

Chapter 6

Minimum Energy Consumption in Multicomponent Distillation

In the evaluation of minimum energy consumption (1st law) we here also discuss minimum entropy production, or lost work (2nd law). This leads us to the reversible Petlyuk arrangement. However the total required heat supply is higher in this case than for the typical (adiabatic) Petlyuk arrangements, but there is a potential for further reduction by use of internal heat integration. This principle can also be applied to general arrangements (not only reversible) and we compare set of alternative distillation arrangements for a given feed example. One interesting result is that it is possible to go below the minimum energy, as given for the extended Petlyuk arrangement presented in Chapter 5, by use of internal heat integration.

We also conjecture that the generalized extended adiabatic (not reversible) Petlyuk arrangement require less energy than any other adiabatic arrangement at constant pressure and without internal heat integration.

6.1 Introduction

In this chapter we use the extended Petlyuk arrangement presented in Chapter 5 as a basis and discuss methods for further reduction of the energy consumption in multicomponent distillation. Some important questions are: Is the directly coupled Petlyuk arrangement always the best? Is it possible to find an ultimate target for minimum energy? Can reversible distillation give further energy reductions? What about internal heat exchange and operation at several pressure levels?

We here define “best” by the following two measures:

1. Minimum Energy (first law efficiency)
2. Minimum Entropy Production (second law efficiency)

The surrounding plant, environmental issues, capital and energy costs have strong influence on the importance of each of these measures to the overall economy. However, we will not consider any economic measure, but present some theoretical distillation arrangements and show by some examples how we can make arrangements, which approach the “best” in both the above senses. We will present the entropy calculations in more detail below.

In section 6.2 we show that for arrangements with adiabatic column sections and no internal heat exchange, the directly (fully thermally) coupled arrangements require less energy than other types of adiabatic column integration. We do not discuss the entropy production in this section, so this can be seen as an introduction and stating of the minimum energy level for a large class of distillation systems.

The reversible Petlyuk arrangement (Petlyuk 1965) plays an important role in this paper, and we show how to compute compositions and vapour flows in section 6.4. The basic theory of reversible distillation is included in Appendix Section 6.10.

In Section 6.5 we compare set of alternative distillation arrangements for a given feed example. One interesting result is that it is possible to go below the minimum energy, as given for the extended Petlyuk arrangement presented in Chapter 5, by use of internal heat integration.

We also briefly discuss operation at several pressure levels in section 6.6.

6.1.1 Some Terms

We first define some important terms used in this chapter. A column *section* is a set of connected *equilibrium stages* and we will mainly discuss sections with infinite number of stages. We introduce the term *adiabatic column section*, as used by Petlyuk et. al. (1964), to denote a column section with constant molar flows and no heat exchange along the section. Thus, the directly coupled columns pre-

sented in Chapter 4 and 5 and typical conventional arrangements contain adiabatic sections. In *non-adiabatic sections* we can supply or remove heat continuously at any stage in the section. A *reversible section* is an infinite non-adiabatic section where the heat is supplied or removed in a way that eliminates mixing irreversibility between neighbouring stages. In a column *arrangement* we put together a number of sections, reboilers and condensers to perform a certain separation task. Sections may be *directly (fully thermally) coupled*, by two-way liquid and vapour streams or may be coupled via condensers or reboilers. We also divide sections at stages with feed streams or side-draw streams. The *Minimum energy* is the minimum required external heat supply to reboilers and to non-adiabatic column sections in order to carry out the specified separation task in a given arrangement and is related to the first law of thermodynamics. Sometimes we use the total requirement for *vaporization* or *vapour flow* as measure of the energy requirement.

We will also discuss the *separation work* (exergy) which is related to the second law, where we also consider the temperature levels of the supplied and removed heat in the system. Reversible distillation requires minimum separation work.

6.1.2 Basic Assumptions

We make the following basic assumptions, which are used throughout this paper:

1. Ideal mixtures with constant relative volatility (α) and constant molar flows (in adiabatic sections)
2. Constant and equal heat of vaporization (λ) for all components
3. Raoult's and Dalton's laws
4. Ideal gas in the vapour phase and negligible liquid volume

In Section 6.10 (Appendix) it is shown that with these assumptions, the temperature-composition-pressure relationship (T - x - P) for a multicomponent mixture (see Petlyuk (1964) for a binary mixture) is given by:

$$\frac{1}{T} = \frac{R}{\lambda} \ln \left(\frac{P_{ref}}{P} \sum_i \alpha_i x_i \right) + \frac{1}{T_{b,r}} \quad (6.1)$$

Here $T_{b,r}$ is the boiling point for the reference component at the reference pressure P_{ref} . However, when considering temperature differences, these constants disappear. The universal gas constant $R=8.31 [JK^{-1}mol^{-1}]$.

In the entropy calculations we assume that the feed and the products are saturated liquids such that the heat supplied equals the heat removed.

6.1.3 Minimum Entropy Production (2nd law efficiency)

The difference between the actual work which can be extracted from a process and the ideal reversible work is the “lost work” given by:

$$W_{lost} = T_0 \Delta S_{total} \quad (6.2)$$

where T_0 is the temperature of the surroundings and ΔS_{total} is the total entropy change (entropy production). The lost work is zero only for a reversible process where $\Delta S_{total} = 0$, but in general we have from the second law of thermodynamics that:

$$\Delta S_{total} = \Delta S_{sur} + \Delta S \geq 0 \quad (6.3)$$

Here, ΔS is the entropy change in the system (e.g. the distillation column) and ΔS_{sur} is the entropy change in the surroundings. Note that ΔS is a state function and is thereby a fixed number for a given separation task. We consider ideal mixtures for which the entropy change when mixing N_c pure compounds at constant pressure and temperature is given by (x denotes mole fraction):

$$\Delta S = -R \sum_{i=1}^{N_c} x_i \ln(x_i) \quad (6.4)$$

However, the entropy change in the surroundings depends on the actual process, and can be calculated from:

$$\Delta S_{sur} = -\oint \frac{dQ}{T} \quad (6.5)$$

where dQ is the actual heat transferred at system temperature T . The integral has to be taken around the system boundaries where heat transfer to the surroundings occur. When the heat is supplied or removed at discrete temperature levels (Q_j at T_j), for example in a reboiler and a condenser of an ordinary distillation column, the integral in (6.5) can be replaced by summation:

$$\Delta S_{sur} = -\sum_j \frac{Q_j}{T_j} \quad (6.6)$$

If we can find a process where the total entropy change (ΔS_{total}) is zero (or $\Delta S_{sur} = -\Delta S$), it is reversible.

A normalized measure of the work loss, or entropy production is given by the *relative entropy production*, defined here as:

$$\frac{\Delta S_{total}}{|\Delta S|} = \frac{\Delta S_{sur} + \Delta S}{|\Delta S|} \quad (6.7)$$

Thus, to check the second law efficiency, we simply need to compute ΔS_{sur} for the actual distillation arrangement. The entropy change ΔS is known from (6.4) when applied to the feed and all the products.

6.1.4 Minimum Energy (1st law)

Even though the net heat supply is zero ($\oint dQ = 0$) it is useful to distinguish between heating ($dQ > 0$) and cooling ($dQ < 0$). We write $dQ = dQ_H + dQ_C$ where the heating $dQ_H = \max(dQ, 0)$ and cooling $dQ_C = \min(dQ, 0)$. Note that the total heat supply equals the total cooling and is given by:

$$Q_H = \oint dQ_H = -Q_C = -\oint dQ_C \quad (6.8)$$

This is a useful measure from an energy point of view (1st law).

Ideally we want to have both ΔS_{total} small (small lost work, i.e. good 2nd law performance) and Q_H small (small total heat supply, i.e. good 1st law performance).

The ideal separation process with a minimum value of Q_H is a reversible process with all the heating at the highest temperature (T_H) and all the cooling at the lowest temperature (T_L). For this process we have from (6.6):

$$\Delta S_{sur} = -\sum \frac{Q_j}{T_j} = -\left(\frac{Q_H}{T_H} - \frac{Q_C}{T_L}\right) = Q_H \left(\frac{1}{T_L} - \frac{1}{T_H}\right) \quad (6.9)$$

Since the process is reversible, we have $\Delta S_{sur} = -\Delta S$, i.e. we have that:

$$Q_{Hmin} = \frac{-\Delta S}{\left(\frac{1}{T_L} - \frac{1}{T_H}\right)} \quad (6.10)$$

The vaporization rate (V) is related to the heat by $Q_H = \lambda V$. For an ideal distillation process with a pure light component at pressure P_L in the top and a pure heavy component in the bottom at pressure P_H , the reciprocal temperature difference is computed by (6.1) and we obtain:

$$\frac{1}{T_L} - \frac{1}{T_H} = \frac{R}{\lambda} \left(\ln \alpha_{LH} + \ln \frac{P_H}{P_L} \right) \quad (6.11)$$

Then the minimum vaporization rate can alternatively be expressed by the relative volatility and pressures:

$$V_{rev,min} = \frac{-\Delta S}{\lambda \left(\frac{1}{T_L} - \frac{1}{T_H} \right)} = \frac{-\Delta S/R}{\ln \alpha_{LH} + \ln \left(\frac{P_H}{P_L} \right)} \quad (6.12)$$

A theoretical reversible process with this behaviour can be obtained by using an ideal heat pump which transforms the required amount of heat from the two extreme temperature levels, to the intermediate temperature levels as required in the reversible distillation column (e.g. in Figure 6.2a at page 186). We could also imagine to adjust the pressure continuously along the column to keep the temperature constant at two levels, but this would also require reversible compression and expansion between stages which is even more “theoretical” than the heat-pump solution.

Any irreversible process that supplies and removes heat at the same temperature levels, e.g. an adiabatic distillation column, will require higher vaporization rate than given by (6.12) according to the second law of thermodynamics.

6.1.5 Summary of some Computation Examples

We show by some numerical examples that it is possible to go below the minimum energy requirement for the typical adiabatic Petlyuk arrangement (to be given in equation 6.13) by use of heat integration, even if we keep constant pressure in the system. In Table 6.1 we have summarized the energy consumption (minimum vapour flow) and the relative entropy production (relative lost work) for some conventional column arrangements and some examples of Petlyuk arrangements with internal heat exchange which are described in Section 6.5. The table is sorted in descending order by the required by external heat supply. Note, however, that the same ordering does not apply to the lost work.

We will in the rest of this paper show in more detail how these results are obtained.

