

ALLOWABLE OPERATING REGIONS OF INTEGRATED DISTILLATION ARRANGEMENTS

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Abstract

The steady state behavior of an integrated three-product distillation column known as the Petlyuk column is examined. When four of the outlet compositions from the column are specified, the steady state solutions have a rather strange nature, including multiplicity. Our explanation to this behavior is based on the solutions of the system when only three of the outlet compositions are specified. These solutions were computed numerically using an arclength continuation method, and their nature is explained in physical terms. Based on the solutions, the effect of adding the fourth specification is explained.

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1 Introduction

The starting point for this work is the paper by Wolff *et al.* (1993,1994), who studied an integrated three-product distillation column known as the Petlyuk column. As the Petlyuk column has five degrees of freedom, they considered using four of these to control four of the outlet compositions from the column, and using the remaining fifth degree of freedom to minimize the energy consumption. They found that the (steady state) energy consumption as a function of the remaining degree of freedom does not have a nice convex shape - as might be expected - but exhibit a strange behavior with two solution branches and a "hole" in the operating range (i.e. there is a range of values for the remaining degree of freedom where no steady state solutions exist). The purpose of this paper is to provide an explanation of the steady state behavior found by Wolff *et al.*

The strategy chosen in order to provide such an explanation is as follows: First, we find all steady state solutions for a case with three outlet compositions specified, and interpret the solutions as geometrical surfaces. These surfaces can be linked to our physical understanding of the column. Then, the effects of adding/changing a fourth specification may be viewed geometrically in terms of the surfaces.

A brief outline of the paper: First, we define some terminology and explain the strange steady state behavior of the column when four outlet compositions are specified. We then describe the mathematical model and the numerical methods. After presenting the computed solution surfaces, we explain their shapes by physical arguments. We then draw a larger picture in terms of these solution surfaces in order to explain the effect of adding/changing specifications on the shape/nature of the solutions. Finally, we discuss the relevance to operation and control.

For readers unfamiliar with the Petlyuk column, we use the rest of this introduction to give a brief review of previous work. The review follows that of Wolff *et al.* (1993,1994).

In 1939, Brugma (1939) proposed a column design similar to the Petlyuk column, but with a reboiler and a condenser also in the prefractionator. This design has been denoted a pseudo-Petlyuk design by previous workers (e.g. Wolff *et al.*, 1994).

Stupin and Lockhardt (1971) study the use of Fenske-Underwood equations for the design of Petlyuk columns.

Tedder and Rudd (1978) examine the energy requirement of various separation arrangements, including the pseudo-Petlyuk column.

Fidowski and Krolkowski (1986) compare the energy requirement of the Petlyuk column to other designs, such as the direct and indirect sequences.

Glinos and Malone (1988) derive analytical expressions for various column designs, including the

Petlyuk column.

Chavez *et al.* (1986) study multiplicity in Petlyuk columns. They have an explanation of the multiplicity in terms of "matching specifications in interlinked columns".

Faravelli *et al.* (1989) examine the resilience of the steady states of the Petlyuk column to changes in the internal flows. Their work is based on the work by Chavez *et al.*

Triantafyllou and Smith (1992) give an overview over the design of Petlyuk columns. They explain how the Petlyuk column may be approximated - from a design point of view - by a regular distillation column and two side strippers.

The only report of an industrial application of a Petlyuk column is from BASF in Germany, as reported by Rudd (1992).

2 The Petlyuk column

The purpose of this section is to describe the column, describe the problem at hand and to introduce some terminology needed for the subsequent discussion.

The column

The Petlyuk column separates a three component mixture of light (A), intermediate (B) and heavy component (C). Fig. 1 shows the system, consisting of a prefractionator and a main column.

