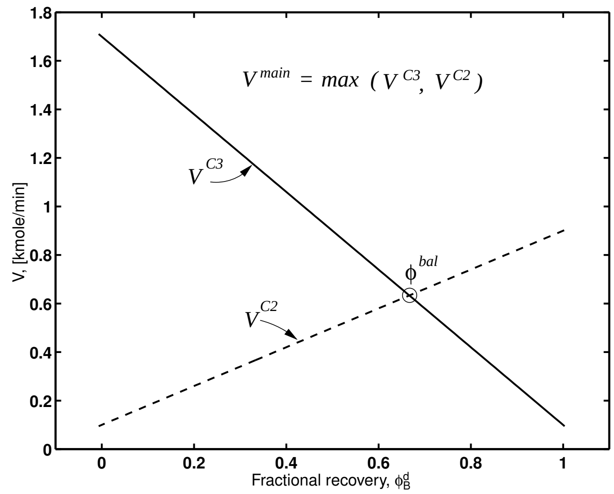


Figure 7: Minimum energy usage V_{min} for the main sidestream column as a function of the fractional recovery of intermediate ϕ_B^d for $\alpha = 4:2:1$ and $z_F = [1/3, 1/31/3]$. The Figure illustrates that the overall V_{min} corresponds to a balanced column and occurs for $\phi_B^d = \phi^{bal}$.

for the prefractionator so that the feed to $C2$ is a saturated liquid, then V_{min}^{C2} will increase and the value for ϕ^{bal} decreases. of columns?

5 Is the Preferred Separation Optimal for the Column Sequence?

In the previous sections we found that the minimum energy usage in a *prefractionator column* usually displays a very sharp minimum at the fractional recovery of intermediate component corresponding to the preferred separation (ϕ^{pref}), whereas the *main column* displays a similar sharp minimum for a balanced column (ϕ^{bal}). The question should then be posed: Which of these values, if any, is best when considering the overall energy consumption in the two columns. As mentioned in the introduction, Petlyuk (1997) and Stichlmair (1988) argue that the prefractionator dominates so that the preferred separation should be used. On the other hand, Triantafyllou and Smith (1992) base their short cut design procedure on the assumption that the main column should be balanced. Using the expressions derived in section 5.2 and 5.3 we will in this section derive approximate analytical equations for the optimality conditions for the prefractionator arrangement in Figure 1 (a) with an infinite number of stages and sharp splits. Based on these expressions we show that the preferred separation is indeed optimal for all cases, but the overall optimum is quite "flat" for values of ϕ_B^d in the range between ϕ^{pref} and ϕ^{bal}.

The overall energy consumption

$$V_{min} = V_{min}^{C1} + V_{min}^{main} \tag{29}$$

can be easily computed as a function of ϕ_B^d using the analytical expressions for V_{min}^{C1} in (17) and (16), and for V_{min}^{main} in (23). Note that we can alternatively use the distillate flow leaving the prefractionator as our independent variable, since there is a unique (linear) mapping from ϕ_B^d to d:

$$d = F \sum_{i=1}^{n} \phi_i^d z_{fi} = F\left(\phi_A^d z_A + \phi_B^d z_B + \phi_C^d z_C\right) \tag{30}$$

Note in particular that for sharp splits the differentials are related by

$$\partial d = z_B \cdot \partial \phi_B^d \tag{31}$$

since the amount of A and C are constant in the distillate, i.e. $\phi_A^d = 1$ and $\phi_C^d = 0$.

5.1 Introductory example

In order to motivate for analyzing the optimality conditions of the preferred separation, we first consider a introductory example where we compute V_{min} as a function of the fractional recovery ϕ_B^d, and compare the results also for the prefractionator V_{min}^{C1} and the main column V_{min}^{main}. We here consider a mixture with a large amount of intermediate B. Figure 8 shows V_{min} as a function of the recovery ϕ_B^d for the example with relative volatilities of $\alpha = 4 : 2 : 1$ and feed compositions $z_F = [0.1, 0.8, 0.1]$. We find for this example that

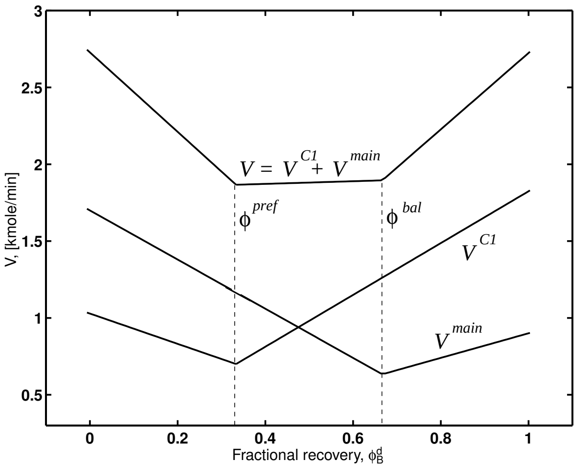


Figure 8: Minimum boilup V as a function of the fractional recover of intermediate ϕ_B^d for $\alpha = 4 : 2 : 1$ and $z_F = [0.1, 0.8, 0.1]$. The Figure illustrates that there is a large region enveloped by ϕ^{pref} and ϕ^{bal}, in which V remains close to the overall minimum.

V_{min} for the sequence of two columns indeed corresponds to using the preferred separation as the initial split. However, as mentioned we find that there is a relatively large region in which the energy usage is almost constant, independent of the recovery, i.e. the decrease in $V^{main} = V^{C3}$ is approximately equal to the increase in V^{C1}. This region covers all intermediate recoveries between ϕ^{pref} and ϕ^{bal}. Note that $\phi^{pref} < \phi^{bal}$ in this case, but for other cases we may have $\phi^{pref} > \phi^{bal}$, as shown in Figure 9. In this case the region with approximately constant V_{min} is between ϕ^{bal} and ϕ^{pref}.

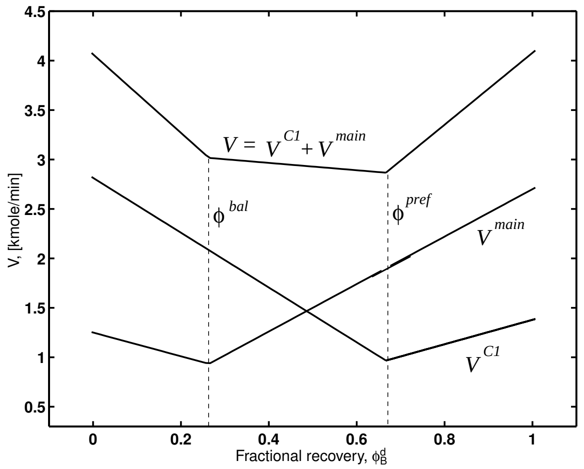


Figure 9: Minimum boilup V as a function of the fractional recover of intermediate ϕ_B^d for $\alpha = 4:3:1$ and $z_F = [0.1, 0.8, 0.1]$. The Figure illustrates that there is a large region enveloped by ϕ^{pref} and ϕ^{bal}, in which V remains close to the overall minimum.

5.2 Analytical results

We here use the previously derived analytical expressions to show that for sharp splits it is always optimal to use $\phi_B^d = \phi^{pref}$ in the prefractionator, i.e. $d = d^{pref}$. We do this by comparing the slopes (i.e. derivatives) for the prefractionator and the main column with respect to the distillate flow, i.e. $\partial V^{C1}/\partial d$ and $\partial V^{main}/\partial d$.

