

Instability of Distillation Columns

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As recently recognized, distillation columns, operating with reflux and boilup as independent inputs, may have multiple steady-state solutions, even in the ideal binary case. Two fundamentally different sources may cause the multiplicity, and in both cases some operating points are found to be unstable. This article provides evidence for the instability and discusses the effect of operating conditions on stability. Increasing the internal flow rates increases the probability of instability; when flows other than reflux and boilup are used as independent inputs, an operating point may become unstable if the level control is not sufficiently tight. In this case, a limit cycle, usually stable, appears as the steady state goes unstable.

Introduction

The dynamic behavior of distillation columns has been studied quite extensively over the past decades and several general qualitative properties have been proposed. One suggested property is that the operating points of distillation columns, at least in the binary case, always are globally asymptotically stable (with level and pressure control). This conjecture is based on results published over the years on the uniqueness and stability of distillation columns (for example, Acrivos and Amundson, 1955; Rosenbrock, 1960, 1962; Doherty and Perkins, 1982; Sridhar and Lucia, 1989). Doherty and Perkins (1982) provide a review of results published on this subject and conclude that multiplicity and instability is impossible in any binary distillation column. However, it is important to realize that all these studies include restrictive assumptions. First, all the studies assume that the flows, for example, reflux L and boilup V , are fixed on a molar rate basis. As Jacobsen and Skogestad (1991) argue, this is rarely the case in operating columns, especially for liquid flows. For instance, fixing the valve position will normally correspond closely to fixing the geometric average of mass and volumetric flow rate.

Secondly, most studies include the assumption of constant molar flows (neglected energy balance). Sridhar and Lucia (1989) include the energy balance in their study, but conclude that also in this case the operating points of binary distillation columns will be unique. They do, however, only study a limited number of configurations (sets of specifications), namely the $Q_D Q_B$ and LB configurations. (The term "configuration" is used in distillation control to denote the two independent variables which remain for composition control.)

In a recent article, Jacobsen and Skogestad (1991) analyze models without the two above mentioned assumptions and show that distillation columns, even in the ideal binary case, may display multiple steady states. They identify two different sources that may cause the multiplicity:

- Most operating columns will have the flows fixed on a mass or volume basis, while the separation is determined by the size of the molar flows. The transformation from mass or volume flows to molar flows is nonlinear due to the composition dependence and may in some cases become singular. A singularity in the input transformation will imply that several solutions exist in terms of the outputs (for example, compositions) for a given specification of inputs (flows).
- When the energy balance is included in the model, even molar inputs may yield multiple solutions. The multiplicity is caused by interactions between flows and compositions through the material and energy balances.

Jacobsen and Skogestad (1991) treat the multiplicity from a steady-state point of view only. In this article we study the dynamics of columns with multiple solutions and provide proof of instability for some configurations (specifications).

It is well known that for the simple distillation columns studied in this article, with given feed stream, two products, and no intermediate heaters or coolers (see Figure 1), there are only two degrees of freedom at steady state, that is, only two independent specifications are possible. A large number of specifications (configurations) are possible for distillation columns. One typical specification is L and D (LD configuration), and others are $Q_D Q_B$, LV , LQ_B , $y_D x_B$ and so on. Note from the last specifications that one may at steady state also specify dependent variables (in this case product compositions). However, in terms of dynamics and control there is a fundamental

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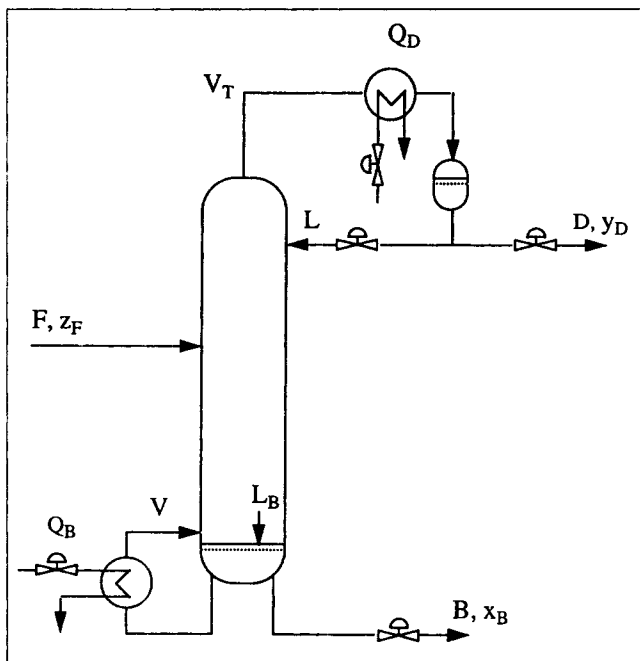