Table 6.1: Comparison of minimum energy (external heat supply) and relative entropy production (lost work) for a set of column arrangements for a given feed

Configuration (Ad: Adiabatic Non: Non-ad.)		External Energy $V_{min} = \Sigma \Delta Q / \lambda$	Relative Entropy Production $\Delta S_{total} / \Delta S $	Comments: (HE: Heat exchange)
Direct Split, no HE (conventional)	Ad	2.072	0.59	C1:A/BC, C2: B/C
Indirect Split, no HE (conventional)	Ad	2.032	1.21	C1:AB/C, C2: A/B
Side Rectifier (directly coupled)	Ad	1.882	0.86	C1:A/BC, C2: B/C1
Side Stripper (directly coupled)	Ad	1.882	1.05	C1:AB/C, C2: A/B Figure 6.1, page 177
Reversible Petlyuk Column	Non	1.667	0.00	Figure 6.2a page 186
Conventional prefrac- tionator arrangement	Ad	1.556	0.63	C1:A/C, C21:A/B, C22: B/C, no HE
Petlyuk Column (typical)	Ad	1.366	0.72	Figure 6.2b page 186 without side-HE
Petlyuk Column + side-HE	Ad	1.366	0.54	Figure 6.2b page 186
Petlyuk + HE across the dividing wall	Ad+ Non	1.222	0.54	Example 2, Section 6.5
Petlyuk + HE from sidestream to feed	Ad	1.181	0.49	Example 3, Section 6.5
Petlyuk + total mid- dle HE	Ad+ Non	1.000	0.26	Example 1a, Section 6.5
Reversible Petlyuk with internal HE	Non	1.000	0.05	Example 1, Section 6.5
Reversible process with only two temper- ature levels	Non	0.793	0.00	Example 0, Section 6.5 Theoretical minimum ref. Section 6.1.4

In the calculation of the numerical values we have assumed constant pressure, and

the following feed data: $F = 1$, $\alpha = [4, 2, 1]$, $z = \left[\frac{1}{3}, \frac{1}{3}, \frac{1}{3} \right]$, $q = 1$.

6.2 The Best Adiabatic Arrangement Without Internal Heat Exchange

Petlyuk (1965) showed that it is possible to devise a reversible Petlyuk arrangement (see Section 6.5) with zero lost separation work and thus requires minimum separation work compared to any other separation process.

However, it has also been conjectured that the adiabatic Petlyuk arrangement, where all the heat is supplied in the bottom reboiler at the maximum temperature, requires minimum energy (V_{min}) compared to any other adiabatic distillation arrangement (without internal heat exchange). However, no proof has been found in the literature (Petlyuk 2000), except for the ternary case. For the ternary case Fidkowski and Krolikowski (1987) showed that the 3-product Petlyuk arrangement always has a smaller vapour flow than any arrangements with side-strippers or side-rectifiers and they showed that these also performed better than the conventional direct and indirect split sequences.

For the generalized adiabatic Petlyuk arrangement presented in Chapter 5, the minimum energy requirement for separation of a feed mixture of N_c components is given by:

$$V_{min}^{Petlyuk} = \max_j \sum_{i=1}^j \left(\frac{\alpha_i z_i F}{\alpha_i - \theta_j} \right), \text{ where } j \in \{1, N_c - 1\} \quad (6.13)$$

where θ_i are the $N_c - 1$ common Underwood roots found from the feed equation (q is liquid fraction in the feed (F) and z is the feed composition):

$$\sum_i \frac{\alpha_i z_i}{\alpha_i - \theta} = 1 - q \quad (6.14)$$

Note that all the heat can be supplied in the bottom reboiler and be removed in the top condenser, but, in some cases, some of the heat may be supplied or removed at the product outlets (Chapter 4 and 5).

In the following we consider adiabatic column sections, and we verify that the adiabatic Petlyuk arrangement is indeed the best distillation arrangement when we regard the total requirement for vaporization at constant pressure, and when we do not consider any internal heat exchange within the arrangement.

6.2.1 Direct Coupling Gives Minimum Vapour Flow

First we will show that the direct (fully thermal) coupling minimises the vapour flow requirement through any column junction.

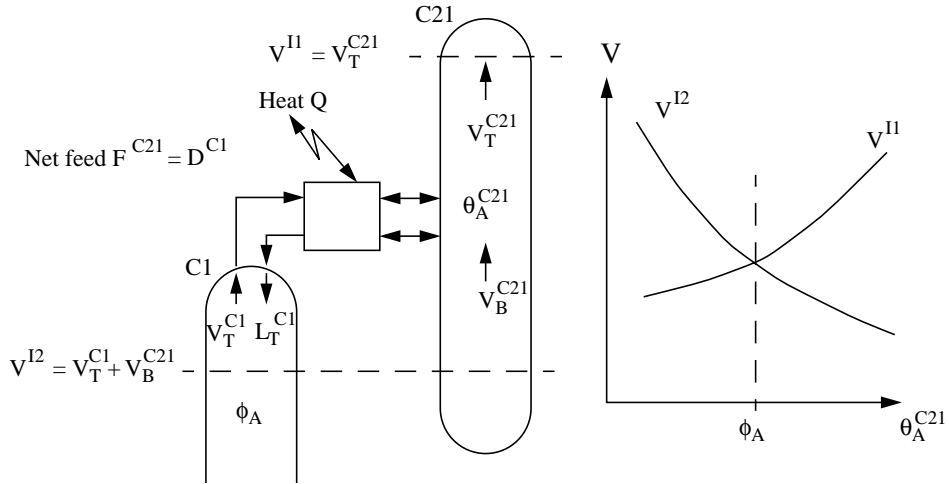


Figure 6.1: General column interconnection junction. The direct (full thermal) coupling gives $\theta_A^{C21} = \phi_A$ which implies $\min(\max(V^{I1}, V^{I2}))$ and a zero external heat exchange at the interconnection ($Q=0$).

Let us consider a general junction at the top of the prefractionator (C1) and the succeeding column (C21) as illustrated in Figure 6.1. To simplify we assume a ternary feed, but similar results can be obtained for any number of components and at any junction in an arrangement.

We assume that the two degrees of freedom in column C1 (e.g. D^{C1} , V_T^{C1}) are fixed. In Chapter 4 we showed that the composition in the recycle flow (L_T^{C1}) from C21 to C1 normally has no effect on the net component flows from C1 to C21. This is so unless a component which would have been removed in an ordinary column (with a condenser) is not introduced in the recycle flow to the directly coupled column. For reasonable operation of the system this will normally not be a problem.

At the interconnection to C21 we allow for supply or removal of heat (still with fixed D^{C1} , V_T^{C1}). This will then only affect the effective liquid fraction (q^{C21}) to column C21 and have no impact on the component flows (w_i^{C1}). Recall that direct coupling implies that the reflux in C1 is taken directly as a side-draw from C21 and that the vapour flow from C1 is fed directly to C21. In this case the external heat exchange is zero, and we obtain an equivalent liquid fraction given by:

$$q_{dc}^{C21} = 1 - V_T^{C1} / D^{C1} \quad (6.15)$$

Note that we always have $q^{C21} < 0$ with direct coupling, which is equivalent to a superheated vapour feed. Heat removal (e.g. a condenser) will increase q^{C21} and heat supply (superheater) will decrease its value.

The most important effect of the direct coupling is that the Underwood roots in the top of C1 “carry over” as the common (minimum energy) Underwood roots for C21 (Carlberg and Westerberg 1989). Thus, $\theta_A^{C21} = \phi_A^{C1}$, which is vital in the following analysis.

For a given operation of the first column (not necessarily at minimum energy), the vapour flow, and net component flows in the top can be related to a certain Underwood root (ϕ), here given by the defining equation in column C1 (we omit the superscript C1 on w and ϕ):

$$V_T^{C1} = \frac{\alpha_A w_A}{\alpha_A - \phi_A} + \frac{\alpha_B w_B}{\alpha_B - \phi_A} + \frac{\alpha_C w_C}{\alpha_C - \phi_A} \quad (\text{note } D^{C1} = \sum w_i) \quad (6.16)$$

Consider now any type of interconnection to the succeeding column (C21). At minimum energy operation in C21 the flow rates are determined by the component distribution and the common Underwood roots. Thus:

$$V_{Tmin}^{C21} = \frac{\alpha_A w_A^{C21}}{\alpha_A - \theta_A^{C21}} + \frac{\alpha_B w_B^{C21}}{\alpha_B - \theta_A^{C21}} + \frac{\alpha_C w_C^{C21}}{\alpha_C - \theta_A^{C21}} \quad (6.17)$$

$$V_{Bmin}^{C21} = \frac{\alpha_A (w_A^{C21} - w_A)}{\alpha_A - \theta_A^{C21}} + \frac{\alpha_B (w_B^{C21} - w_B)}{\alpha_B - \theta_A^{C21}} + \frac{\alpha_C (w_C^{C21} - w_C)}{\alpha_C - \theta_A^{C21}}$$

The common Underwood roots can be found from the feed equation of C21 (6.18) and will depend on the external heat through the feed quality. The net component flow and net distillate flow in C1 are constants.

$$\frac{\alpha_A w_A}{\alpha_A - \theta^{C21}} + \frac{\alpha_B w_B}{\alpha_B - \theta^{C21}} + \frac{\alpha_C w_C}{\alpha_C - \theta^{C21}} = (1 - q^{C21}) D^{C1} \quad (6.18)$$

Note that for any reasonable operation of columns, all net component flows are positive in the top sections and negative in the bottom sections. This implies that the minimum vapour flow in the top section will increase as the common Underwood root increase and the vapour flow in the bottom section will decrease.

In the following we fix the operation of column C1 such that V_T^{C1} and all w_i , and thereby all ϕ_j are constant, and we want to find the value of the common Underwood root in C21 (θ_A^{C21}) which minimize the maximum vapour flow rates through any of the intersections above or below the feed junction (see Figure 6.1):

$$\min_{\theta_A^{C2I}} \left(\max(V^{I1}, V^{I2}) \right) \text{ where} \quad (6.19)$$

$$V^{I1} = V_{Tmin}^{C2I} \text{ and } V^{I2} = V_T^{C1} + V_{Bmin}^{C2I} \quad (6.20)$$

A typical dependency of V^{I1} and V^{I2} as a function of θ_A^{2I} is shown in Figure 6.1, and we see that the analytical solution is given by:

$$\arg(\min_{\theta_A^{C2I}} \left(\max(V^{I1}, V^{I2}) \right)) = \phi_A \quad (6.21)$$

Proof

For normal operating conditions, we have $\frac{dV^{I1}}{d\theta_A^{C2I}} > 0$ and $\frac{dV^{I2}}{d\theta_A^{C2I}} < 0$.