First consider the prefractionator $C1$. Using equations (17) and (22) we thus obtain for $d > d^{pref}$, (i.e. $\phi_B^d > \phi^{pref}$

$$\begin{aligned} \frac{\partial V^{C1}}{\partial d} &= \frac{V^I - V^{pref}}{d^I - d^{pref}} \\ &= \frac{z_A - (1 + z_A z_C)}{z_B(\alpha_{AC} - \alpha_{BC})(1 + z_A z_C)} \\ &\quad + \frac{(z_B + z_C)(\alpha_{AC} - 1)}{z_B(\alpha_{AC} - \alpha_{BC})(1 + z_A z_C)(\alpha_{BC} - 1)} + 1, \; (d > d^{pref}) \end{aligned} \tag{32}$$

Conversely we obtain using equations (16) and (21) for $d < d^{pref}$ (i.e. $\phi_B^d < \phi^{pref}$)

$$\begin{aligned} \frac{\partial V^{C1}}{\partial d} &= \frac{V^D - V^{pref}}{d^{pref} - d^D} \\ &= \frac{(\alpha_{AC} - 1)(z_A + z_B)}{f z_B (\alpha_{AB} - 1)(\alpha_{BC} - 1)} + \frac{z_C - 1}{f z_B(\alpha_{BC} - 1)} - 1, \; (d < d^{pref}) \end{aligned} \tag{33}$$

Next consider the main column, for which we obtain from (25) and (26)

$$\frac{\partial V^{main}}{\partial d} = \frac{\partial V^{C3}}{\partial d} = -\frac{\alpha_{BC}}{\alpha_{BC} - 1}, \; (d < d^{bal}) \tag{34}$$

and

$$\frac{\partial V^{main}}{\partial d} = \frac{\partial V^{C2}}{\partial d} = \frac{1}{\alpha_{AB} - 1}, \ (d > d^{bal}) \tag{35}$$

Note that these derivatives are *exact* for sharp splits and binary mixtures, since the minimum energy usage for the main column is equal to the *binary* separation requiring the largest energy. Our objective is now to determine whether the preferred separation is always optimal for the directly coupled Prefractionator. A simple analysis shows that we must consider the slope in the "flat" region for the two cases of

(1) $d^{pref} > d^{bal}$

and

(2) $d^{pref} < d^{bal}$

Case 1 : $d^{pref} > d^{bal}$ (Figure 10)
In this case the upper feed controls in the "flat" region and the preferred separation is *not* optimal if for $d^{bal} < d < d^{pref}$ we have $\partial V / \partial d$ positive, i.e. the overall energy usage (V) is smaller for some value of d than for d^{pref}. From (32) and (35) this is the case if and only if

$$\frac{1}{\alpha_{AB} - 1} > \frac{(\alpha_{AC} - 1)(z_A + z_B)}{f z_B (\alpha_{AB} - 1)(\alpha_{BC} - 1)} + \frac{z_C - 1}{f z_B (\alpha_{BC} - 1)} - 1 \tag{36}$$

After substituting for $\alpha_{AC} = \alpha_{AB}\alpha_{BC}$ and $z_C = 1 - (z_A + z_B)$ and some algebra, we find that the condition is equivalent to

$$\frac{(z_A + z_B)(\alpha_{AC} - 1 - \alpha_{AB} + 1) - f z_B (\alpha_{BC} - 1)(\alpha_{AB} - 1 + 1)}{f z_B (\alpha_{AB} - 1)(\alpha_{BC} - 1)} < 0 \tag{37}$$

$$\Downarrow$$

$$\frac{\alpha_{AB}(z_A + z_B)(\alpha_{BC} - 1) - \alpha_{AB} f z_B (\alpha_{BC} - 1)}{f z_B (\alpha_{AB} - 1)(\alpha_{BC} - 1)} < 0 \tag{38}$$

$$\Downarrow$$

$$\frac{\alpha_{AB}(z_A + (1 - f) z_B)}{f z_B (\alpha_{AB} - 1)} < 0 \tag{39}$$

Since the nominator is always larger than zero, we consider only the denominator. After substituting for $f = 1 + B/100$ we derive

$$\frac{\partial V}{\partial d} > 0 \quad \text{iff} \quad z_A < (f - 1) z_B = \frac{z_B^2}{100} \tag{40}$$

which is satisfied only when z_A is very small. From this result we see that the preferred separation is optimal in almost all cases, but we are lead to believe that there may exist some limiting cases with z_A small where the preferred separation is not optimal for the column sequence. However, it should be noted that (40) is based on the analytical expressions of (Glinos and Malone 1988) which are are not quite exact. In order provide the *exact* optimality conditions for the preferred separation, one will have to use exact methods such as Underwood's method as shown by Fidkowski and Krolikowski (1986).

To verify the optimality condition (40) we give in Figure 10 numerical results for an example where we also applied Underwood's method to compute V^D and V^I used in equations (17) and (16). According to condition (40) we have that for this case ($z_A < z_B^2/100$), the true V_{min} should not correspond to the preferred separation. However, using Underwood's method instead of the approximate expressions by Glinos and Malone (1984), we find that the preferred separation indeed gives the true V_{min}. Thus it is for all separations, at least

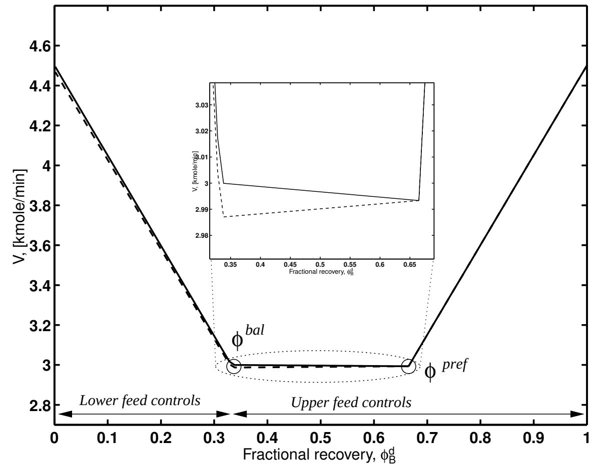


Figure 10: Analytical results for boilup V as a function of the fractional recover of intermediate ϕ_B^d for $\alpha = 4 : 3 : 1$ and $z_F = [0.005, 0.99, 0.005]$. The Figure shows that the overall V_{min} corresponds to using the preferred separation (ϕ_{pref}) as the initial split. The solid line is obtained using the approximate equations by Glinos and Malone (1984), and the dashed line gives the exact solution obtained from Underwood's method.

for sharp splits, optimal to use the *preferred separation* in the prefractionator. More importantly, the Figure also illustrates that there is a large region of recoveries enveloped by ϕ^{pref} and ϕ^{bal}, in which V remains close to the overall minimum. This is not stressed in any of the previous works, and is of great import for practical operation to be discussed later.

Case 2 : $\phi^{bal} > \phi^{pref}$ **(Figure 11)**
In this case the lower feed controls in the "flat" region between ϕ^{pref} and ϕ^{bal}, and From (33) and (34) the preferred split is *not* optimal if and only if

$$\frac{\alpha_{BC}}{\alpha_{BC}-1} > \frac{z_A - (1 + z_A z_C)}{z_B(\alpha_{AC} - \alpha_{BC})(1 + z_A z_C)} + \frac{(z_B + z_C)(\alpha_{AC} - 1)}{z_B(\alpha_{AC} - \alpha_{BC})(1 + z_A z_C)(\alpha_{BC} - 1)} + 1 \tag{41}$$

In this case it becomes somewhat difficult to extract simple algebraic conditions as was the case for upper feed control, i.e. equation (40). However, after considering numerically a large range of mixtures, we have in fact not found any case for which condition (41) is satisfied. Nevertheless, we find as shown in Figure 11 that for low values of α_{BC} for which ϕ^{bal} is large, there may be very large regions in which V_{min} is relatively constant. Note that we in this case did not find any significant differences between the approximate equations and Underwood's method. One may note, although hardly surprising, that as $\alpha_{BC} \to 1$ we have that $\phi^{bal} \to 1$ and $\phi_{pref} \to 0$, hence the lower feed controls for all recoveries.