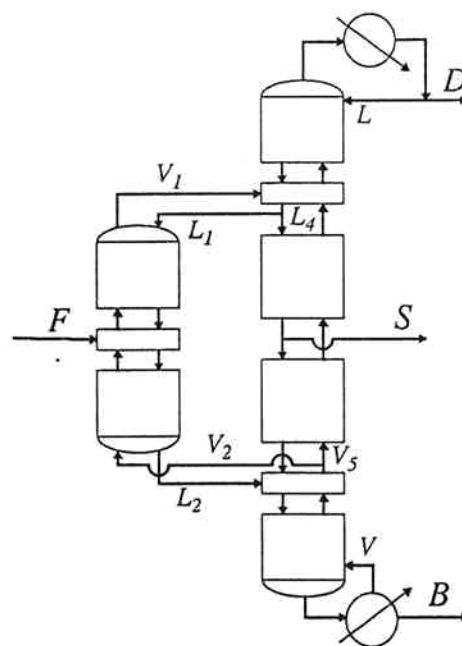


Figure 1: Petlyuk column

The main column resembles an ordinary distillation column in that there is a reboiler at the bottom,

producing a vapor flow rate, V , and a condenser at the top, yielding a liquid reflux, L . The prefractionator, however, does not have a separate reboiler and condenser. Instead it takes a fraction, $R_V \equiv V_2/V$, of the vapor stream, V , from the main column to use as boilup, and it takes a fraction, $R_L \equiv L_1/L$, of the liquid stream, L , to use as reflux.

The feed to the system, F , is placed at the middle of the prefractionator. The products are withdrawn from the main column: the light component in the distillate flow, D ; the intermediate component in the side draw, S ; and the heavy component from the bottom, B .

Note that *in practice* the prefractionator and the main column may be built in a single shell using a dividing wall.

Degrees of freedom and operating objectives

It is assumed that the holdups (condenser level, reboiler level and pressure) are already controlled. The system then has five degrees of freedom, which may be represented by the parameters given above: L , V , S , R_L and R_V . Possible objectives (specifications) for the operation of the column might be to control the compositions of the outlet streams and to minimize the energy consumption.

If the relative volatilities are reasonably large, there will be almost no heavy component at the top of the main column, and almost no light component at the bottom; it is therefore unnecessary to specify these two quantities. Hence, we need only one degree of freedom to control each of the top and bottoms compositions, and two for the side composition (possibly one if we don't care whether the impurity is light or heavy), a total of four (three) degrees of freedom. The remaining degree of freedom (two degrees) could then be used to minimize the energy consumption. For subsequent discussion we distinguish between two different sets of specifications:

1. *Three compositions specified.* In this case three degrees of freedom are used to specify one concentration in each outlet stream, i.e. $x_{A,top}$, $x_{B,side}$ and $x_{C,bottom}$. The remaining two degrees of freedom are used for energy minimization.
2. *Four compositions specified.* In some cases the side product may be specified - not in terms of the purity of the intermediate (B), $x_{B,side}$ - but rather in terms of both a maximum amount of light impurity (A) and a maximum amount of heavy impurity (C). Specifying these two impurities is equivalent to specifying, in addition to the three specifications above, the ratio of light to heavy impurity in the side stream, $x_{A,side}/x_{C,side}$. In this case, there is only one degree of freedom left for energy minimization.

Solutions and solution manifolds

We define a *solution* to be an operating point which satisfy the specifications on the compositions, not taking the energy minimization into account. Since there are remaining degrees of freedom there will be a family of solutions; we will denote such a family a *solution manifold*.

For the first specification set above, "Three compositions specified", there are two degrees of freedom left (five degrees of freedom minus three specifications). Hence, the solution manifold is twodimensional, i.e. a surface. For the second specification set, "Four compositions specified", there is one degree of freedom left, i.e. the solution manifold is a curve. Note that, in general, the solution manifold does not have to be connected; it may consist of several parts, or branches.

Observe that the solution manifold of the second specification set is part of the solution manifold of the first set. Hence, by calculating the solution manifold of the first specification set, we have also calculated the solution manifold of the second.