This implies that $\min \left(\max(V^{I1}, V^{I2}) \right)$ is found when $V^{I1} = V^{I2}$.

By applying $\theta_A^{C2I} = \phi_A$ in equations (6.16-6.20) we obtain $V^{I1} = V^{I2}$.
Q.E.D.

In conclusion, minimization of the vapour rate through any intersection (I1 or I2) is found when the common Underwood roots in column C2I equal the actual roots in the top section of C1. This is exactly what we obtain with a direct coupling. Note that the proof does not require the first column to be operated at minimum energy and that it is valid for any distribution of components in C1.

6.2.2 Implications for Side-Strippers and Side-Rectifiers

A direct implication of the result in Section 6.2.1 above is that arrangements with side-strippers (like in Figure 6.1 with a direct coupling) or side-rectifiers, will always have a lower total need for vaporization than the corresponding indirect split or direct split configurations. This was also shown by Fidkowski and Krolkowski (1987) for the ternary case, but it is straightforward to extend the result in Section 6.2.1 to the general multicomponent case.

6.2.3 The Adiabatic Petlyuk Arrangement is Optimal

The result in Section 6.2.1 gives rise to the following conclusion:

We assume constant relative volatilities, constant molar flows, constant pressure and no internal heat integration. Then the generalized adiabatic Petlyuk arrangement has the lowest need for vaporization compared to any other adiabatic distillation arrangement for separation of an arbitrary feed mixture into its pure components when

This result is based on the simple argument that at any junction where we might consider another type of connection than the direct coupling, the required vapour flow through the junction, and thereby through a cross-section of the whole arrangement, will increase.

We have not presented a complete proof, so the above conclusion is a conjecture. However, for the ternary case, it has been proved by Fidkowski and Krolikowski (1987), when considering conventional arrangements and side-strippers as alternative configurations.

A qualitative explanation is that the direct (full thermal) coupling can be regarded as ideal heat integration. For example when a side stripper configuration is used instead of an indirect split configuration, the direct coupling replaces a condenser (which in practice has an inevitable loss). This is probably the background for the term “full thermal coupling” used by many authors. However, here we will use the term “direct coupling” which relates to that both the vapour and liquid flows are coupled directly between two columns. In addition, we obtain reversible mixing at the junctions when we keep the vapour and liquid flows in the junctions at equilibrium.

6.3 Entropy Production in Adiabatic Arrangements

6.3.1 Adiabatic Column (Section)

We consider a column (or just a section) with constant vapour flow (V), $Q_H = \lambda V = -Q_C$ and known composition and pressure in the top (T) and bottom (B). Then equation (6.6) combined with (6.11) gives:

$$\Delta S_{sur} = \lambda V \left(\frac{1}{T_T} - \frac{1}{T_B} \right) = RV \ln \left(\frac{\sum (\alpha_i x_{i,T}) P_B}{\sum (\alpha_i x_{i,B}) P_T} \right) \quad (6.22)$$

Note that this expression is independent of the heat of vaporization, the boiling point temperatures and the absolute pressure. In the case of constant pressure and when all the heat is supplied in the bottom where only the heavy key (H) appear, and is removed in the top where only the light key appear (L), (6.22) is simplified to:

$$\Delta S_{sur} = RV \ln \alpha_{LH} \quad (6.23)$$

(Note that this could be found from equation (6.12) when replacing $V_{rev,min}$ with V and ΔS with $-\Delta S_{sur}$, and constant pressure).

Example: We may apply (6.23) to adiabatic binary distillation. Combined with Kings formula for V_{min} (for $q = 1$) we obtain:

$$\Delta S_{sur} = R \frac{(1 + (\alpha - 1)z)}{\alpha - 1} F \ln \alpha \quad (6.24)$$

For the feed: $F = 1$, $\alpha = 2$, $z = 0.5$, we obtain $\Delta S_{sur} = R \cdot 1.5 \cdot \ln 2$. The actual entropy change $\Delta S = -R \cdot \ln 2$ (6.4), and the relative entropy production $(\Delta S_{sur} + \Delta S)/|\Delta S| = 0.5$.

6.3.2 Adiabatic Petlyuk Arrangements

The entropy production in adiabatic Petlyuk arrangements (see Figure 6.2b at page 186) can be found by the expression for single adiabatic sections. We may use combinations of (6.23) for nonsharp products or (6.23) for sharp product splits. Consider a ternary case (components ABC with relative volatilities $\alpha_C, \alpha_B, \alpha_A$ and saturated liquid feed) where we have sharp product splits and that we have the possibility to change the vapour flow at the sidestream stage (S) (by supply or removal of heat). Then, given by the minimum vapour flows (V) in the reboiler (B) and condenser (T):

$$\begin{aligned} \Delta S_{sur} &= \lambda \left(V_B \left(\frac{1}{T_S} - \frac{1}{T_B} \right) + V_T \left(\frac{1}{T_T} - \frac{1}{T_S} \right) \right) \\ &= \lambda R (V_B \ln \alpha_{BC} + V_T \ln \alpha_{AB}) \end{aligned} \quad (6.25)$$

When the vapour flow is constant through the arrangement we have $V_B = V_T = V$, ($V_{min}^{Petl} = \max(V_{Bmin}, V_{Tmin})$), and from (6.25) or directly from 6.23) we obtain:

$$\Delta S_{sur} = RV \ln \alpha_{AC} \quad (\text{where } \ln \alpha_{AC} = \ln \alpha_{BC} + \ln \alpha_{AB}) \quad (6.26)$$

Note that $\max(V_B, V_T)(\ln \alpha_{BC} + \ln \alpha_{AB}) \geq V_B \ln \alpha_{BC} + V_T \ln \alpha_{AB}$. From this it is simple to see that the entropy production when we have constant vapour flow through the arrangement as given by (6.26) is always larger than the expression in (6.25) where we allow for supply or removal of heat at the sidestream stage.

6.4 Reversible Distillation

Reversible multicomponent distillation have been described by Grunberg (1956), Petlyuk et. al. (1964 and 1965) and Fonyó (1974ab). An overview is also found in the textbook by King (1980). Here we show in detail how to compute flow rates and composition profiles in a reversible arrangement for separation of multicomponent feed, and we will relate this to the adiabatic Petlyuk arrangement.

The sources of irreversibility in a distillation process come from mixing of fluids with different composition and temperature. This may happen at the feed stage, in the top or bottom when fluid is returned to the column, and also inside the column when there is a composition difference between neighbouring stages. In adiabatic distillation, there will always be a set of stages inside the column sections with significant composition differences resulting in irreversible interstage mixing, even with infinite number of stages.

To obtain reversible operation we consider infinite non-adiabatic sections. We assume that at any stage (n), there is a local pinch zone with constant composition (i.e. $x_{n+1} = x_n$) such that we obtain reversible mixing between neighbouring stages. Then the material balance for component (i) at an arbitrary stage is given by:

$$Vy_i - Lx_i = w_i \quad (6.27)$$

Note that the net component flow w_i is always constant inside any type of section. From (6.27) we get the requirement for vapour flow through the stages:

$$\text{Top: } V_T = \frac{w_i - x_i D}{y_i - x_i} \quad \text{Bottom: } V_B = \frac{w_i + x_i B}{y_i - x_i} \quad (6.28)$$

Note that this expression is valid for any component (i). The net product flows are trivially given by $D = \sum w_{i,T}$ and $B = -\sum w_{i,B}$.

A characteristic of the *reversible distillation* column is that some of the heat is supplied continuously along the bottom (stripping) sections and removed along the top (rectifying) sections, as opposed to the conventional adiabatic arrangements, where there are no heat exchange along the column sections.

In Appendix Section 6.10 we present the reversible distillation theory in more detail. We show that when the vapour flow is set according to (6.28), the entropy to the surroundings is described by:

$$dS_{sur} = R \sum_i (w_i d(\ln x_i)) \quad (6.29)$$

For any given reversible arrangement, it is then trivial to show that integration of (6.29) along all the reversible sections gives $\Delta S_{sur} = -\Delta S$ (where ΔS is given by (6.4)). In the appendix, this is shown in detail for a reversible binary column. Below, we treat the reversible Petlyuk arrangement more carefully.

6.4.1 The Reversible Petlyuk Arrangement

In multicomponent distillation, Petlyuk (1965) and Fonyó (1974) list two main properties which limits the possibilities for reversible operation:

1. We cannot remove more than one component in each end of a 2-product column. This is due to the fact that we need a pinch zone across the feed stage, and this is only possible for so-called Class 1 separations (Shiras 1950). If we relate this to the V_{min} -diagram (ref. Chapter 3), reversible operation is only possible at the preferred split, where we remove exactly one feed component in each end, or in the triangle below, where all components may be distributed to both ends.
2. We require that the top reflux is in equilibrium with the vapour flow leaving. This is not possible with an ordinary condenser unless the liquid flow rate is zero. Similarly the vapour flow into the bottom must be in equilibrium with the liquid flow out. This cannot be done reversibly in a single condenser or reboiler in the case of more than 2 components.

Note that both these limitations can be overcome with a Petlyuk arrangement. We already have reversible mixing in all junctions and by using infinite reversible sections, the whole arrangement becomes reversible. In the following we will present a procedure for computing reversible heating or cooling along the sections, and the corresponding composition profiles. Figure 6.2a illustrates the results for a given ternary example.

The vapour and liquid rates through the feed stage can be found by assuming a pinch at the feed stage, for example by applying Underwood's (1948) equations for minimum reflux for the stages immediately above and below the feed. The recoveries (or net component flows w_i) of all components are completely determined when we specify two independent variables at the feed stage.

Away from the feed stage we can express the compositions and flow rates as a function of a single free variable, e.g. the composition of the component which is to be removed in that section. (This can be shown by inspecting the equations involved). When we remove the most extreme volatile component (k) in a section (its composition has to approach zero before the end and the material flow $w_k=0$) we may apply equation (6.28/6.60) for $i=k$. Then the following expression applies for both the top and the bottom section (with different components k and net flows w_i , of course):

$$V(x) = -\sum_i w_i / \left(\frac{\alpha_k}{\sum_i \alpha_i x_i} - I \right) \quad (\text{note } D = \sum_i w_{i,T} \text{ and } B = -\sum_i w_{i,B}) \quad (6.30)$$

Interestingly, the composition of an arbitrary component (i) on a certain stage depends linearly on the mole fraction of the component (k), which is to be removed, and its mole fraction at the feed stage of that column.

$$x_i = x_{i,F} \left(\frac{I - x_k}{I - x_{k,F}} \right), \text{ for } i \neq k \quad (6.31)$$

Here k refers to the least volatile component in the top section, and the most volatile component in a bottom section. Equation (6.31) can be proved by inserting (6.31) into (6.30), and verifying that the material balance (6.27/6.58) is fulfilled for all component flows.