Case 3 : $d^{pref} = d^{bal}$ **(Figure 12)**
A special limiting case is when $\phi^{pref} = \phi^{bal}$. Such cases are obtained by equating expressions (9) and (27). It becomes somewhat complicated to derive simple conditions for when this may occur in the general case. However, for cases where the A/B split and B/C split are equally difficult so that $\alpha_{AB} = \alpha_{BC}$ we derive after some algebra that $\phi^{pref} = \phi^{bal}$ if and only if

$$z_A = \frac{1 + z_B(\alpha - 2)}{2} \tag{42}$$

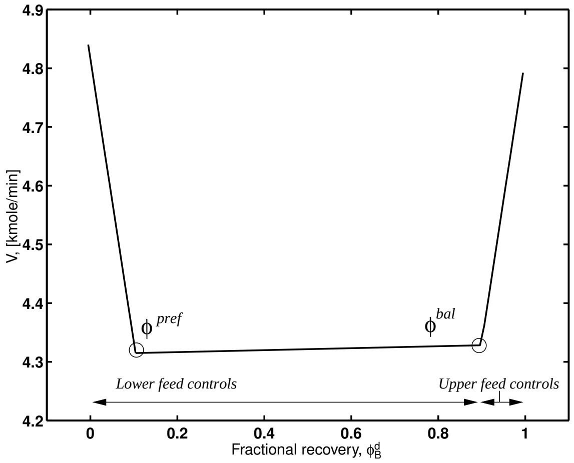


Figure 11: Analytical results for V_{min} as a function of the fractional recover of intermediate ϕ_B^d for $\alpha = 4 : 1.3 : 1$ and $z_F = [0.005, 0.99, 0.005]$. The Figure illustrates a large region enveloped by ϕ_{ref} and ϕ^{bal} where V_{min} stays relatively constant.

Note that equation (42) may not apply for all compositions z_B, since we must require $z_A \leq 1$. For the particular case where $\alpha = 2$ the equality $\phi^{pref} = \phi^{bal}$ occurs always for $z_A = 0.5$. Figure 12 illustrates one such case with a sharp minimum where $\phi^{pref} = \phi^{bal}$. Finally we also recognize that the "flat" regions become smaller if a total condenser is used in the prefractionator ($C1$), since ϕ^{bal} moves closer to ϕ^{pref}. This is because the region in which the upper feed controls becomes larger so that ϕ^{bal} is decreased, i.e. V^{C2} increases with a term proportional to Fz_A.

6 Implications for Operation

We have so far shown in terms of analytical and numerical results that for sharp splits it is optimal to use the preferred separation as the initial split. This preferred separation corresponds to a certain fractional recovery of the intermediate in the distillate from the prefractionator. However, we have also shown that there usually is a range of recoveries between ϕ^{pref} and ϕ^{bal} for which the energy usage (boilup) remains close to the minimum. For practical operation we want to maintain operation in this "flat" region. Again we have three cases

$\phi^{pref} < \phi^{bal}$ In this case we want to maintain $\phi_B^d \geq \phi^{pref}$ ($d \geq d^{pref}$). With reference to Figure 2 we see that to achieve this in the prefractionator, we want to keep the top product (y_C^d) at a given composition, and *overpurify* in the bottoms, i.e. a bottoms product almost completely depleted in the low boiler A. This means that it may be sufficient to use only one point control in the *top* of the prefractionator (i.e. use reflux for control) and set the the boilup in the bottoms at a value which is equal to or higher than the optimum value corresponding to $d = d^{pref}$..

$\phi^{pref} > \phi^{bal}$ This is the reversed case, for which we want to maintain $\phi_B^d \leq \phi^{pref}$ ($d \leq d^{pref}$). This may be achieved by controlling the bottom composition (x_A^b) and *overpurify* in the top of the prefractionator, i.e. a distillate product almost completely depleted in the high boiler C. In this case one may use one point control in the *bottoms* of the prefractionator, e.g. use reflux for control and set the boilup in the bottoms at a value which is equal to or higher than its optimal value corresponding to $d = d^{pref}$.

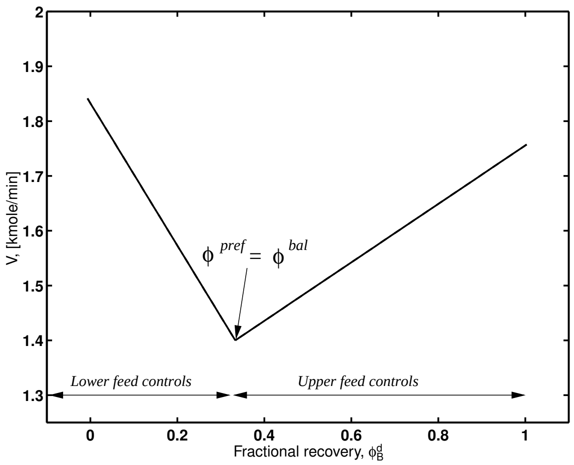


Figure 12: Analytical results for boilup V as a function of the fractional recover of intermediate ϕ_B^d for $\alpha = 4:2:1$ and $z_F = [0.5, 0.1, 0.4]$. The Figure shows that there is a sharp minimum V_{min} where $\phi^{pref} = \phi^{bal}$.

$\phi^{pref} = \phi^{bal}$ In this case V_{min} has a sharp minimum, so there is no "flat" region in which we can operate the column. This case may pose great difficulties for practical operation if one wants to achieve the minimum energy usage. Tight control is most likely needed in both ends of the prefractionator, i.e. use both reflux and boilup for control purposes.

Similar results are expected to hold also for the Petlyuk column, but there the vapor split (R_V) takes the role of the boilup to the prefractionator. These results show the importance of knowing whether ϕ^{pref} is smaller or larger than ϕ^{bal}.

7 Optimal Splits for the Petlyuk Column

As shown in Figure 1 (b) the Petlyuk columns shows a strong resemblance with the prefractionator arrangement studied above. Thus, one may expect that there is a region of recoveries of the intermediate component, approximately between ϕ^{bal} and ϕ^{pref}, for which V_{min} remains almost constant. Indeed, this is the case, and for sharp separations we have in fact a region in which V_{min} is exactly constant. This is shown in the insightful analysis of Fidkowski and Krolikowski (1986).