A typical solution manifold

It is not always easy to predict what type of solutions to expect, not even qualitatively. For example, consider the second specification set, with four compositions specified. One might intuitively think that the energy consumption as a function of the remaining degree of freedom, X say, would be a nice convex function with a well defined minimum. However, as was shown by Wolff *et al.* [17], the numerical solution typically looks like the curve shown in Fig 2, exhibiting two solution branches and a hole in the operating range. In the figure, the remaining degree of freedom, X , is the vapor split fraction, R_V . Other choices of X yield similar results.

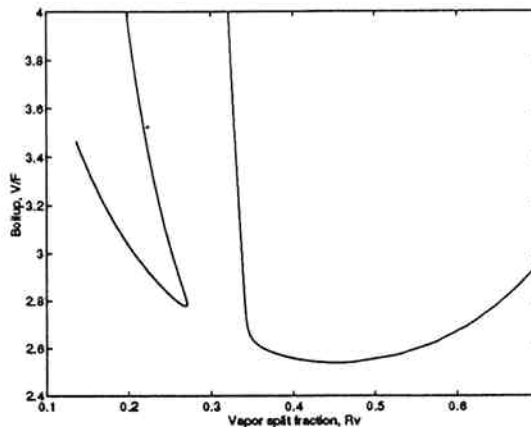


Figure 2: Energy consumption, V/F , as function of R_V . Four compositions specified.

Before explaining why the energy consumption as a function of the remaining degree of freedom has

such a shape, we describe the simple mathematical model and the numerical methods used to support our arguments. We compute all solutions of the model for the case with three specifications.

3 Mathematical model and numerical solution

3.1 Model of the column

The model is simplified as much as possible to reveal some essential features of the Petlyuk column. It is assumed (Fig. 1):

- Eight equilibrium stages in the prefractionator, sixteen in the main column;
- Constant relative volatilities 4:2:1;
- Constant molar flows;
- Total condenser; and
- Reboiler taken as one equilibrium stage.

The mass balances for equilibrium stage "i" are taken as:

$$L \cdot x_{c,i+1} - V \cdot y_{c,i} = L \cdot x_{c,i} - V \cdot y_{c,i-1} \quad (1)$$

where L and V are the liquid and vapor molar flows through the stage; $x_{c,i}$ and $y_{c,i}$ are the liquid and vapor mole fraction of component "c", where "c" refers to any of the three components A, B or C .

The vapor mole fraction of component "c", $y_{c,i}$, is given by the vapor equilibrium:

$$y_{c,i} = \frac{\alpha_c x_{c,i}}{\sum_{c=A,B,C} \alpha_c x_{c,i}} \quad (2)$$

The mixer and splitter models mix or split either liquid or vapor. The mass balance for liquid becomes:

$$\sum_{in} F_{in} x_{c,in} = \sum_{out} F_{out} x_{c,out} \quad (3)$$

where the F 's are molar flows and "in" and "out" are indices over the inlet and outlet streams. This expression determines a mixer; for a splitter more specifications are needed: the split fraction of the outlet flow rates, i.e. R_L or R_V , and that the outlet flows have the same composition. The mass balance for vapor is the same as for liquid when x is replaced by y .

The reboiler model is taken as an equilibrium stage with the vapor flow rate specified. The condenser is supposed to be a total condenser, i.e. the condenser model simply converts vapor to liquid with no change in compositions.

Now that the models of the column elements - the stages, mixers, splitters, reboiler and condenser - are established, we describe how these were combined to an equation system.

3.2 The equation system

The model described in the previous section was solved using the equivalent of McCabe-Thiele stepping: The vapor and liquid compositions at the middle of the prefractionator and the main column are guessed, and the system is solved stage by stage towards the top and the bottom of the column. Of course, the guess is in general wrong, so there will be a mismatch - residuals - in the splitters at the top and bottom of the prefractionator, as well as in the reboiler and in the condenser. One may consider this stage-by-stage procedure to be equivalent to an equation system $f(z) = 0$, where:

- z : unknowns consisting of five degrees of freedom - L, S, V, R_L, R_V - and eight guesses (four streams times two mole fractions), a total of 13 unknowns.
- f : function values consisting of three specifications and eight mismatches (four locations times two mole fractions), a total of eleven function values.