Now we extend this to compute flow rates and compositions in the whole arrangement. The compositions in all feed junctions are found by applying (6.31) recursively for every section. The limiting pinch composition in the top of the first column (the prefractionator), when the heavy component ($i=H$) is removed, can be found by setting $x_H=0$.

$$x_{i,PT}^{CI} = \frac{x_{i,F}}{I - x_{H,F}} \quad (6.32)$$

Similarly, when $i=L$ denotes the light component, the composition in the liquid leaving the bottom (where $w_L = 0, x_L = 0$) becomes:

$$x_{i,PB}^{CI} = \frac{x_{i,F}}{I - x_{L,F}} \quad (6.33)$$

When the direct (fully thermal) coupling is between pinch zones at the same composition, the mixing will be reversible. Then the “pinch”-compositions given by (6.32 and 6.33) express the “feed-stage” composition in the succeeding columns and we simply reuse the same equations for that column, but with one component removed from its feed. Going through the whole arrangement, we find that at an arbitrary junction (column Cx) the “feed composition” is uniquely determined by the first feed stage composition (x_F), and the range of components present at the junction ($L \leq i \leq H$).

$$x_{i,F}^{Cx} = \frac{x_{i,F}}{H \sum_{j=L} x_{j,F}}, \text{ and } x_{i,F}^{Cx} = 0 \text{ for } i < L \text{ and } i > H \quad (6.34)$$

Example: With a ternary saturated liquid feed (components ABC)

$$x_F = z \text{ and (6.34) gives: } x_{A,F}^{C21} = \frac{z_A}{z_A + z_B}, \quad x_{B,F}^{C22} = \frac{z_B}{z_B + z_C}$$

The net component flows at the feed stages in each column can again be computed by using the preferred split in every column inside the arrangement.

The entropy change to the surroundings is given by equation (6.29/6.62) for each section. For our ternary example shown in Figure 6.2a, we may write the total entropy change as:

$$\Delta S_{sur} = \Delta S_{sur}^{C1T} + \Delta S_{sur}^{C1B} + \Delta S_{sur}^{C21T} + \Delta S_{sur}^{C21T} + \Delta S_{sur}^{C22T} + \Delta S_{sur}^{C22T} \quad (6.35)$$

Since we have pinch in the junction regions, the terms involving any intermediate junction composition will cancel out due to the material balance for each net component in the junction. For example the terms involving the contribution from the light component at the feed junction at C21 are given as:

$$w_{A,T}^{C1} \ln x_{A,T}^{C1} + (w_{A,B}^{C21} - w_{A,T}^{C21}) \ln x_{A,F}^{C21} = (w_{A,T}^{C1} + w_{A,B}^{C21} - w_{A,T}^{C21}) \ln x_{A,F}^{C21} = 0$$

Thus, the total entropy change in the surroundings will depend only on the product and feed compositions and we obtain $\Delta S_{sur} = -\Delta S$, which proves that we have a reversible process.

The solution for a ternary case is illustrated in Figure 6.2a. Numerical values are shown for the compositions and flow rates in the end of every section.

The solution procedure is summarized below:

- The net component flows are computed for the prefractionator (C1) feed by specifying a sharp A/C split and pinch across the feed stage (preferred split) (ref. Chapter 4).
- The net component flow rates in columns C21 and C22 are trivial since we require pure products, and the junction feed flows are given from C1.
- All junction compositions are computed from (6.34)
- Vapour flow in each end is computed from (6.30)

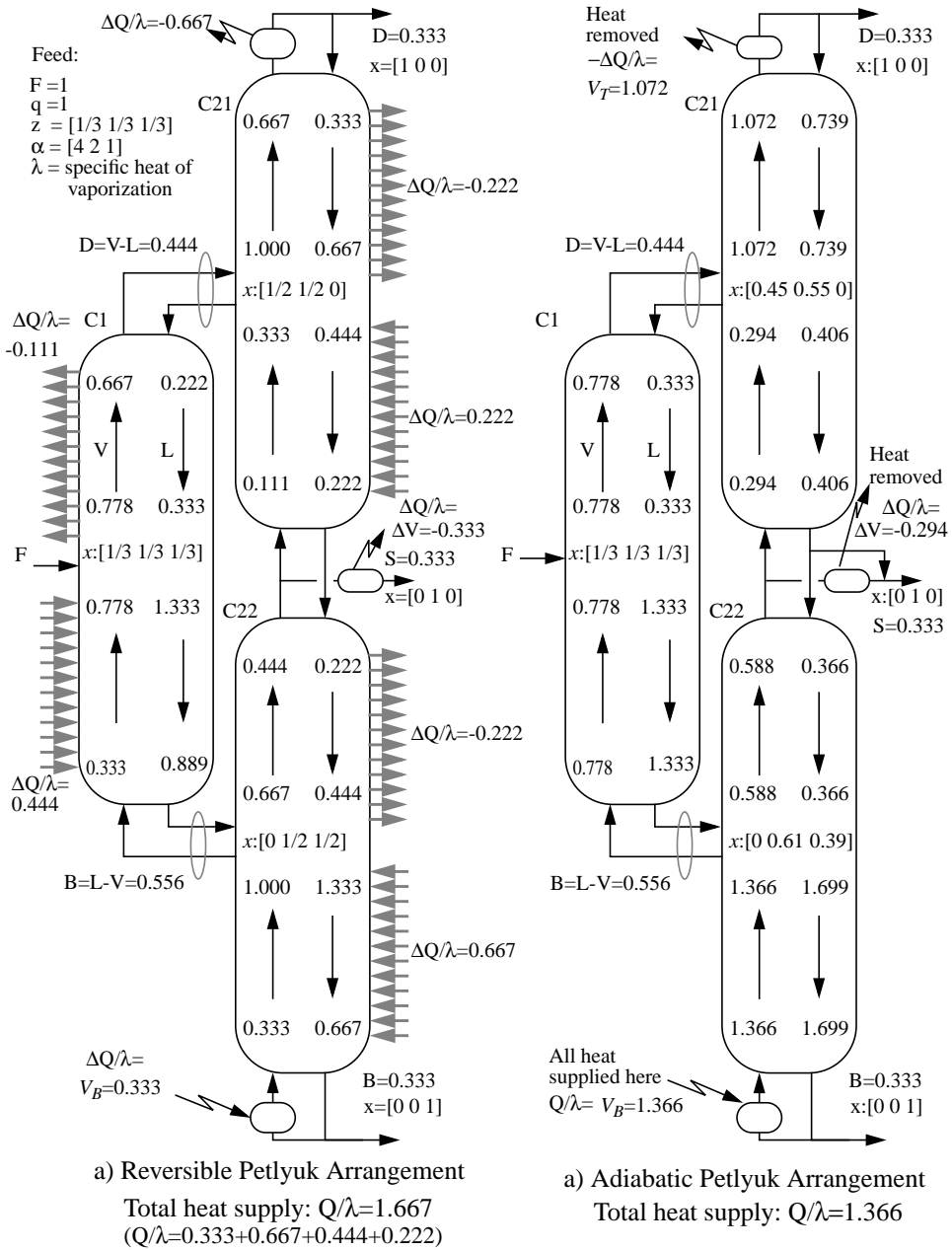


Figure 6.2: The reversible Petlyuk arrangement (a), has heating and cooling along the sections. In the (irreversible) adiabatic Petlyuk arrangement (b), all heat is supplied in the bottom reboiler. Minimum required internal flow rates and the resulting junction compositions (x) are shown for the given feed. (The heat removal at the side-stage for the adiabatic arrangement (b) is optional).

6.4.2 Comparing Reversible and Adiabatic Arrangements

The internal flows and pinch zone compositions for the adiabatic Petlyuk arrangement shown in Figure 6.2b are computed by the methods presented in Chapters 3 and 4. The composition profile in the prefractionators are shown in Figure 6.3. Recall from Chapter 3 that the adiabatic profile also follow straight lines from the feed to the pinch composition at the ends in the triangular diagram.

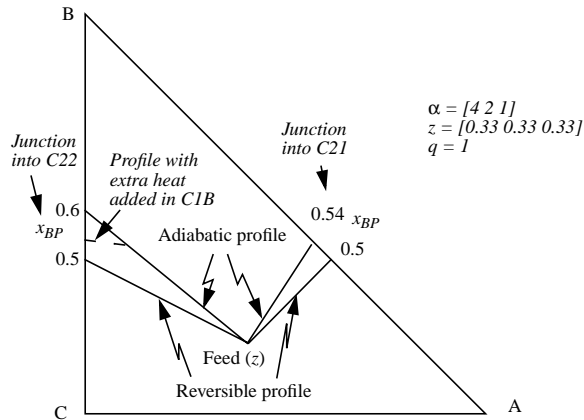


Figure 6.3: Composition profiles in the prefractionators (C1) for the adiabatic and reversible arrangements shown in Figure 6.2.

However, in the reversible arrangement the pinch composition at each end of C1 results in a lower vapour flow requirement in the succeeding columns, compared to the adiabatic arrangement. This is easy to see qualitatively: In C22 the mole fraction of C is increased at its feed stage, leading to a lower boilup ratio, and similarly in C21 the mole fraction of the light A is increased at the feed stage, leading to a lower reflux ratio.

Note that even if the reversible arrangement in Figure 6.2a is thermodynamically optimal with respect to minimising the lost work, the numerical example shows that the total heat supply is higher than for the (irreversible) adiabatic arrangement in Figure 6.2b.

The most important similarities and differences between the reversible and the adiabatic arrangements are listed below:

- The flows through the prefractionator feed stage and the recoveries of feed components from the prefractionators are identical.
- The pinch zone compositions at the ends of the prefractionator differ and result in a lower requirement for energy in the succeeding columns for the reversible prefractionator.
- The total amount of required heat supply for vaporization is higher in the given reversible arrangement for our example.

6.5 A Case Study: Petlyuk Arrangements with Internal Heat Exchange

We here show by a set of numerical examples that by use of internal heat integration it is possible to reduce the external heat supply requirement below the well known minimum vapour flow values given by Fidkowski and Krolikowski (1986) for the ternary case, and generalized in equation (6.13) (Chapter 5) to any multi-component case.

The basis for this possibility is that the temperature range in some of the column sections overlaps, and some of the removed heat can be utilized as heat supply to other sections and in that way reduce the need for external heat supply.