To complete the foregoing analysis of the prefractionator column we here present the main results from this work. Through a careful analysis of the Underwood equations, Fidkowski and Krolikowski (1986) show that the minimum reflux for a sharp split between components A, B and C is given by

$$\left(\frac{L}{F}\right)_{min} = max\{\frac{z_A\theta_1}{\alpha_{AC}-\theta_1}, \frac{z_A\theta_2}{\alpha_{AC}-\theta_2} + \frac{\alpha_{BC}z_B}{\alpha_{BC}-\theta_2}\} \tag{43}$$

and corresponding for the minimum boilup

$$\left(\frac{V}{F}\right)_{min} = max\{\frac{\alpha_{AC}z_A}{\alpha_{AC}-\theta_1}, \frac{\alpha_{AC}z_A}{\alpha_{AC}-\theta_2} + \frac{\alpha_{BC}z_B}{\alpha_{BC}-\theta_2}\} \tag{44}$$

where θ_1 and θ_2 are the solutions of the Underwood equation (14). These roots may be computed for the *absolute* minimum solution for the prefractionator, which as previously discussed corresponds to the preferred separation. They then carry over to the solutions for the upper and lower part of the main sidestream column. The authors further show that V_{min} is constant between the fractional recoveries ϕ^{pref} given in equation (9) and the recovery ϕ^R given by

$$\phi^R = \frac{L_{min}\left(\alpha_{AC} - \alpha_{BC}\right) - Fz_A\alpha_{BC}}{L_{min}\alpha_{AC} - \left(L_{min} + Fz_A + Fz_C\right)} \tag{45}$$

As noted by Carlberg and Westerberg (1989) this constant minimum reflux *region* is constituted by 4 different sets of specifications for the recoveries and Underwood roots. We thus have that for Petlyuk columns with a sufficiently large number of stages, one may operate that column at any value between ϕ^{pref} and ϕ^R without any increase in the energy usage. For completeness, we also note that Carlberg and Westerberg (1989) extended the analysis for the Petlyuk column also to multicomponent mixtures with an arbitrary number of components.

We may further comment that ϕ^R for the Petlyuk column has the same significance as ϕ^{bal} for the prefractionator arrangement. However we stress that for the latter we found that the minimum energy usage is always smaller for ϕ^{pref} (although only slightly in many cases). We may compare the extent of the "flat" regions in the prefractionator arrangement and the "constant" region of the Petlyuk column. The difference between these depend only on the recoveries of ϕ^{bal} and ϕ^R, since the other limiting value is that of ϕ^{pref} which is the same for both columns. In Table 2 we give values of ϕ^{bal} and ϕ^R obtained from equations (27) and (45) for a feed composition of $z_F = [0.1, 0.8, 0.1]$ and different volatilities. We also give V_{min} for the two column arrangements.

Table 2: Comparison of prefractionator arrangement and Petlyuk column for sharp separations of a ternary mixture with $z_F = [0.1, 0.8, 0.1]$

Volatilities	ϕ^R	ϕ^{bal}	ϕ^{pref}	$V_{min}^{petlyuk}$	$V_{min}^{prefrac}$
$\alpha = 4:2:1$	0.6535	0.6667	0.3333	1.828	1.867
$\alpha = 4:3:1$	0.2691	0.2639	0.6667	2.830	2.867
$\alpha = 4:1.3:1$	0.9038	0.9740	0.1000	3.924	3.967

8 Preferred Separation in Real Columns Using a Finite Number of Stages

The analytical results presented in the previous sections apply to the special case of sharp splits and infinite columns. To verify the analytical results, and to examine the impact of "finite" columns, we now consider numerical simulations for simple, but detailed stage by stage models of distillation columns. The assumptions are the same as for the analytical results; i.e. constant molar flows and constant relative volatilities. Our objective is thus to establish whether using the preferred separation yields the minimum energy inputs for complex columns with a finite number of trays and finite purities. Results are presented both for the prefractionator arrangement and the Petlyuk column in Figure 1.

8.1 Optimal split-sequence for sharp splits

In this section we present numerical results from nonlinear simulations of staged columns assuming constant molar flows and constant relative volatilities. The process data for the simulations are given in Table 3. Here $N_i = 30$ denotes the number of stages in each of the six column sections, giving a total of 180 stages for all arrangements. The product purities

Table 3: Data for ternary separations in real ("finite") column

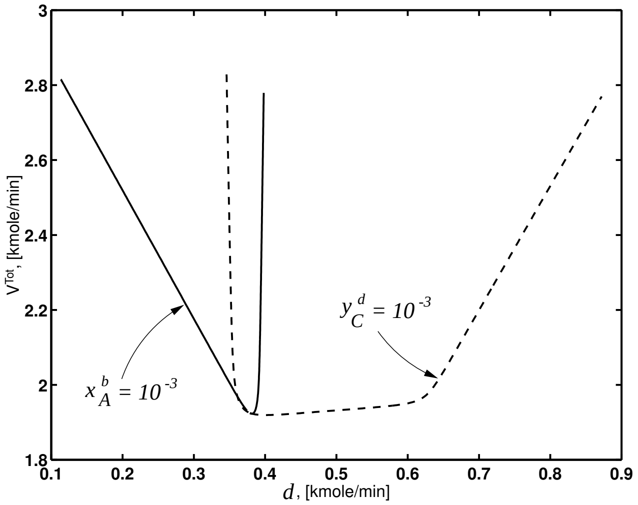
Number of stages	$Ni = 30$
Feed compositions	$z_F = [0.1, 0.8, 0.1]$
Molar feed	$F = 1$ [kmol/min]
Relative volatility	$\alpha = 4 : 2 : 1$
Purity spec.'s	$x_i^P = 99.8\%$ $x_A^b \leq 10^{-3}$ $y_C^d \leq 10^{-3}$

are given by $x_i^P = 99.8\%$ whereas x_A^b and y_C^d denote the purities of A and C in the bottoms and distillate flows from the prefractionator ($C1$), for which we for sharp splits chose the value of $\epsilon = 10^{-3}$. Note that we for all cases plot the boilup versus the distillate flow. This has however no practical implications since there is a unique (linear) mapping from the recoveries to the distillate given by equation (30).

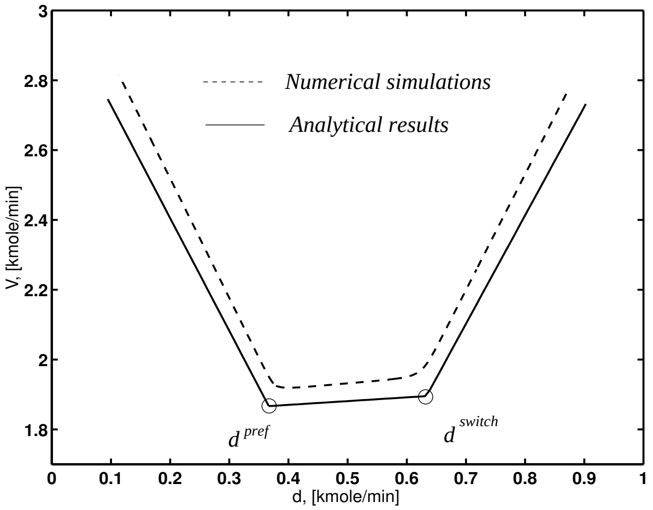
For the arrangements in Figure 1 we have 5 DOFs at steady state. Since three of these are consumed in order to keep the product purities at the respective set points, there is only one DOF left if we are to vary d freely. This last DOF is here used to keep either the purity of A in the bottoms (x_A^b) or C in the distillate (y_C^d) at their set-points of $\epsilon = 10^{-3}$. The procedure for obtaining the desired numerical results can then be outlined as follows. We first compute the minimum boilup using the gradient projection method discussed in (Christiansen 1997), for which we optimize with respect to the two remaining DOFs. Using this optimum as an initial point we then obtained the uniquely defined solution corresponding to $x_A^b = y_C^d = 10^{-3}$, for which all the DOFs are consumed. To determine whether the optimum corresponds to the preferred separation also for (real) columns with a finite number of stages, we then obtained solutions for different values of d by continuation along lines of constant x_A^b and y_C^d. We thus use the value of d corresponding to $x_A^b = y_C^d = 10^{-3}$ in order to switch between the solutions branches. Note also that as for the initial example in Figure 4, the impurity specifications are satisfied as inequalities for all other values of d.