Figure 1. Two product distillation column.

difference between independent variables (“inputs” in control) and dependent variables (“outputs” in control). In an operating column only the former may be specified directly, while the latter only may be specified indirectly through the manipulation of the former, for example, through feedback control. Mathematically, we require any dynamic model to be causal and this is satisfied only when independent variables are specified. Also, in the dynamic case we usually have at least three additional degrees of freedom because the pressure (vapor holdup) and the reboiler and condenser levels (liquid holdups) may vary dynamically. When studying the dynamic behavior of distillation columns in this article, we assume that the pressure and the two levels are controlled, that is, we are studying a partly controlled system. This is reasonable since otherwise all distillation columns are unstable because the two levels behave as pure integrators. In any case, usually the levels and pressure are tightly controlled so that we practically are left with two degrees of freedom also in the dynamic case.

We will assume that the cooling Q_D always is used for pressure control and restrict ourselves to consider as independent variables the flows L , V , D , and B (note that boilup rate V is closely related to heat input Q_B). These flows may be specified on a molar basis (in which case we use no subscript) or on a mass basis (in which case we use subscript w). Typically, the product flows D and B are used to control the levels, which leaves L and V as independent variables and we get the LV configuration. This is the most widespread configuration in industry and in this article we mainly discuss this configuration. However, there are also other possibilities for controlling the levels and therefore many possible configurations (see, for example, Skogestad and Morari, 1987).

We review the results presented in Jacobsen and Skogestad (1991) on steady-state multiplicity caused by singularities in the input transformations. We provide evidence for the instability of some of the operating points for this case and discuss the effect of operating conditions on the stability of distillation

columns. The multiplicity and instability caused by singularities in the input transformations is independent of the energy balance and, for simplicity, we therefore assume constant molar flows. We then include the energy balance in the model and provide evidence for the instability that may result from interactions between flows and compositions through the material and energy balances. The effect of operating conditions on this type of instability is then discussed.

At the end of the article we consider the D_wV and L_wB_w configurations. We show that while steady-state multiplicity is unlikely, they may have unstable operating points if level control is slow. It is shown that the instability in this case results from a Hopf bifurcation and that a limit cycle hence appears as the steady state goes unstable.

In this article we concentrate on the theoretical aspects of the dynamic behavior of distillation columns. In a separate article (Jacobsen and Skogestad, 1991b) we study the practical implications of the observed instability on operation and control.

Steady-State Multiplicity in Ideal Distillation with L_wV Configuration

We give here a brief review of the results on multiplicity caused by singularities in the input transformation presented in Jacobsen and Skogestad (1991). By “ideal” we mean that the thermodynamic behavior is ideal and that we have constant molar flows. Specifically, we assume that the vapor-liquid equilibrium (VLE) is described by constant relative volatility α :

$$y_i = \frac{\alpha x_i}{1 + (\alpha - 1)x_i} \quad (1)$$

and that at steady state we have for all stages (except at feed locations):

$$V_i = V_{i+1}; \quad L_i = L_{i+1} \quad (2)$$

where the subscript denotes the stage number. With assumption 2 the energy balance is not needed. Note that in order to make our dynamic model more realistic we have included liquid flow dynamics (Table 1) so that dynamically $L_i \neq L_{i+1}$. However, the flow dynamics do not affect the stability and all analytical results we present are therefore valid also if the flow dynamics are neglected. Throughout the article we also assume negligible vapor holdup and constant pressure.