The examples are based on the ternary case in Figure 6.2. For all cases we have:

$$\Delta S = -R \sum z_i \ln z_i = -8.31 \cdot \frac{3}{3} \ln \frac{1}{3} = -9.13 [JK^{-1} mol^{-1}] \quad (6.36)$$

6.5.1 Example 0: Theoretical Minimum Energy Limit

Recall that the absolute minimum heat supply in a reversible process is when all the heat is supplied at the highest temperature, and is removed at the lowest temperature as shown in (6.12). For the given feed ($F = 1$), the correspondingly minimum need for vaporization by external heat supply in a hypothetical reversible “distillation” process is:

$$V_{rev,min} = -\frac{\sum z_i \ln z_i}{\ln \alpha_{LH}} = -\frac{(3/3) \ln 1/3}{\ln 4} = 0.7925 \quad (6.37)$$

6.5.2 Example 1: Internal Heat Exchange in the Reversible Arrangement

The total need for heat supply for vaporization in all three bottom sections of the reversible arrangement in Figure 6.2a, is given by:

$$\frac{Q_v}{\lambda} = V_{BF}^{C22} + \Delta V^{C1B} + \Delta V^{C21B} = 1 + \frac{4}{9} + \frac{2}{9} = \frac{15}{9} = 1.667 \quad (6.38)$$

However, by careful inspection of the available heat from cooling ($dQ_c(T)$) and required heating ($dQ_H(T)$) at each temperature (T) for the given case we find that the heat required in the middle sections C1B and C21B can be supplied by the heat removed from C22T and C1T and the sidestream condenser (the details of this procedure is not shown, but ordinary Temperature-Heat diagrams can be

used). Thus, in theory, minimum heat supply, when the possibilities for internal heat exchange case is utilized, is given by the requirement for heat supply in C22B only: $V_{BF}^{C22} = 1.0$. We may also calculate the entropy to the surroundings by (6.29) for sections C22B and C21T and along the middle sections, where the external $dQ=0$, we use (6.5 or 6.50) directly (this part yields the two first terms):

$$\Delta S_{sur}' = \frac{V_{TF}^{C21}}{T_F^{C21}} - \frac{V_{BF}^{C22}}{T_F^{C22}} - R(w_{C,B}^{C22} \ln x_{C,F}^{C22} - w_{B,T}^{C21} \ln x_{B,F}^{C21}) = 9.60 \quad (6.39)$$

We get a little entropy production ($\Delta S_{sur}' > -\Delta S$) due to some irreversibility in the heat exchanger system since we take out some heat which is returned at slightly lower temperature levels. However, the separation in the column is not affected.

Note that in the general case we might get $\Delta S_{sur}' < -\Delta S$ from (6.39). However, this would only imply that we are unable to obtain full heat exchange in the middle sections, and (6.39) would then not represent the complete $\Delta S_{sur}'$.

All the external heat is now supplied in the bottom reboiler and along the bottom section of C22, and is removed along the top of C21 and in the top condenser. The internal flows and compositions will be identical to the case in Figure 6.2a.

Example 1a . We may alternatively operate the top and bottom as adiabatic sections and supply all the required heat in the bottom remove it in the top (we still consider the same heat exchange in the middle sections). The entropy change in the surroundings will in this case simply be given by equation (6.23):

$$\Delta S_{sur} = R V_B^{C22} \ln \alpha_{LH} = 8.31 \cdot 1.00 \cdot \ln 4 = 11.52 \quad (6.40)$$

6.5.3 Example 2: Heat Exchange Across the Dividing Wall

Usually, capital costs prevent distributed reboilers and condensers along the sides of column sections. However, in a Dividing Wall Column (DWC) the sections are closely integrated, and it is interesting to study if we may get positive energy effect for free, from the potential of heat transfer across the dividing wall. The idea is to distribute the heat transfer inside the Petlyuk arrangement, and possibly obtain reduced energy consumption in the reboiler.

The beneficial directions are always from section C1B to C22T and from C1T to C21B. However, the temperature profiles along the sections may not always give a driving force in the beneficial directions. In addition, the heat transfer surface area may not be sufficient. This implies that the desired heat transfer coefficient will depend on feed properties and flow rates. Lestak et al. (1994) considered heat

transfer across the wall for a given case, and it was found that it could be beneficial to allow for heat transfer along a part of the dividing wall and insulate the other part. But the overall reduction in heat input was found to be small.

In our example we may try to operate only sections C1B and C22T reversibly. The upper parts (C1T, C21T and C21B) will then require the same flows as in the adiabatic arrangement. Note that the demand in C1B is higher than the available heat in C22T, so we can never reach fully reversible operation in C1B. However, by using equations (6.46) and (6.49) we can adjust the feed junction composition to C22 until we get a heat balance ($\Delta V_T^{C22} = \Delta V_B^{C1}$). This occurs at $x_B = 0.571$, which gives $V_B^{C1}/F = 0.611$ and the need for boilup is reduced with about 8%, from $V_B^{C22}/F = 1.366$ in the pure adiabatic case to $V_B^{C22}/F = 1.222$.

An alternative approach is to use forced heat integration at one, or more, pairs of stages. For example by using a part of the vapour flow from a stage in C22T as a heating fluid at a suitable stage in C1B. This may give better flexibility in operation compared to a passive heat transfer across the wall, but it will be more expensive in implementation and operation.

6.5.4 Example 3: Pre-heating of the Feed by Heat Exchange with the Sidestream

The result obtained when all the available internal heat is utilized is not very realistic. However, it is possible to obtain positive results with a much simpler approach. In the adiabatic arrangement in Figure 6.2b, a large portion of the sidestream is available as vapour. If the feed temperature is below the boiling point of the pure intermediate, it is possible to transfer heat from the sidestream to the feed. Stage temperatures for the example is obtained from equation (6.52) where we assume that the heavy component boiling point is $T_{b,C} = 310K$ and heat of vaporization $\lambda = 25kJ/kg$ (which is close to n-pentane properties at 100kPa).

With equimolar feed, the feed stage temperature $T_F = 285.1K$ while the pure B-component boiling point is $T_{b,B} = 289.3K$. However, as the amount of vapour in the feed is increased, the feed stage temperature will be increased, and the amount of sidestream available as vapour will be decreased. The vapour flows have been computed from the V_{min} -diagram methods presented in chapter 4.

For the given case we find that with a liquid fraction $q = 0.7620$, all the heat from condensing the sidestream is transferred to the feed. The resulting liquid feed stage composition is changed from $x_F = [0.333, 0.333, 0.333]$ to $x_F = [0.2778, 0.3398, 0.3825]$ and the feed stage temperature becomes $T_F = 287.0K$, which is still below $T_{b,B}$. The important mole fraction of the intermediate B-component in pinch zone at the feed junction to C22 is reduced from $x_{BP} = 0.608$ to $x_{BP} = 0.560$ and the vaporization rate in the reboiler is reduced from $V_B^{C22}/F = 1.364$ to $V_B^{C22}/F = 1.181$.

The total need for vaporization per unit feed in both columns in a conventional direct split configuration is 2.032 in this case. The savings in reboiler duty compared with the plain Petlyuk arrangement is 32.8% and can in theory be increased to 41.8% when we heat the feed by condensing the vapour portion of sidestream.

This kind of heat integration is only possible when the feed temperature is below the sidestream temperature, and excess vapour is available at the sidestream stage. However, if there are other available streams in the plant at suitable temperature levels, the heat exchange does not have to be done directly between the sidestream and the feed stream, but with other streams.

Unlike the ideal heat exchange within the reversible arrangement, heat exchange with the sidestream can be implemented in practice, and we may get significant positive effects for units with realistic size and efficiency.

6.5.5 Summary of the Examples

The main results have been summarized in Table 6.1. The numbers should speak from themselves, but here are some observations:

- The Adiabatic Petlyuk column has the lowest energy consumption compared to the arrangement without heat integration. This will also be a general result (ref. Section 6.2 and Fidkowski and Krolikowski (1987)).
- The heat-integrated arrangements have even lower energy consumption than the Adiabatic Petlyuk column.
- The conventional direct split configurations require less separation work than the typical Petlyuk arrangement where all the heat is supplied in the bottom and is removed in the top. Note also that the different reversible arrangements may have quite different requirements for total heat supply
- However, the Petlyuk column with an heat exchanger at the sidestream stage (Figure 6.2b) has lower lost work than any of the arrangements without heat integration (this result may be case specific).
- For the given feed data, the extremely simple configuration with heat transfer to the feed from condensation of the available sidestream vapour flow (Ex 3) seems very promising, both with respect to small minimum energy requirement, small lost work, and possibility for practical realization.

However, we cannot make general conclusions from Table 6.1 since the data are only valid for the particular feed used in the example.

6.6 Operation at Several Pressure Levels

The pressure has a large impact on distillation and is widely used in the industry to get a suitable temperature range in each column. This is important for optimal use of available utilities, and for optimal heat integration within a plant. We will not go into much detail on this issue, but we will look at three examples below where we combine heat exchange and different pressure levels. Note that we here assume no loss related to the compression or expansion or in the heat exchangers.

6.6.1 Example 1: Feed Split (Binary Case)

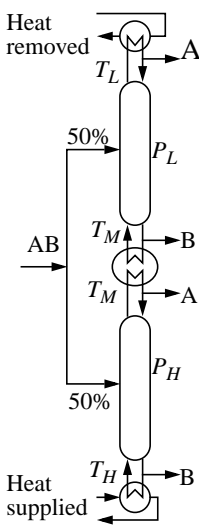


Figure 6.4: Binary separation at two pressures

The configuration in Figure 6.4 shows a case where the feed is split into two streams where 50% is separated at high pressure and 50% at low pressure. The pressures are adjusted so the temperature in the reboiler of the low-pressure column is equal to the condenser temperature of the high pressure column (T_M), and we assume perfect heat exchange. (Note that in practice we would require a certain temperature difference).

If the columns are adiabatic, it is trivial to find that the minimum boilup (V_{min}) in the high-pressure reboiler will be 50% less than in a single column since the feed is reduced by 50% (we assume constant relative volatility).