8.2 Prefractionator arrangement

In Figure 13 we give numerical results for the prefractionator arrangement in Figure 1, using the data in Table 3. Figure 13 (a) illustrates an important feature with respect to practical operation. We find that the energy usage stays relatively constant in a region where we keep the composition in the top constant, i.e. $y_C^d = 10^{-3}$. As shown in Figure 13 (b), this region corresponds roughly to the region between the preferred separation (d^{pref}) and the balance main column (d^{bal}). Figure 13 (b) also illustrates the comparison between the numerical results and the analytical results shown in Figure 8. The difference owes mainly to the fact that the simulations correspond to a column with a finite number of stages. An important issue to bear in mind, is that by introducing direct coupling between columns $C2$ and $C3$, we have that only a certain fraction of the impurities that enter from the distillate (C) or bottoms (A) of the prefractionator $C1$ will appear as impurities in the sidestream product B. This situation is different from a prefractionator arrangement with three columns, in which case any impurity either in the top or bottoms from the prefractionator leaves in the intermediate product streams. The impact of this direct coupling becomes even more pronounced as we decrease the *product* purity of the intermediate, and hence allow a larger fraction of impurities to enter from the top and/or bottoms of $C1$. This is treated later when considering non-sharp splits. We now proceed to give results for the Petlyuk column.



(a) Constant y_C^d, x_A^b



(b) Analytical vs. numerical results

Figure 13: Boilup for prefractionator arrangement in Figure 1 as a function of the intermediate distillate flow d with $\alpha = 4:2:1$ and $z_F = [0.1, 0.8, 0.1]$. The solid line in Figure (a) corresponds to $x_A^b = 10^{-3}$ and the dashed line to $y_C^d = 10^{-3}$. Figure (b) gives comparison between numerical and analytical results.

8.3 The Petlyuk column

As previously discussed one may use the Underwood equations to obtain exact analytical solutions for the minimum reflux conditions in Petlyuk columns (Fidkowski and Krolikowski 1986). However, we have not derived any *explicit* analytical results in this paper for the Petlyuk column, although its analogy to the prefractionator is close. We thus expect that the main results for the prefractionator carry over to the Petlyuk column. Note that for the Petlyuk column the *net* distillate flow and fractional recovery are given by

$$d = R_V V - R_L L \tag{46}$$

and

$$\phi_B^d = \frac{R_V V y_B^d - R_L L x_B^{d'}}{F z_B} \tag{47}$$

Here R_V and R_L denote the vapor and liquid split ratio from the main column to the prefractionator, y_B^d the vapor composition of B leaving the prefractionator and $x^{d'}$ the liquid composition entering in the top of the prefractionator. We should comment that ϕ_B^d in this case may extend outside the range $[0, 1]$ and that the (net) distillate flow may become negative. In Figure 14 (a) we show the energy usage V versus d for the Petlyuk column, when using the remaining DOF to fix either the impurity of light in the bottoms (solid line) or heavy in the top (dashed line).

We recognize that instead of consuming the last DOF for purity control, we may of course also use it for optimization purposes. To obtain the "true" optimal solutions for each value of d, we may optimize using for instance the values along constant x_A^b and y_C^d as initial guesses. The optimized curve is given in Figure 14 (b). The results indicate that the overall minimum boilup is constant within a large region of distillate flows, and for this example close to the curve given by $y_C^d = 10^{-3}$. According to the discussion in section 6 this is as expected, since $\phi^R > \phi^{pref}$ so that the lower feed controls. These results thus confirm that the findings of Fidkowski and Krolikowski (1986) easily carry over also to columns with a finite number of stages. We furthermore expect that the opposite situation applies to mixtures for which the upper feed controls. Hence if the upper feed controls, the boilup V should be relatively insensitive to changes in d along the line of constant x_A^b.

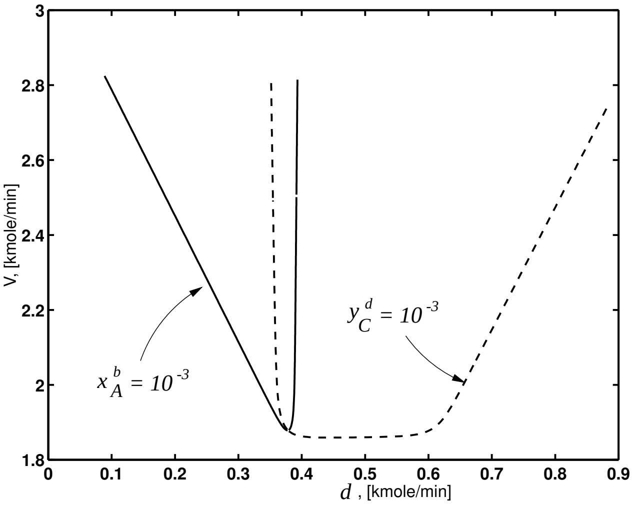
Comparison with results from the literature

We may now compare the results to the analytical results by Fidkowski and Krolikowski (1986). According to the authors V is constant (at least for infinite number of stages) in the region between ϕ^{pref} and ϕ^R. Computing ϕ^{pref} and ϕ^R from equations (9) and (45) we find for a sharp A/C split that they correspond to distillate flows of $d^{pref} = 0.3934$ and $d^R = 0.6228$. Comparing these to the optimized curve in Figure 14, we find excellent agreement which confirms the applicability of the analytical results also to columns with a finite number of stages. Note also that from equation (44) we obtain $V_{min} = 1.83$ and from the simulations we computed $V_{min} = 1.86$.

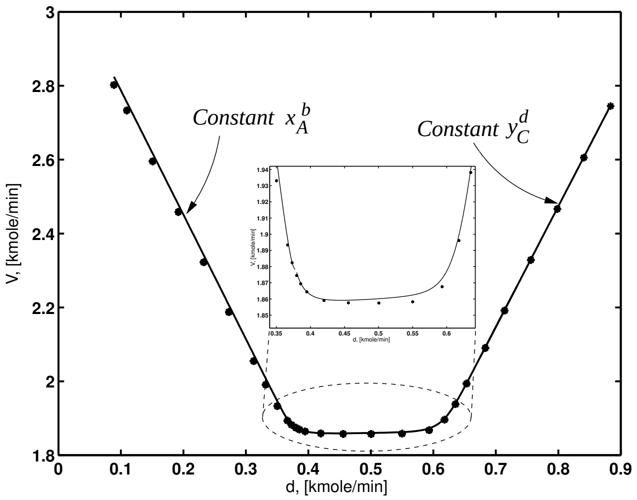
Furthermore we recognize that the regions plotted for constant x_A^b and y_C^d correspond to the qualitatively different regions characterized by Carlberg and Westerberg (1989). For instance we have that the left part in Figure 14, where the light component is kept constant at the upper bound $x_A^b = 10^{-3}$, and the distillate is practically depleted in the heavy component (i.e. $y_C^d \leq 10^{-3}$), corresponds to what Carlberg and Westerberg (1989) denote as region I. Note that keeping x_A^b at the upper bound, implies that the recovery of A in the top is at its lower bound. The other regions correspond to where y_C^d increases and finally reaches its upper bound of $y_C^d = 10^{-3}$.

Implications for operation

The implications for operation of the Petlyuk column are in general the same as for the prefractionator arrangements in section 5.5. Furthermore, since there is a region in which



(a) Constant x_A^b, y_C^d



(b) Optimized V

Figure 14: Boilup for Petlyuk column as a function of the net distillate flow d. The solid line in (a) corresponds to $x_A^b = 10^{-3}$ and the dashed line to $y_C^d = 10^{-3}$. The solid line in (b) represents lines for constant x_A^b and y_C^d and the circles the optimized (minimized) solutions.