It is seen that there are two more unknowns than equations, which means that there exists a two-parameter family of solutions, i.e. a *two-dimensional surface*.

To find the solution surface, a continuation method was used.

3.3 A simple continuation scheme

Continuation methods are well established; therefore the method used will only be briefly discussed here. An easily accessible book on the subject is the book by Seydel, 1988 ([12]). The method used here is not particularly sophisticated and was chosen for its simplicity.

The equation system to be solved is on the form $f(z) = 0$ with two more variables than equations, i.e. the solutions form a surface. Since a surface may be constructed from the curves lying in it, a family of such solution curves are found. The overall procedure is:

1. First find an initial solution z_0 (with flow splits $R_{L,0}, R_{V,0}$).
2. Temporarily make an additional specification to get only **one** more variables than unknowns. The choice of additional specification is almost arbitrary; here, we fix the ratio $(R_L - R_{L,0}) / (R_V - R_{V,0})$.
3. Solve the resulting equation using arclength continuation to find a solution curve lying in the solution surface.

4. Repeat for various values of the ratio $(R_L - R_{L,0})/(R_V - R_{V,0})$, each time starting at z_0 .
5. Construct the solution surface from the computed curves.

The arclength continuation method used in this procedure is as follows: We are to solve $f(z) = 0$, now with only one unknown more than the number of equations. Assume that we have already calculated a set of points $z_0, z_1 \dots z_n$ on the solution curve, and that we desire to find a point z_{n+1} a distance approximately δ from z_n . First linearize to get the equation for the tangent of the solution curve: $0 = J \cdot \Delta z$, where $J \equiv \frac{\partial f}{\partial z^T}$ is the Jacobian of f . It is seen that any deviation, Δz , on the tangent is in the *null space* of the Jacobian. The hyperplane orthogonal to the tangent is spanned by the *row space* of the Jacobian. This suggests the following two step procedure:

1. A prediction of the next point z_{n+1} is found by taking a small step, δ , in the direction of the nullspace of the Jacobian, $\mathcal{N}(J_n)$ (tangent direction). The Jacobian was computed numerically, using central differences.
2. A correction is made iterating in the row space of the Jacobian, $\mathcal{R}(J_n^T)$, by Newton-Rapson like iterations using the pseudoinverse of the Jacobian, J_n^+ , evaluated at the previous point, z_n :

$$z_{n+1}^k = z_{n+1}^{k-1} - J_n^+ \cdot f(z_{n+1}^{k-1}), \quad k = 1, 2, 3 \dots \quad (4)$$

3.4 Numerical results

As the solutions forms a two parameter family - i.e. a surface - one may parametrize it by almost any two variables. Here, we use the liquid and vapor split fractions, R_L and R_V , as parameters. Any other quantity, Q say, may thus be considered a function of these two parameters, formally $Q = Q(R_L, R_V)$.

Figs. 3 and 4 show a typical result as a function of the parameters, R_L and R_V . Fig. 3 show the calculated energy requirement, represented by the boilup V/F ; Fig. 4 show the calculated ratio of light to heavy component in the side stream, $x_{A,side}/x_{C,side}$. As can be seen, the energy surface is a nice convex function with a well defined energy minimum, whereas the ratio of light to heavy component in the side stream has the shape of a saddle.

4 Discussion of results

In this section we first make a physical argument as to why the surfaces ought to have the shape they have. We then explain the connection between the

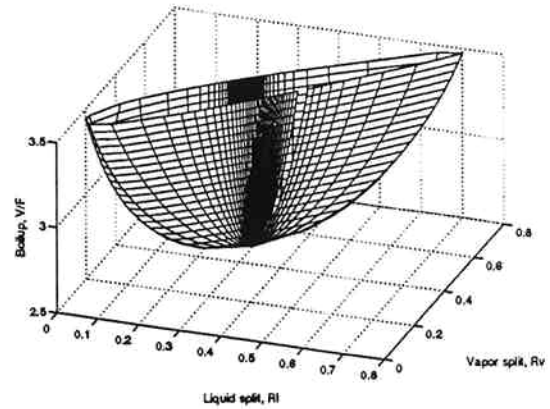


Figure 3: Calculated energy consumption, V/F , as a function of R_L and R_V . Three compositions specified.