Jacobsen and Skogestad (1991) provide an example of steady-state multiplicity in a column separating a mixture of methanol and *n*-propanol. The column has mass reflux L_w and molar boilup V as independent variables, that is, L_wV -configuration. Data for the column are given in Table 1. Some steady-state solutions are given in Table 2, and we see that for a specification of mass reflux $L_w = 50.0$ kg/min and molar boilup $V = 2.0$ kmol/min there are three possible solutions II, III and IV in terms of compositions. The multiplicity is graphically illustrated in Figure 2.

The observed multiplicity is caused by the transformation between the actual flow rates (mass) and the molar flow rates which determine separation. For a binary mixture the trans-

Table 1. Data for Methanol-Propanol Column

z_F	F	α	N	N_F	M_1	M_2
0.50	1	3.55	8	4	32.0	60.1

- Feed is saturated liquid.
- Total condenser with saturated reflux.
- Liquid holdups are $M_{Li}/F=0.5$ min, including reboiler and condenser except for D_wV configuration where $M_B/F=M_D/F=5.0$ min.
- Liquid flow dynamics: $L_i=L_{i0}+(M_{Li}-M_{Li0})/\tau_L$, $\tau_L=M_{Li0}/3L_{i0}$
- Negligible vapor holdup.
- Constant pressure (1 atm).

formation between mass reflux, L_w , and molar reflux, L , is given by:

$$L = L_w/M; \quad M = y_D M_1 + (1 - y_D) M_2 \quad (3)$$

where y_D is the mole fraction of light component in the distillate product. M_i denotes the molecular weight of the individual components and M denotes the molecular weight of the reflux and distillate product. One might expect the molar reflux to increase monotonically with the mass reflux, that is, $(\partial L/\partial L_w)_V > 0$. However, because M is a function of composition, y_D , and thereby of L_w , this might not be the case. Assuming molar boilup V fixed and differentiating $L_w = LM$ on both sides with respect to L yields:

$$\left(\frac{\partial L_w}{\partial L}\right)_V = M + L(M_1 - M_2) \left(\frac{\partial y_D}{\partial L}\right)_V \quad (4)$$

Here, the steady-state value of $(\partial y_D/\partial L)_V$ is usually positive (it may be negative when the energy balance is included as discussed later). For $M_1 < M_2$ (the most volatile component has the smallest molecular weight), which is usually the case, the second term on the righthand side of Eq. 4 will then be negative and $(\partial L_w/\partial L)_V$ may take either sign. The transformation from L_w to L will be singular when $(\partial L_w/\partial L)_V = 0$, that is, $(\partial L/\partial L_w)_V = \infty$. A singular point corresponds to a limit point, around which there locally exist two steady-state solutions (see, for example, Golubitsky and Schaeffer, 1985). Jacobsen and Skogestad (1991) state that solutions with $(\partial L_w/\partial L)_V < 0$ (middle branch in Figure 2) correspond to unstable operating points, but they do not prove this rigorously.

Instability with L_wV -Configuration

Methanol-propanol example

The "maximum eigenvalue" (the eigenvalue with the largest real part) at selected operating points of the methanol-propanol

Table 2. Steady-State Solutions for Methanol-Propanol Column with $V=2.0$ kmol/min and $L_w=48$ to 53 kg/min*

	L kmol/min	D kmol/min	L_w kg/min	y_D	x_B
I	1.064	0.936	48.00	0.534	0.00310
II	1.143	0.857	50.00	0.584	0.00350
III	1.463	0.537	50.00	0.9237	0.00780
IV	1.555	0.445	50.00	0.9969	0.104
V	1.650	0.350	53.00	0.9984	0.233

*Constant molar flows (no energy balance); see also Figure 2.