V_{min} is constant, one may in fact choose any of the operating points within this region without increasing the energy usage compared to the overall minimum. The Petlyuk column thus proves to be a rather flexible arrangement, in that optimal operation is rather insensitive to changes and disturbances (at least for sharp splits). Although it may appear obvious, we should emphasize that the minimum energy usage is not independent of both DOFs in the prefractionator. Hence it is still required to use (at least) one point control in the prefractionator to maintain operation in the vicinity of the optimum. Furthermore, we acknowledge some important differences between the prefractionator and the Petlyuk column, owing to the direct coupling between the prefractionator and the main (sidestream) column. From a practical point of view it is most likely difficult to use on–line control of the vapor split R_V, so that one may in practice choose to design the column so that R_V during operation is within the optimal region.

Since some means for control in the prefractionator is needed, one may use the liquid split R_L for control. The probably easiest situation in terms of on–line operation, is thus when the lower feed controls, for which one can use R_L to control the impurity of the light component in the top (and overpurify in the bottoms by having R_V sufficiently large). If the upper feed controls, it is somewhat more difficult to control the prefractionator, since one must use the liquid split in the top to control the bottoms composition.

8.4 Optimal split-sequence for non-sharp splits

To investigate the impact of non-sharp separations on the optimal split–sequence, we present in this section numerical results for cases when we decrease the *product* purity of the intermediate component B. Thus, we examine the impact of the separation in the downstream (main) column on the optimal split for the prefractionator. This is in general a problem that does not have a unique solution, since we have two DOFs for the prefractionator and may specify any two of the six recoveries (i.e. top and bottom for all three components).

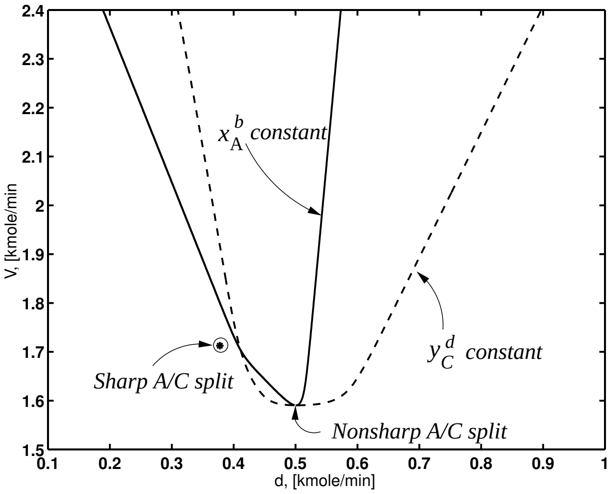
For the numerical examples we use the column data given in Table 3, but now we decrease the product purity of intermediate to $x_B^P = 98\%$. In Figure 15 we show that reducing x_B^P moves the optimum away from the preferred separation. In fact, the overall minimum energy usage does not correspond to using a sharp A/C split in the prefractionator! This is hardly surprising, since reducing the product purity of B allows for a certain amount of impurity to enter over the top and bottom of the prefractionator. It is thus possible to carry out a non-sharp separation in the prefractionator which reduces the required energy input. Importantly, we see that for the Petlyuk column that there is a "constant" region also for non-sharp separations, which is about as large as for the sharp split case. For this example, where the lower feed controls, one may in practice fix the vapor split at the optimal value and use one-point control in the top where the liquid split controls the top composition.

In figure 16 we compare the operating lines for constant y_C^d with the optimized curve, i.e. using the last DOF to minimize V for each value of d.

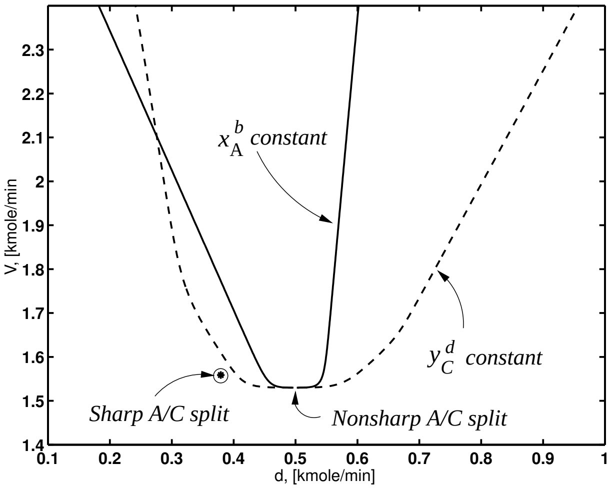
In Table 4 we give results from numerical optimizations using the data in Table 3, where minimum energy inputs are given for different column arrangements and intermediate purities x_B^P. In the table we have in terms of rigor also included data for a heat integrated implementation of a three–column prefractionator arrangement. It is interesting to observe

Table 4: Minimum energy usage for prefractionator arrangements and petlyuk column for different intermediate product purities

	$x_i^P = 99.8\%$	$x_B^P = 98\%$	$x_B^P = 95\%$
Heat integrated	$V_{min} = 1.92$	$V_{min} = 1.64$	$V_{min} = 1.25$
Prefractionator	$V_{min} = 1.91$	$V_{min} = 1.59$	$V_{min} = 1.20$
Petlyuk column	$V_{min} = 1.86$	$V_{min} = 1.53$	$V_{min} = 1.08$



(a) Prefractionator



(b) Petlyuk column

Figure 15: Boilup for prefractionator arrangement (a) and Petlyuk column (b) as a function of the intermediate distillate flow d for non-sharp A/C split and intermediate product purity of $x_B^P 98\%$. The solid lines correspond to $x_A^b = 2 \cdot 10^{-4}$ in Figure (a) and $x_A^b = 2 \cdot 10^{-5}$ in Figure (b). The dashed lines correspond to $y_C^d = 3 \cdot 10^{-2}$ in Figure (a) and $y_C^d = 1.4 \cdot 10^{-2}$ in Figure (b).

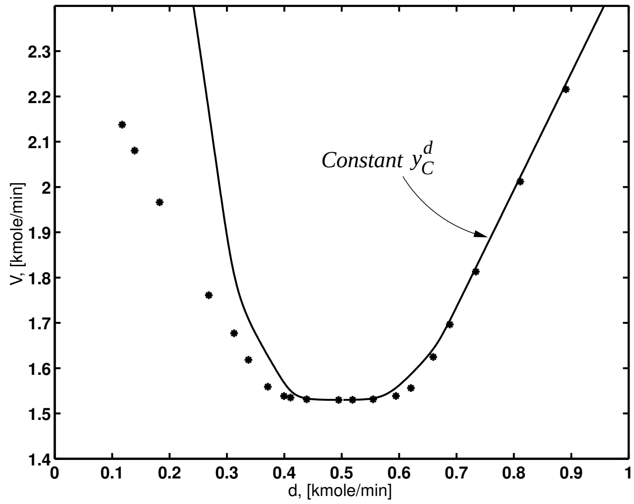


Figure 16: Boilup for Petlyuk column as a function of the intermediate distillate flow d for non-sharp A/C split. The solid lines correspond to controlling the composition in the top(i.e. $y_C^d = 1.4 \cdot 10^{-2}$) whereas the circles illustrate the optimized V for each value of d .

that the savings for the Petlyuk column relative to the prefractionator arrangements increase as the intermediate purity x_B^P is decreased. For $x_B^P = 95\%$ the additional savings are as large as 10%. One may also note that conventional arrangements such as the direct or indirect schemes require, a minimum energy usage of $V_{min} = 2.73$ for *sharp separations* of the given mixture in an infinite column. This value is easily obtained from Underwood's method.