However, we may also check the requirements for a reversible system. The required pressure ratio is $P_H/P_L = \alpha$ which is found from equation (6.1) by equating the boiling points of pure light component at high pressure and pure heavy component at low pressure at (T_M), thus the relation between temperature spans are given by:

$$\frac{1}{T_L} - \frac{1}{T_H} = 2\left(\frac{1}{T_M} - \frac{1}{T_H}\right) = 2\left(\frac{1}{T_L} - \frac{1}{T_M}\right) \quad (6.41)$$

Minimum separation work will of course be identical for any separation process, but minimum required vaporization in reversible distillation depends on the temperature span as given by (6.12). For the temperature span in Figure 6.4 we obtain:

$$V_{rev,min} = \frac{-\sum z_i \ln z_i}{\log \alpha + \log P_H/P_L} = \frac{1}{2} \left(\frac{-\sum z_i \ln z_i}{\log \alpha} \right) \quad (6.42)$$

This result is 50% below the requirement for a single reversible column. Thus the efficiency of the adiabatic system $V_{rev,min}/V_{min}$ will be the same in the two cases, provided that the heat exchange between the columns is ideal.

This is an example where we trade heat supply with temperature span, but without changing the consumption of separation work. A typical application area is if a plant has available hot utility at high temperature, e.g. from an exothermal process like a methanol reactor. Then the configuration in Figure 6.4 is a practical arrangement that make the best use of the available energy.

6.6.2 Example 2: Double Effect Direct Split (DEDS)

For the ternary case we may consider a heat-integrated direct split configuration (double effect column). Figure 6.5 shows an example where the upper column (C1) performs a sharp A/BC split and the lower (C2) the B/C split. Column C2 is operated at a higher pressure so reboiling in column C1 can be obtained by full heat integration with the condenser in column C2. The required minimum pressure ratio (for ideal heat exchange) can be found from (6.1):

$$P_H/P_L = a_B(z_B + z_C)/(\alpha_B z_B + \alpha_C z_C) \quad (6.43)$$

The temperature levels can be computed from (6.1) for a given reference pressure level and corresponding boiling point of the reference component, and a given heat of vaporization.

The required vapour flow from the bottom reboiler in column C2 can be found by Underwood's equations for C1 and King's formula for C2 (Chapter 2):

$$\frac{V_{Bmin}^{C2}}{F} = \max\left(\frac{\alpha_A z_A}{\alpha_A - \theta_A} + (1 - q), \frac{\alpha_B z_B + \alpha_C z_C}{\alpha_B - \alpha_C}\right) \quad (6.44)$$

A double effect indirect split (DEIS) will be equivalent. Then the first column will normally be operated at high pressure, and will carry out the sharp AB/C split.

We may operate both columns at their minimum vapour flow at the same time, by adding or removing additional external heat in the middle heat exchanger so both expressions on the right hand side in (6.44) are fulfilled.

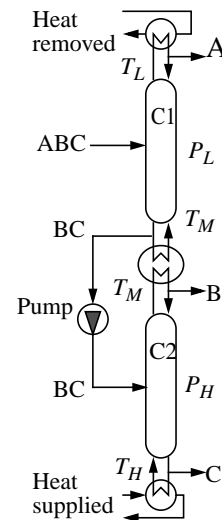


Figure 6.5: Double effect Direct Split (DEDS) configuration for 3-product separation

6.6.3 Example 3: Double Effect Prefractionator Column (DEPC)

Consider an arrangement with a separate conventional prefractionator column, but where the condenser in the prefractionator is directly heat integrated with the reboiler of the main column as shown in Figure 6.6 (Bildea and Dimian 1999), (Fonyó et. al. 1999) (Emtir et.al. 1999). In order to get direct heat transfer, the pressure in the prefractionator must be higher than in the main column. Note that all the external heat is supplied in one reboiler and is removed in one condenser, just as for the Petlyuk column. However, the main difference is operation at more than one pressure level.

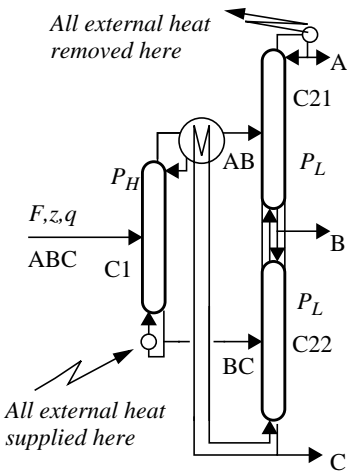


Figure 6.6: Separate prefractionator arrangement with double effect heat exchange (DEPC)

For any given split in C1 (distribution of the middle B-component) we will obtain a certain product composition in the top ($x_{A,T}^{C1}, x_{B,T}^{C1}$). The pressure ratio can be found from (6.1) by equating the temperatures in top of C1 (A+B components) and the bottom of C22 (pure C):

$$P_H/P_L = (\alpha_A x_{A,T}^{C1} + \alpha_B x_{B,T}^{C1})/\alpha_C \quad (6.45)$$

This ratio is obviously higher than for the direct split case (DFDS: 6.43) since for DSPC we need to have the same temperature for the mix of all the light A+ some B from C1 and pure C in C22, while for DEPS we only adjust the pressure until the temperature of the mix of all B+C from C1 equals pure C in C2.

The minimum vapour flow (when we consider total condensation in C1) is the maximum of the minimum requirement in each of the three columns. The absolute minimum will be determined by the requirement for the preferred prefractionator split ($V_{min}^{A/C}$). However, in cases where one of the main columns has a higher requirement, we have to supply some extra heat, either in the reboiler to C1 or to C22 or C21 directly.

6.6.4 Relation to the Petlyuk Column and the V_{min} -diagram

Minimum vapour flow in an adiabatic Petlyuk column is determined by the highest peak in the V_{min} -diagram (Chapter 4) as shown in Figure 6.7. Originally, this diagram characterize minimum vapour flow and feed component distribution in a two-product column (Chapter 3), and interestingly we also find the lower boundaries for minimum vapour flow in the double effect columns directly from the same diagram. The peaks correspond to minimum vapour flow for sharp product splits (A/BC and AB/C) and the “valley” in the middle correspond to the preferred

A/C split where the intermediate component is distributing. The diagram in the figure show a case where the A/BC split is simpler than the AB/C split, thus in selection between DEDS or DEIS, the preferred choice will be DEDS (note that this rule does not apply to conventional DS and IS configuration without heat integration, where we compare the sum of vapour requirement in both columns for the two configurations).

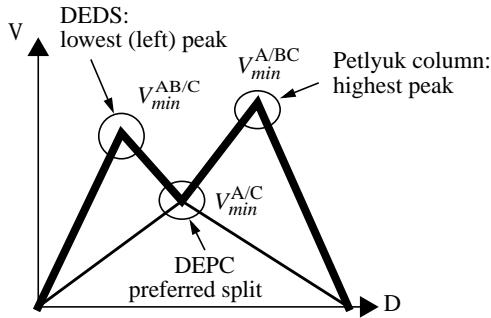


Figure 6.7: Minimum energy for the Petlyuk column, the DEDS and the DEPC illustrated in a V_{min} -diagram for a ternary feed (ABC)

The results are summarized in Table 6.2 below, where we also give data based on the same feed as used for the examples in Table 6.1.

Table 6.2: Relation between minimum vapour flows

Configuration	V_{min}		Diagram	$\Delta S_{total}/ \Delta S $
Petlyuk column, (with side HE)	$= \max(V_{min}^{A/BC}, V_{min}^{AB/C})$	= 1.366	= Highest peak	0.72 (0.54)
DEDS or DEIS (with extra HE)	$\geq \min(V_{min}^{A/BC}, V_{min}^{AB/C})$	= 1.072	= Lowest peak	0.64 (0.59)
DEPC	$\geq V_{min}^{A/C}$	= 0.778	= Preferred split	0.63

Note that we do not have an equality for the V_{min} -expression in the table for the double effect columns when the split in column C1 is very simple and require very low vapour flow compared to the split in next column(s).

The numbers in parenthesis apply to cases where we utilize the possibility for heat exchange at an intermediate temperature level. At the sidestream stage for the Petlyuk column, and in the heat exchanger in the DEDS column in order to reach minimum vapour flow in all sections simultaneously. For the DEPC configuration, the given feed data actually results in the same the minimum vapour flow in all three internal columns.

As in the binary feed split example, the double effect columns are also configurations where we trade an increased temperature range for a lower amount of heat supply. The temperature range in the DEDS/DEIS configurations will be larger than for the Petlyuk column, and the DEPC will have the largest range.

Due to the operation at several pressure levels, and thereby at different temperature ranges, we must be careful when comparing minimum vapour flows with the results for constant pressure in Table 6.1.

However, the entropy production (lost work) in the double effect columns are the same as for the corresponding configurations without heat integration. This implies that we do not improve the energy consumption (total heat supply) related to the theoretical reversible process with all heat exchanged at the most extreme temperatures.

6.7 Discussion

6.7.1 Plant-wide Issues

The general principle for any internal heat exchange configuration is to approach a reversible process, and at the same time approach the case where most of the external heat supply and heat removal occur at the most extreme temperatures.

However, heat integration should be considered on a plant-wide basis, and not only within a isolated plant segment, and as in all design problems, the investment cost must always be considered in comparison to the potential energy cost savings.

6.7.2 Heat Exchange at the Sidestream Stages

As discussed in Chapter 4 and 5, the adiabatic Petlyuk column has normally different minimum vapour requirements above and below the sidestream stages. It follows trivially from equation (6.6) that if these differences in vapour flow rates are obtained by adding or removing heat at the sidestream stages, the separation work will be reduced, thus the 2nd law efficiency will be improved. However, the total requirement for vaporization will not be affected.

Note that many authors generally state that Petlyuk arrangements has a higher net work consumption than conventional systems (e.g Carlberg and Westerberg 1989), but this is normally without considering any heat exchange at the sidestream stage. Data for the example in Table 6.1 is an example where we reach an opposite conclusion when we utilize the possibility for removing heat at the intermediate temperature level at the sidestream stage. (Here the relative entropy production ($\Delta S_{total}/|\Delta S|=0.54$ for the column in Figure 6.2b while the direct split configuration has $\Delta S_{total}/|\Delta S|=0.59$).

6.7.3 Non-Uniqueness of Heat Supply in Reversible Columns

The entropy change in the reversible system is unique, but there are many possible reversible process paths, and each will result in a different amount of heat consumed by vaporization. Note that the total entropy expression is independent of the junction compositions, thus if we can alter the junction compositions by a reversible sub-process, we can find alternative reversible paths.