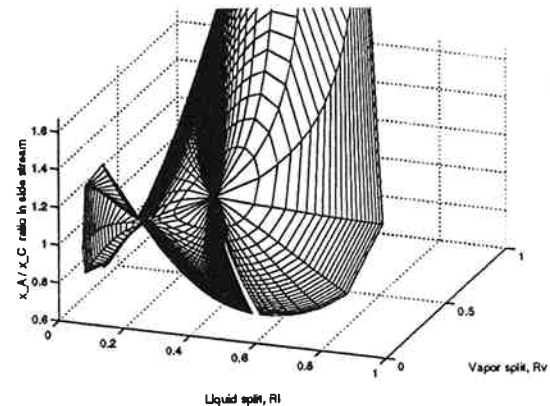


Figure 4: Calculated ratio of light to heavy impurity in side stream. Three compositions specified.

shape of these surfaces and the effect of specifying e.g. the side stream impurity.

Shape of the energy surface

First, consider the calculated energy requirement as a function of the liquid and vapor split fractions, R_L and R_V , Fig. 3. The energy surface looks as we might intuitively expect, with a well defined minimum. Any excursion from the minimum point costs in terms of energy. Too high R_L and R_V means that we use all the vapor and liquid in the prefractionator, not leaving much to the main column, which is stupid. A similar argument applies to low R_L and R_V , not leaving much for the prefractionator. High R_L and low R_V means that the liquid to vapor ratio is too high in the prefractionator and too low in the main column, again stupid. A symmetrical argument applies to low R_L and high R_V .

Shape of the $x_{A,side}/x_{C,side}$ surface

Next, consider Fig. 4 showing the calculated ratio of light to heavy component in the side stream, $x_{A,side}/x_{C,side}$ as a function of the liquid and vapor split fractions, R_L and R_V .

A change in one of the split fractions has an effect on both the prefractionator and the main column. For example, consider a *decrease* in the liquid split fraction, R_L (while keeping R_V and the other three product compositions constant). This affects the purity of the sidestream in two ways:

1. By degrading the separation in the prefractionator: The liquid to vapor ratio, L/V , in the prefractionator *decreases*, and if it becomes sufficiently low, there is a breakthrough of heavy component, C , at the top of the prefractionator into the main column. This heavy component ends up in the side stream, *reducing* the ratio of light to heavy component in the side stream, $x_{A,side}/x_{C,side}$.
2. By improving the separation in the main column: The liquid to vapor ratio, L/V , in the main column *increases*, which by itself increases the concentration of light component, A , in the side stream, and *increases* the ratio of light to heavy component in the side stream, $x_{A,side}/x_{C,side}$.

The two effects are thus *competing*, indicating that the ratio $x_{A,side}/x_{C,side}$ may have a *maximum* along a line of constant R_V . This is indeed the case and may be explained as follows: If R_L is decreased towards zero starting from a relatively high value, then the ratio $x_{A,side}/x_{C,side}$ will first *increase* due to the second effect, but a further reduction of R_L will *decrease* $x_{A,side}/x_{C,side}$ due to the first effect (breakthrough of heavy component). A similar argument along a line of constant R_L (varying the vapor split fraction, R_V) yields a *minimum* along this direction.

To summarize: The $x_{A,side}/x_{C,side}$ surface has a maximum along a line of constant R_V and a minimum along a line of constant R_L , that is, the surface has the shape of a *saddle*.