A useful feature of the optimization procedure is that we may obtain the sensitivities for the energy usage with respect to the product purities, by computing the Lagrangian multipliers at the optimal solutions. The details of the calculation procedure is given here in (Christiansen 1997). In table 5 we give Lagrangian multipliers for two product purities for the Petlyuk column. Using the information contained in these multipliers proves very

Table 5: Lagrangian multipliers for the intermediate product purity

$x_i^P = 99.8\%$	$V_{min} = 1.86$	$\lambda = 43$
$x_B^P = 98\%$	$V_{min} = 1.53$	$\lambda = 15$

useful, although we should emphasize that the results in general apply only locally since it is based on approximating the optimal surface as a quadrature. For instance we find that for $x_B^P = 0.98$ $\lambda = 15$. Hence if x_B^P is reduced to $x_B^P = 0.95$, we have that the *predicted* reduction in the energy usage is $\Delta V = 15 \cdot (0.98 - 0.95) = 0.45$. Compared to the results in table 4 we thus find excellent agreement since $\Delta V = 1.53 - 1.08 = 0.45$! For the sharp split case, the results are however not that accurate. This is is however as expected since there is a large *relative* decrease from $x_B^P = 0.998$ $x_B^P = 0.98$. Thus it is correct for smaller reductions in x_B^P.

9 Prefractionator or Petlyuk Column?

We have in this paper discussed some important features of the prefractionator arrangements and the Petlyuk column, which proves useful for both operation and design. We may summarize these findings by comparing certain advantages and drawbacks for the two designs.

1. The Petlyuk column always give a lower V_{min} as shown by Fidkowski and Krolikowski (1990), although the difference is usually small. The lower energy usage owes to supplying all heat (boilup) in the bottoms of the Petlyuk column, and all cooling (reflux) in the top, so that one in fact increases the internal flows in *all sections*. For the prefractionator arrangement a given part of the energy input is "only" used in the prefractionator column.
2. The Petlyuk column has a region between certain recoveries for the intermediate component, given by $\phi_B^d \in [\phi^{pref} \phi^R]$, where V_{min} is constant. The prefractionator on the other hand displays a "flat" region where V_{min} may increase only little for changes in the recoveries in the region $\phi_B^d \in [\phi^{pref} \phi^{bal}]$ where V_{min}. This has important implications for operation, since one may use control in only one end of the prefractionator column and "overpurify" in the other end. The Petlyuk column thus have a slight advantages in terms of flexibility, since one may allow for operation within a certain range of recoveries, without paying a penalty of increased energy input.
3. For operation and control it is also important to recognize that it is probably easier to control *external* flows (i.e. reflux and boilup) rather than manipulating the internal splits (vapor R_V and liquid R_L). For the prefractionator arrangement it thus proves useful to have an external condenser and reboiler compared to the directly coupled flows in the Petlyuk column. The latter may also act in favor of the prefractionator arrangement, since "two–point" control (i.e. in both column ends) is easier to implement.
4. The Petlyuk column is in general more favorable with respect to energy loads ("first law effects") than energy levels ("second law effects"). Since the Petlyuk column consumes all heat at the highest temperature (reboiler) and all cooling at the lowest temperature (bottoms), the utilities may be more expensive than for the prefractionator arrangement where some of the boilup and reflux is supplied at less "extreme" levels.
5. For retrofit and revamping studies, where columns and heat exchangers often are available on cite, the prefractionator arrangement may have some advantages. This owes also to the possibility for using heat integration of the intermediate utility (prefractionator column), which is not possible for the fully integrated Petlyuk column. Hence if utility from some other process stream is available at the level required for the prefractionator column, the "overall" energy consumption may in fact be smaller than the Petlyuk column. One may also operate the two columns in the prefractionator arrangement under different pressure, so as to take advantage of different utility levels on a large scale.
6. An issue that favors the Petlyuk column is that it may be implemented in a single shell using a dividing wall as suggested by Wright (1949). Thus one may also considerably reduce the capital costs and the literature indicates savings in the order of 30% (Smith 1995). However, for such dividing wall columns the aspects of operation and control may become even more crucial. Recent industrial practice however indicates that these are issue which may be resolved.
7. Finally we give a comment on the claim by some authors (e.g. Carlberg and Westerberg (1989)) that the Petlyuk column is only favored when the temperature difference between the heat sources and sinks are large. Although this argument based on "second law effects" (levels) certainly applies, one should at the same time recognize that the "first law" savings (loads) for the Petlyuk column is the largest when the relative volatilities are small, i.e. the temperature differences are small. Hence it is important to always keep in mind this important trade off. However, for close boiling mixtures we also acknowledge that a very large number of stages is required for the Petlyuk column, so that the pressure drop should also be taken into account.

10 Discussion and Conclusions

In this paper we have proposed analytic expressions to obtain the minimum energy usage (V_{min}) for prefractionator arrangements. These expressions allows one to obtain V_{min} analytically for any split of the intermediate component in the prefractionator. Furthermore we have addressed the issue of using the preferred separation as the initial split for multicomponent separations. We have shown that for sharp splits of ideal mixtures, the preferred separation yields the true overall V_{min}. An equally important observation in terms of implications for practical operation, is that there may exist a (large) region of splits in the prefractionator for which V_{min} stays relatively constant. Interestingly we find that using the idea of the preferred separation (Stichlmair 1988) suggest operating the column at one end in this "constant" region, whereas the idea of balancing the main column (Triantafyllou and Smith 1992), suggest operation in the other end. In practical operation it is however usually best to use an intermediate value, because column operation is then relative insensitive to changes.

To verify the significance of the analytical results, we also studied the importance of using the preferred separation for columns with a finite number of stages and for non-sharp separations. For sharp splits we found good agreement between the analytical and numerical results. Results from numerical simulations also indicate that using direct coupling between column sections introduces a degree of flexibility in the column, which proves favorable in terms of operation. For the prefractionator arrangement we find that there is a region of recoveries for which the minimum energy usage stays relatively constant, i.e. there is a "flat region". For the Petlyuk column there is similarly a region where the minimum energy in fact stays constant. This constant region has been showed in the literature to hold for infinite columns and sharp splits, and in this paper we present numerical simulations that shows that it holds also for Petlyuk columns with a finite number of stages and non-sharp separations. Numerical results are also presented which indicate that the fractional savings of the Petlyuk column in fact increases as the purity of the intermediate product is decreased.

Based on the results presented in this paper, we find that in order to maintain operation in the vicinity of the optimum, it may for some cases suffice to use only "one-point control" in the prefractionator. This means that one may overpurify in one end of the prefractionator, and control the composition in the other end to keep this at its optimum value. For the Petlyuk column we may for instance fix the vapor split R_V and use the liquid split R_L to control either the heavy impurity in the top of the prefractionator or the light impurity in the bottom depending on whether the upper or lower feed controls. This finding is supported by simulations where we find that the energy surface is "flat" in certain regions, within which optimal operation should take place.

In order to project our results onto possible directions for future research, we believe that the results presented in this paper may be quite easily extended to multicomponent mixtures of more than three components. In particular we believe that the concepts of the preferred separation and balancing sidestream columns should prove to be very useful in the analysis of other complex distillation arrangements. We expect that using the concept of the preferred separation, which gives the optimal distribution of intermediate components for a pseudo-binary split (for the Petlyuk column it gives *one* of the optimal solutions), may be used to decompose a multicomponent separation to that of a sequence of pseudo-binary splits.