Consider the ternary feed case again. The net component flow rates are given by the conditions at the feed stage. At the bottom of section C1, we have only B and C present. We omit the column superscript in the equations below where the net flow rates (w_B and w_C) are computed for the bottom of C1 (note that $w_A=0$). The reversible vapour flow (6.60) in the bottom of C1 and up to the feed stage in C22 can then be expressed as a function of x_B at the feed junction (where $x_A=0$). Note that these expressions also are valid for the adiabatic arrangement since it is based on a pinch assumption in the zone around the junction.

$$V_B^{C1}(x_B) = \frac{\alpha_C w_B}{(\alpha_B - \alpha_C)x_B} - \frac{\alpha_B w_C}{(\alpha_B - \alpha_C)(I - x_B)} + (w_B + w_C), x_A = 0 \quad (6.46)$$

In the process of removing the light component A, we have already shown that reversible operation follows a linear profile (Figure 6.3) towards the pinch composition given by (6.33). However, after the light A is removed completely, we may add a new subsection at the bottom of C1, denoted C1B'. By exchanging heat along the side of C1B' to obtain the vapour flow given in (6.46) we may alter the final junction composition x_B in both directions. We may reduce it by further heating until we reach the limiting composition when the vapour flow into the bottom of C1B' becomes zero. Then

$$x_B|_{v=0} = \frac{w_B}{w_B + w_C} \quad (6.47)$$

This is exactly the same condition as when we use an adiabatic prefractionator with its own reboiler, and saturated liquid feed to C22.

The effect on vapour flow in C22 can also be expressed as a function of x_B . In the bottom (C22B) the net flow of component C is identical to the net flow in the prefractionator, $w_B=0$, and the vapour flow up into the feed stage becomes:

$$V_{BF}^{C22}(x_B) = -\frac{\alpha_B w_C}{(\alpha_B - \alpha_C)(I - x_B)} + w_C \quad (6.48)$$

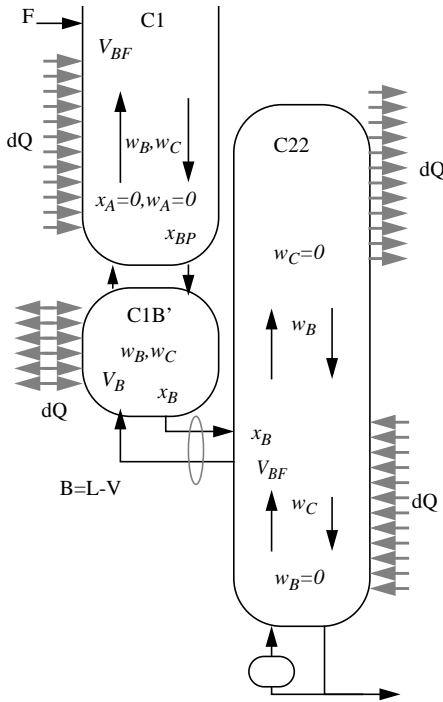


Figure 6.8: Modified reversible arrangement. In section C1B' we have only B and C present, and we may alter the composition x_B into C22, and thereby V_{BF}^{C22} , by heating or cooling reversibly, but without affecting the total net work consumption in the system.

Note that this is exactly the same expression that will determine minimum boilup in an adiabatic column ($w_C = (I - z)F$, $x_B = z$, $q = I$ gives Kings's formula for V_{min}).

An interesting issue, however, is how the total amount of vaporization flow in the integrated system is affected by the choice of x_B in a reversible arrangement. The total amount of vaporization in the bottoms of C22 and C1, is given by

$$V_{BF}^{C22} + (V_{BF}^{C1} - V_B^{C1}) = V_{BF}^{C1} + \frac{\alpha_C(-w_B)}{(\alpha_B - \alpha_C)x_B} + (-w_B) \quad (6.49)$$

Note that the material flows w_B and w_C are negative in the bottom sections and the vapour flow at the feed stage of C1 (V_{BF}^{C1}) is independent on x_B . From (6.49) we see that if we reduce x_B by adding more heat along C1B', the total amount of vaporization in C1 (including C1B') and C22 increase.

This suggest that we should cool along C1B', but, we still have to supply the same heating along the bottom of C1B. The picture is changed, however, if we allow internal heat exchange inside the system. Thus, if the heat removed in C1B' is reused in C1B we may still apply (6.49). Then the limiting point of operation is when $V_B^{CI} = V_{BF}^{CI}$, which gives the same flows as in the adiabatic case.

Thus, this confirms that it is possible to find a reversible arrangement with exactly the same heat supply requirement, and the same external flows and junction compositions as in the adiabatic Petlyuk arrangement, but this requires that we allow internal heat exchange inside the system. In this example we cool along C1B' and supply the same amount of heat back along C1B which seems a bit strange.

6.7.4 Practical Issues

In the design of an optimal separation system, we must apply a cost function which properly reflects the real operational costs and investment cost. Thus, there is no particular configuration which is optimal for a given separation task. In some cases, the optimal solution of a given cost criterion will be close to minimum energy, and in other cases closer to minimum lost work. This is dependent of prices on equipment, energy, raw materials and products, and on the available temperature ranges for heating and cooling utilities within a plant.

Zero lost work (entropy production) may be an unrealistic target since the capital cost of realizing a (close to) reversible process may become very high (infinite column sections, infinite number of heat exchangers infinite area in heat exchangers e.t.c.). For simulations of reversible columns we really need "infinite" number of stages. In adiabatic distillation columns, "infinite" number of stages can be simulated in practice with $N \approx 4xN_{min}$

Proper control and operation of a given distillation system is very important to obtain the full potential in terms of low energy consumption, product quality and volume. This is particularly important for complex arrangements like the Petlyuk arrangements, and for closely heat integrated arrangements.

6.8 Conclusion

Reversible distillation gives valuable insight in the energy requirement for multi-component distillation. However, even if the reversible arrangement is optimal with respect to entropy production or lost work (2nd law), we may have irreversible (adiabatic) systems with less energy consumption (1st law). Without considering any internal heat exchange, the adiabatic Petlyuk arrangement seems to achieve less minimum energy requirement than any other distillation system.

Internal heat exchange can be used for further reduction of the minimum external energy (heat supply). Heat exchange can be applied in any system where the temperature ranges where we need heat supply overlaps temperature ranges where heat is available. Thus, it can be applied for the ideal reversible arrangement as well as for the adiabatic Petlyuk column and also for conventional column arrangements. With a given total temperature range, the minimum energy target can be defined by a theoretical reversible distillation process where all the energy is supplied at the highest temperature and removed at the lowest temperature.

We have briefly discussed how pressure can be used to adjust temperature range in a distillation system, but this issue is by no means fully covered in this work, and it seems clear that this is an important area for further studies.

6.9 References

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6.10 Appendix: Reversible Distillation Theory

In a reversible process the total entropy production is zero, thus, for reversible distillation we trivially have $\Delta S_{sur} = -\Delta S$. However, we here show in more detail how we can obtain a reversible distillation process by applying heating and cooling along infinite non-adiabatic column sections in a particular way. Then, by computing ΔS_{sur} from (6.5) we verify that this process really is reversible by showing that we obtain $\Delta S_{sur} = -\Delta S$.

In a distillation column where the feed and products are saturated liquids, the heat supplied equals the heat removed, thus $\oint dQ = 0$. For entropy calculation in distillation sections, we need to relate heat flow and temperature to flows and compositions, and in the following we will deduce some useful expressions. A similar procedure is presented by Petlyuk (1964) for a binary mixture, but here we present a general procedure for multicomponent mixtures.

By applying integration by parts (Petlyuk 1964), we obtain:

$$\Delta S_{sur} = -\oint \frac{dQ}{T} = -\frac{1}{T} \underbrace{\oint dQ}_{=0} + \oint \left(\left(\underbrace{\int dQ}_{\neq 0} \right) d\left(\frac{1}{T}\right) \right) = \oint Q d\left(\frac{1}{T}\right) \quad (6.50)$$

We express all variables as function of composition, thus we consider $Q(x)$ and $T(x)$. The integration path will then be from the bottom to the top of the column and (6.50) can be expressed as:

$$\Delta S_{sur} = \int_{x_B}^{x_T} Q(x) \frac{d\left(\frac{1}{T(x)}\right)}{dx} dx \quad (6.51)$$

6.10.1 Temperature-Composition-Pressure Relationship

Let us first find an expression for the temperature and the differential $d(1/T(x))$.

We consider ideal components obeying Raoult's law; $p_i = Py_i = x_i p_i^o$, where p_i is the partial pressure of component i , p_i^o is the vapour pressure, x , y are the liquid and vapour phase mole fractions. According to Dalton's law, the total pressure $P = \sum p_i = \sum x_i p_i^o$. The relative volatility for any component related to a reference component is then given by $\alpha_i = p_i^o / p_r^o$. We divide the pressure with $p_r^o(T)$ and obtain: $P/p_r^o(T) = \sum (p_i^o(T)/p_r^o(T))x_i = \sum \alpha_i x_i$. Note that the temperature relation is through the vapour pressure of the reference component, given by: $\ln(p_r^o(T)) = \ln P_{ref} + (\lambda/R)(1/T_{b,r} - 1/T)$. Here $T_{b,r}$ is the boiling point of a pure reference component at the reference pressure (P_{ref}). This expression is obtained by integration of the Clausius-Clapeyron equation (Chapter 2) when we assume constant heat of vaporization (λ), negligible liquid volume and the ideal gas law. Then the temperature-composition-pressure relationship for the multicomponent mixture is given by (ref. Petlyuk (1964) for binary mixture):

$$\frac{1}{T} = \frac{R}{\lambda} \ln \left(\frac{P_{ref}}{P} \sum_i \alpha_i x_i \right) + \frac{1}{T_{b,r}} \quad (6.52)$$

We need $d(1/T)$ in (6.50) and by derivation of (6.52) we obtain:

$$d\left(\frac{1}{T}\right) = \frac{R \sum_i \alpha_i dx_i}{\lambda \sum_i \alpha_i x_i} - \frac{R dP}{\lambda P} \quad (6.53)$$

The vapour liquid equilibrium (VLE) is given by:

$$\frac{y_i}{x_i} = \frac{\alpha_i}{\sum_j \alpha_j x_j} \quad (6.54)$$

In the following we assume constant pressure. We can substitute the VLE into (6.53) and the relation between $d(1/T)$ and the compositions is then given by:

$$d\left(\frac{1}{T}\right) = \frac{R}{\lambda} \sum_i \left(\frac{y_i}{x_i} \right) dx_i = \frac{R}{\lambda} \sum_i (y_i - x_i) \frac{dx_i}{x_i} \quad (6.55)$$

Note that we have used the fact that $\sum dx_i = 0$ (when the sum is taken over all components) to obtain the expression with the factor $(y_i - x_i)$. This form is useful when we shall evaluate the integral (6.50) as we will show in (6.61).