Combining the surfaces

Now that the general shape of the solution surfaces has been established, we explain what this means in terms of specifying e.g. the $x_{A,side}/x_{C,side}$ ratio. Consider Fig. 5, showing an *idealized* picture of the solution surfaces (functions of the liquid and vapor split fractions, R_L and R_V). Fig 5c shows the $x_{A,side}/x_{C,side}$ ratio, while Fig 5b shows the energy consumption. The two surfaces have been placed above each other on purpose, to emphasize that points on the two surfaces that have the same arguments R_L and R_V correspond to each other. In other words,

corresponding points on the two surfaces are placed *above* one another.

Specifying the $x_{A,side}/x_{C,side}$ ratio is equivalent to finding the intersection of the saddle, Fig 5c, with a horizontal plane. Since the surface is a saddle, this intersection yields *two* branches of intersection curves (marked with 'O' in the figure). The corresponding branches on the energy surface are found by projecting these two curves upward onto the energy consumption surface, Fig 5b. The original figure with a "hole" in the operating range, Fig. 5a, is found by projecting the branches onto the $R_V - V/F$ plane.

This explains the effect of adding the fourth specification on the outlet composition. It can be seen geometrically the effect of changing the value of the fourth specification; this corresponds to translating the horizontal plane upwards or downwards. Thus we have obtained a qualitative picture of the effect of the fourth specification.

5 General discussion

A comment on the saddle shape

Since the flow split fractions R_L and R_V represent some sort of trade-off between the column sections - i.e. larger internal flows in one column section means less in another - it should not come as a surprise to find maxima or minima along lines of constant R_L or lines of constant R_V . Also, since the Petlyuk column has a near top-bottom symmetry, one would expect R_L to have a similar effect on the $x_{A,side}/x_{C,side}$ ratio as R_V has on the *reciprocal* of this quantity, i.e. $x_{C,side}/x_{A,side}$. This indicates strongly that the $x_{A,side}/x_{C,side}$ ratio as a function of R_L and R_V has the shape of a saddle; at least it should not be very surprising. One may conjecture that such saddle shapes should be quite common in integrated column arrangements. Adding a specification on a saddle-like quantity would then imply multiple solution branches and complex multiple solutions.

Operation and Control

As shown by Wolff *et al.* (1993,1994), it is relatively easy to control the Petlyuk column for the case with three compositions specified, but the fourth specification complicates matters. It is not obvious how to design a good feedback control system in this case. However, systems with more manipulated variables (in our case, the degrees of freedom) than outputs (in our case, the outlet concentrations) normally do not have fundamental limitations in achievable control performance (i.e. bandwidth limitations). Since