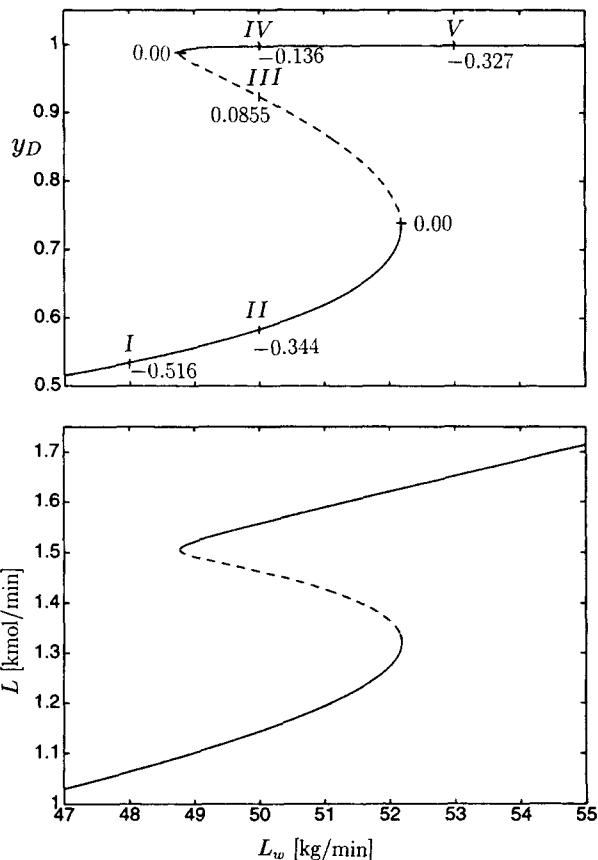


Figure 2. Methanol-propanol column with constant molar flows: multiple steady-states for L_wV configuration.

Mass reflux L_w is varied while molar boilup V is fixed at 2.0 kmol/min. On upper plot, corresponding maximum eigenvalue is shown at some steady-state solutions. Broken line represents unstable solutions.

column with constant molar flows is indicated in Figure 2. The eigenvalues were obtained by linearizing a nonlinear dynamic model with two states per tray (fraction of light component and liquid holdup). The steady-state values used for the liquid holdups are given in Table 1. Note that the nonlinear model is used in all simulations. From Figure 2 we observe that the maximum eigenvalue at the upper and lower branches (positive slope) is negative, implying stability, while the one at the intermediate branch (negative slope) is positive, implying instability of the operating points. There is only a single eigenvalue in the RHP at any unstable operating point. The eigenvalue at the singular points, which correspond to limit points, are zero as expected. The open-loop instability at the intermediate branch is illustrated by the nonlinear simulations in Figure 3 which shows the responses in top composition y_D to small changes in mass-reflux L_w (keeping boilup V fixed) starting from equilibrium at the unstable operating point III (initial holdups are as given in Table 1). The simulations indicate that the two stable solutions II and IV have equally large regions of attraction as seen from the unstable solution III.

Below we provide evidence for the observed instability and discuss under which operating conditions instability is most likely to occur.

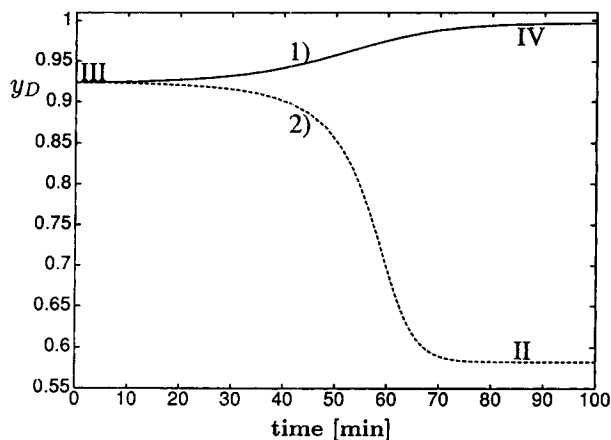


Figure 3. Nonlinear open-loop simulation of methanol-propanol column starting at unstable operating point III in Figure 2.

1) = increase in mass reflux L_w of 0.01 kg/min; 2) = decrease in mass reflux L_w of 0.01 kg/min. Boilup $V = 2.0$ kmol/min. Constant molar flows.