Nomenclature

A, B, C - Component indices

D - Notation for direct split

d - Distillate flow rate [kmol/min]

f - Parameter in minimum reflux expression, i.e. $f = 1 + z_B/100$
F - Feed flow rate [kmol/min]
I - Notation for indirect split
L - Reflux flow rate [kmol/min]
N - Number of theoretical stages
q - Feed enthalpy
R_L - Vapor split fraction in Petlyuk column
R_V - Liquid split fraction in Petlyuk column
S - Separation factor
t - Time [min]
V - Boilup from reboiler [kmol/min]
x_i - Liquid mole fraction of component i
x_i^P - Product composition of component i
y_i - Vapor mole fraction of component i
z_i - Mole fraction of component i in feed

Greek letters
α_{ij} Relative volatility between components i and j
Δ - Deviation variable
ϵ - Upper bound on impurity mole fractions
λ - Lagrangian multiplier
∂ - Derivatives
ϕ_i^d - Fractional recovery of component i in distillate
ϕ^{pref} - Fractional recovery of component B for the preferred separation
ϕ^{bal} - Fractional recovery of component B for balanced main column
θ_i - ith root of Underwood equation

Sub– and superscripts
bal - Balanced column
$main$ - Main sidestream column in prefractionator arrangements
F - Feed stage
min - Minimum flow conditions for infinite number of stages
$pref$ - Preferred separation
P - Product

References

Carlberg, N. A. and A. W. Westerberg (1989). Temperature-Heat Diagrams for Complex Columns. 3. Underwood's Method for the Petlyuk Configuration. *Ind. Chem. Eng. Res.* **28**(9), 1386–1397.

Cerda, J. and A. Wersterberg (1981). Shortcut Methods for Complex Distillation Columns. 1. Minimum Reflux. *Ind. Chem. Eng. Process Des. Dev.* **20**(3), 546–557.

Christiansen, A. C. (1997). Studies on Optimal Design and Operation of Integrated Distillation Columns. PhD thesis. The Norwegian University of Science and Technology.

Fidkowski, Z. and L. Krolikowski (1986). Thermally Coupled system of Distillation Columns: Optimization Procedure. *AIChE journal* **32**(4), 537–546.

Fidkowski, Z. and L. Krolikowski (1990). Energy requirements of nonconventional distillation systems. *AIChE Journal* **36**(8), 1275–1278.

Glinos, K. and M.F. Malone (1984). Minimum Reflux, Product Distribution, and Lumping Rules for Multicomponent Distillation. *Ind. Eng. Chem. Process Des. Dev.* pp. 764–768.

Glinos, K. and M.F. Malone (1988). Optimality Regions for Complex Column Alternatives in Distillation Systems. *Chem. Eng. Res. Des.* **66**(3), 229–240.

King, C. J. (1971). *Separation Processes.* McGraw-Hill Chemical Engineering Series. McGraw-Hill Book Company.

Morud, J. (1995). Studies on the Dynamics and Operation of Integrated Processes. PhD thesis. University of Trondheim. The Norwegian Institute of Technology. Norway.

Nikolaides, I. P. and M. F. Malone (1988). Approximate Design and Optimization of a Thermodynamically Coupled Distillation with Prefractionaton. *Ind. Chem. Eng. Res.* **27**(5), 811–818.

Petlyuk, F. B. and V. M. Platonov (1964). Thermodynamically Reversible Multicomponent Distillation. *Khim. Prom.* (10), 723.

Petlyuk, F. B., V. M. Platonov and D. M. Slavinskij (1965). Thermodynamically Optimal Method for Separating Multicomponent Mixtures. *Int. Chem. Eng.* **5**(3), 555–561.

Petlyuk, F. B., V. M. Platonov and V. S. Avetlyan (1966). Optimum Arrangements in the Fractionating Distillation of Multicomponent Mixtures. *Khim. Prom.* **42**(11), 865.

Smith, R. (1995). *Chemical Process Design.* Wiley.

Stichlmair, J. (1988). Distillation and rectification. *Ullmann's Encyclopedia of Industrial Chemistry* **B3**, 4–1 – 4–94.

Stupin, W. J. (1970). The Separation of Multicomponent Mixtures in Thermally Coupled Distillation Systems. PhD thesis. University of Southern California, USA.

Triantafyllou, C. and R. Smith (1992). The Design and Optimization of Fully Thermally Coupled Distillation Columns. *Trans. Inst. Chem. Eng.* **70**(Part A), 118–132.

Underwood, A. J. V. (1948). Fractional Distillation of Multicomponent Mixtures. *Chem. Eng. Prog.* **44**(8), 603–614.

Wolff, E. A. and S. Skogestad (1995). Operation of Integrated Three–Product (Petlyuk) Distillation Columns. *Ind. Chem. Eng. Res.* **34**(6), 2094–2103.

Wright, R. O. (1949). U.S. Patent 2,471,134.

A Fractional Recoveries for the "Preferred" Separation

The underlying assumption in the expressions for minimum reflux presented by King (1971) and Stichlmair (1988), is the occurrence of a pinch at the feed point for *all* components in a multicomponent mixture. In the main body of the paper we showed how one may find expressions for cases where the feed is saturated liquid. We here show how to derive similar expressions for *saturated vapor feeds.*

A.1 Preferred separation for saturated liquid and vapor feeds

Assuming that all components pinch at the feed point under limiting flow conditions (i.e. minimum reflux), we find using the component balances around each stage that the minimum reflux ratio is given by

$$\left(\frac{L}{D}\right)_{min} = \frac{x_{Di} - y_{Fi}}{y_{Fi} - z_i} \tag{48}$$

This can easily be illustrated from McCabe–Thiele diagrams for each component. For a *saturated vapor feed* we have from (King 1971)

$$\left(\frac{V}{F}\right)_{min} = \frac{\alpha_{LH}\phi_L^d - \phi_H^d}{\alpha_{LH} - 1} \tag{49}$$

If we then equate expression (49) twice for the sharp split between components A and C ($\phi_A^d = 1$ and $\phi_C^d = 0$), and the corresponding split between A and B, we obtain

$$\frac{\alpha_{AC}}{\alpha_{AC} - 1} = \frac{\alpha_{AB} - \phi_B^d}{\alpha_{AB} - 1} \tag{50}$$

which gives

$$\phi_{vapor}^{pref} = \frac{\alpha_{AC} - \alpha_{AB}}{\alpha_{AC} - 1} \tag{51}$$

If we assume that all components pinch at the feed point also for *non-sharp* separations, we may use the formulas given above also for this case. For saturated liquid feeds and non-sharp separations we thus have (King 1971)

$$\left(\frac{L}{F}\right)_{min} = \frac{\phi_A^d - \alpha_{AB}\phi_B^d}{\alpha_{AB} - 1} = \frac{\phi_A^d - \alpha_{AC}\phi_C^d}{\alpha_{AC} - 1} \tag{52}$$

Given the recoveries of A and C, we may thus obtain the fractional recovery of B *exact* for the preferred separation also when the purities are not high, i.e.

$$\phi_{liquid}^{pref} = \frac{\phi_A^d(\alpha_{BC} - 1) + \alpha_{BC}\phi_C^d(\alpha_{AB} - 1)}{\alpha_{AC} - 1} \tag{53}$$

For vapor feeds use equation (49) and obtain

$$\phi_{vapor}^{pref} = \frac{\phi_A^d(\alpha_{AC} - \alpha_{AB}) + \phi_C^d(\alpha_{AB} - 1)}{\alpha_{AC} - 1} \tag{54}$$

These equations then reduce to (9) and (51) in the special case of sharp splits.