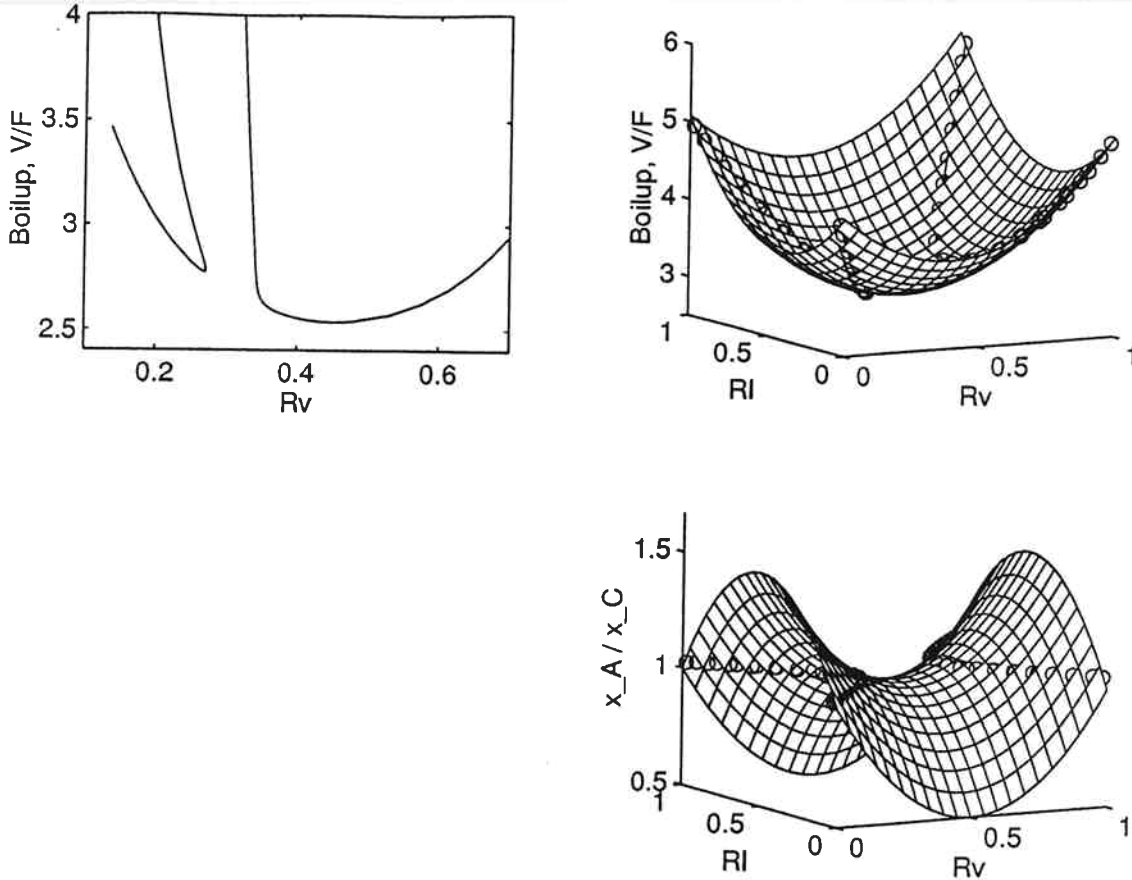


Figure 5: Relation between three specifications and four specifications. Note: surfaces have been idealized.

there is one excess degree of freedom when four outlet compositions are specified, we do not expect any fundamental limitations in the achievable performance in this case. The design of such controllers is an area of future research.

A comment on thermodynamics

In this work, we have used a very simple model with constant relative volatilities; still, this system exhibits a rather complex steady state behavior. More rigorous thermodynamics yield similar results (Wolff *et al.*, 1993, 1994). However, it should be noted that more complex thermodynamics might complicate the steady state behavior even further.

6 Conclusion

Complex steady state behavior of Petlyuk columns - with multiple solutions and a "hole" in the operating range - was explained by physical/geometrical arguments. The numerical method used for supporting the arguments was an arclength continuation method applied to a simple mathematical model of a Petlyuk column.

The nature of the solutions are similar to what is obtained by more rigorous models.

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Nomenclature

B	Bottoms flow rate [mole/time]
D	Distillate flow rate [mole/time]
$f(z)$	equation system of Petlyuk column
F	Feed flow rate [mole/time]
J	Jacobian matrix of $f(z)$
L	Liquid reflux flow rate in top of main column [mole/time]
L_1	Liquid flow rate in top of prefractionator [mole/time]
R_L	Liquid split fraction at top of prefractionator, L_1/L [-]
$R_{L,0}$	R_L in the vector z_0 [-]
R_V	Vapor split fraction at bottom of prefractionator, V_2/V [-]
$R_{V,0}$	R_V in the vector z_0 [-]
S	Side stream flow rate [mole/time]
V	Vapor flow rate from reboiler [mole/time]
V_2	Vapor flow rate in bottom of prefractionator [mole/time]
x	Liquid mole fraction [-]
y	Vapor mole fraction [-]
z	unknown vector in equation system $f(z)$
z_0	Starting value of z for continuation algorithm
Greek	
α	relative volatility [-]
Subscripts	
<i>bottom</i>	indicates bottoms flow
<i>c</i>	index over component A (light), B (intermediate), C (heavy)
<i>in</i>	index over inlet streams
<i>out</i>	index over outlet streams
<i>side</i>	indicates side stream
<i>top</i>	indicates distillate flow