6.10.2 The Reversible Vapour Flow Profile

Now, we need to express the heat supply as a function of composition along a section. When we consider ideal components with equal and constant heat of vaporization (λ), the heat input is directly related to vaporization and condensation:

$$dQ = \lambda dV = -\lambda dL \quad (6.56)$$

where L and V are the local liquid and vapour flow rates through the stages. Thus we can apply $Q(x) = \lambda V(x)$ together with $d(1/T(x))$ from (6.55) in the entropy expression in (6.50). Note that the contribution from reboilers and condensers is given by discrete terms like $\Delta Q/T$ as in (6.6).

To find the function $V(x)$ which gives reversible operation we study the internal properties of a column section. To avoid irreversible mixing, we assume infinite number of stages so that we can consider any local stage (n) as belonging to a zone of constant composition such that $x_{i,n} = x_{i,n+1}$. (Such a pinch condition is usual to assume also in adiabatic distillation for computing of minimum flows, but only in a certain zone of the section). The material balance on an arbitrary stage (n) inside a section is given by:

$$V_n y_{i,n} - L_{n+1} x_{i+1,n} = w_i \quad (6.57)$$

We use the pinch assumption, omit the stage index and obtain:

$$V y_i - L x_i = w_i \quad (6.58)$$

Here w_i is the net molar flow of component i (defined positive upwards, and into feed stages). The total net flow is obtained by taking the sum over all components:

$$V - L = \sum_i w_i \quad (6.59)$$

Note that in the top we have $D = \sum_i w_{i,T}$ and in the bottom $B = -\sum_i w_{i,B}$

By eliminating the liquid flow (L) we can express the vapour flow at any stage in a top or a bottom section as function of the stage composition for any given component (i):

$$\text{Top: } V_T = \frac{w_i - x_i D}{y_i - x_i} \quad \text{Bottom: } V_B = \frac{w_i + x_i B}{y_i - x_i} \quad (6.60)$$

6.10.3 Entropy Production in a Reversible Section

Note that the component flows (and thus also D and B) are always constant inside a given section. Then equation (6.60 top) combined with (6.55) in (6.50) gives:

$$\begin{aligned} \Delta S_{sur} &= \oint \lambda V d\left(\frac{I}{T}\right) = \oint \lambda \left(\frac{w_i - x_i D}{(y_i - x_i)} \right) \left(\frac{R}{\lambda} \sum_i (y_i - x_i) \frac{dx_i}{x_i} \right) \\ &= R \oint \sum_i \left((w_i - x_i D) \frac{dx_i}{x_i} \right) = R \oint \left(\sum_i \left(w_i \frac{dx_i}{x_i} \right) - \underbrace{D \sum_i dx_i}_0 \right) \end{aligned} \quad (6.61)$$

Note that the expression for vapour flow (V) in (6.60) is valid for any component (i) so we could use it inside the summation and cancel the factors $(y_i - x_i)$.

The final expression is independent of both relative volatility and heat of vaporization. We will get the same expression for both the top and bottom sections:

$$\Delta S_{sur} = R \oint \sum_i \left(w_i \frac{dx_i}{x_i} \right) \text{ or on differential form: } dS_{sur} = R \sum_i \left(w_i \frac{dx_i}{x_i} \right) \quad (6.62)$$

The net component flows (w_i) are always constant in a section, thus the integral in (6.62) gives us the logarithmic terms ($\ln x_i$) that appear in the system entropy expression (6.4). For a given separation case, we have to find the net component flows (w_i), but this is usually quite simple for the given set of specifications.

We show the application of the equations for reversible vapour flow and entropy production in detail for a binary case below, and discuss some of its properties (a multicomponent case is treated in Section 6.4.1).

6.10.4 Reversible Binary Distillation

The net component flows in each section of a binary distillation column are uniquely given by the two product specifications, here as the composition of the light component (x_D, x_B) (We use $x_1 = x$ and $x_2 = 1 - x$). Thus, $w_{1,T} = x_D D$, $w_{2,T} = (1 - x_D)D$ and $w_{1,B} = -x_B B$, $w_{2,B} = -(1 - x_B)B$

From (6.59) and (6.60), the resulting vapour and liquid flows in the top becomes:

$$V_T = D \frac{x_D - x}{y - x}, \quad L_T = D \frac{x_D - y}{y - x} \quad \text{and} \quad \frac{L_T}{V_T} = \frac{x_D - y}{x_D - x} \quad (6.63)$$

Similarly we find for the bottom:

$$V_B = B \frac{y - x_B}{y - x}, \quad L_B = B \frac{x - x_B}{y - x} \quad \text{and} \quad \frac{L_B}{V_B} = \frac{y - x_B}{x - x_B} \quad (6.64)$$

The entropy change of the surroundings as given in (6.62) becomes:

$$\Delta S_{sur} = R \left(\sum_i \left(\int_{x_{i,F}}^{x_{i,D}} w_{i,T} d(\ln x_i) \right) + \sum_i \left(\int_{x_{i,B}}^{x_{i,F}} w_{i,B} d(\ln x_i) \right) \right) \quad (6.65)$$

Note that $x_{i,F} = z_i$ and $z_i F = x_{i,D} D + x_{i,B} B$, thus we obtain:

$$\begin{aligned} \Delta S_{sur} = R [& D(x_D \ln(x_D) + (1 - x_D) \ln(1 - x_D)) + \\ & B(x_B \ln(x_B) + (1 - x_B) \ln(1 - x_B)) - \\ & F(z \ln(z) + (1 - z) \ln(1 - z))] / F \end{aligned} \quad (6.66)$$

and we find that $\Delta S_{sur} + \Delta S = 0$ by computing the actual state change of the system entropy ΔS from equation (6.4). Thus, we conclude that by applying the liquid and vapour flow rates as in (6.63) and (6.64), we have reached a reversible system, and it follows from the second law of thermodynamics that no further reduction in net work can be obtained.

The following two characteristics of reversible operation are very important for minimum energy results and will also be valid for the multicomponent case:

- We have to supply heat below the feed and remove heat above the feed. (See Figure 6.9)
- The vapour flow has its maximum through the feed stage and this flow rate is identical to the minimum vapour flow in an adiabatic column (with constant molar flows). (See Figure 6.9 and the McCabe diagram in Figure 6.10)

General comments and observations:

- Note that for feasible (positive) liquid flow rates in the top, the highest liquid composition in the top is really not x_D , but x_D^+ which is in equilibrium with $y_D=x_D$. Then $L_T=0$ and $V_T=D$. However, raising the purity from x_D^+ on the top stage to x_D in the product can be done by equilibrium vaporisation and then a direct condensation of the vapour to liquid, which is a reversible process for a binary mixture. And since the integral to x_D^+ is shown to give the reversible work expression, and the process on the final stage and condensation of the product is also reversible, we may integrate (6.65) to x_D even if the equations gives a negative L_T in for $x>x_D^+$.
- Note that the local slope of the operating line (L_T/V_T) at $[x,y]$ is the same as the line through $[x_D,x_D]$ and $[x,y]$, as illustrated in the McCabe-Thiele diagram in Figure 6.10.
- For pure products, the flow expressions can be simplified (shown for the top):

$$V_T = D \frac{\alpha}{(\alpha - 1)x}, L_T = D \frac{1}{(\alpha - 1)x} \text{ and } \frac{L_T}{V_T} = \frac{y}{\alpha x} \quad (6.67)$$

The slope L/V at the top becomes $1/\alpha$ and $L_T = Fz(\alpha - 1)$, while for non-pure product, the slope at the top is zero at $[x_D^+,y_D]$ and $L_T = 0$

For nonsharp-splits the liquid flow into the top is zero (see Figure 6.9), but as we approach pure products, the heat exchange close to the top will approach the amount required in the condenser for sharp splits (given by equation (6.67) for $x=1$). The behaviour in the bottom is equivalent.

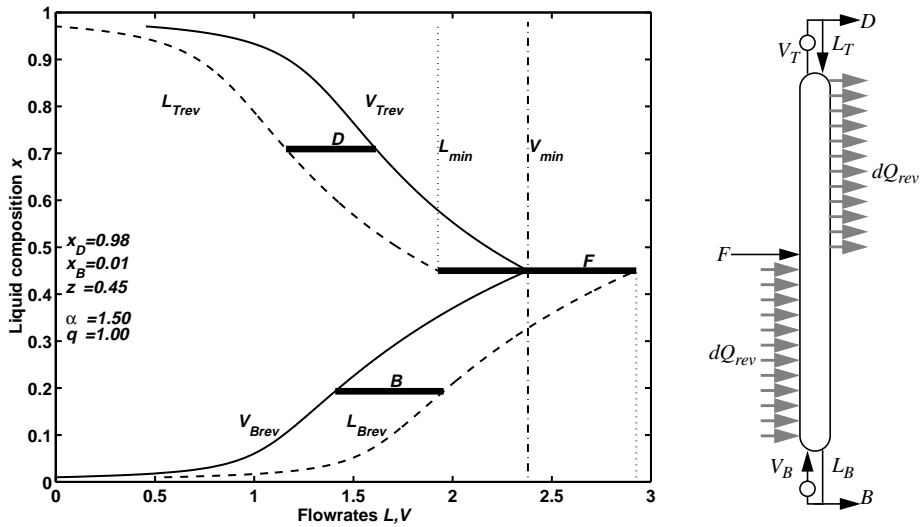


Figure 6.9: Reversible binary distillation: Internal flow rates (L_{rev}, V_{rev}) as function of composition. Minimum flow rates in an adiabatic column are also indicated (L_{min}, V_{min}). Note that $V_T \cdot L_T = D$ and $L_B \cdot V_B = B$

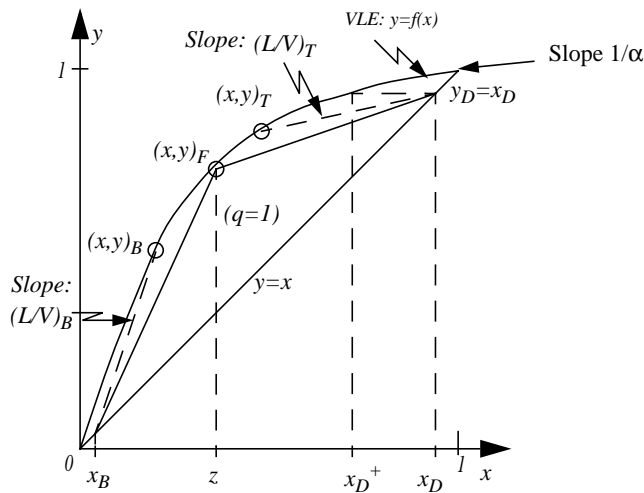


Figure 6.10: Reversible binary distillation in a McCabe-Thiele diagram. The operating point is always at the equilibrium curve anywhere in the column.