Conditions for instability

One-Stage Column. It is useful to study the simplest case for which the above-mentioned instability may occur (Jacobsen and Skogestad, 1991). To this end, consider the simple column in Figure 4 with one theoretical stage (the reboiler) and a total condenser (of course, such a column will never be operated in practice because the reflux is simply wasting energy and has no effect on separation). Assume binary separation, liquid feed, constant holdup in the reboiler (M_L) and negligible holdup in the condenser. The dynamic model of the column becomes:

$$M_L \frac{dx_B}{dt} = Fz_F - Dy_D - Bx_B \quad (5)$$

Here $D = V - L$ and $D + B = F$ and with L and V as independent variables we get:

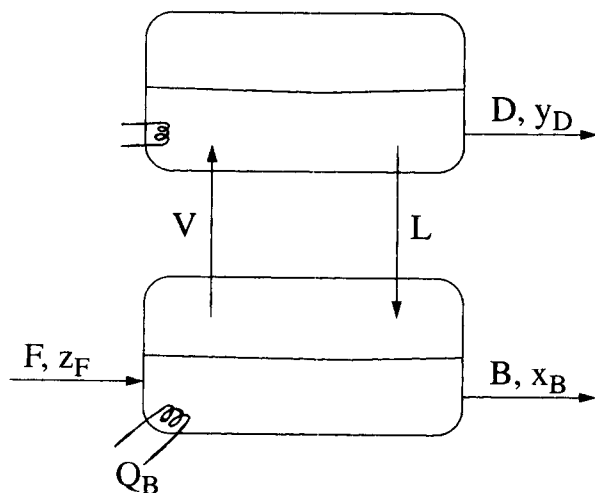


Figure 4. One-stage column with total condenser.

$$M_L \frac{dx_B}{dt} = F(z_F - x_B) + L(y_D - x_B) + V(x_B - y_D) \quad (6)$$

Linearization, Laplace transformation and introduction of deviation variables assuming F , z_F , and V constant yields:

$$sM_L dx_B(s) = -D^* dy_D(s) - B^* dx_B(s) + (y_D^* - x_B^*) dL(s) \quad (7)$$

where s is the Laplace variable and a superscript * shows that these are the nominal steady-state values (this superscript is deleted in the following to simplify notation). Linearizing the expression for the vapor-liquid equilibrium (for example, expression 1 for the case of constant relative volatility) yields:

$$dy_D = K(x_B) dx_B \quad (8)$$

where K is the local slope on the equilibrium curve. Equation 7 then becomes:

$$dx_B(s) = \frac{y_D - x_B}{M_L s + a} dL(s) \quad (9)$$

where

$$a = KD + B \quad (10)$$

As all terms in Eq. 10 are positive, the eigenvalue (pole) of the linearized system, $-a/M_L$, is always negative, implying that all operating points are stable when molar reflux L and molar boilup V are used as independent variables.

Now consider mass reflux L_w as an input instead of molar reflux $L = L_w/M$. By linearizing Eq. 3 we obtain for binary mixtures:

$$dL(s) = \frac{1}{M} dL_w(s) + L \frac{M_2 - M_1}{M} dy_D(s) \quad (11)$$

Substituting Eq. 11 into Eq. 9 and using Eq. 8 we obtain the following transfer-function between liquid composition, $dx_B(s)$, and mass reflux $dL_w(s)$

$$dx_B(s) = \frac{y_D - x_B}{M_L s + a_w} \frac{dL_w(s)}{M} \quad (12)$$

where

$$a_w = KD + B - (y_D - x_B) \frac{M_2 - M_1}{M} KL \quad (13)$$

The operating point is unstable for $a_w < 0$. We now want to compare this condition for instability with the condition for a negative slope for $(\partial L_w / \partial L)_v$ given in Eq. 4. At steady-state we have $s = 0$ and Eqs. 8, 9, and 10 yield:

$$\left(\frac{\partial y_D}{\partial L} \right)_v (0) = \frac{y_D - x_B}{D + B/K} \quad (14)$$

which inserted into Eq. 4 yields:

$$\left(\frac{\partial L_w}{\partial L} \right)_v (0) = \frac{M}{KD + B} a_w \quad (15)$$

and we find for the simple one-stage column that instability ($a_w < 0$) is equivalent to a negative steady-state slope for $(\partial L_w / \partial L)_v$.

Multistage Column. The dynamic model of a multistage column will in general be of relatively high order and it is difficult, if not impossible, to calculate the eigenvalues analytically. In order to assess the stability of a multistage column we will therefore employ the Routh-Hurwitz stability criterion and derive a sufficient condition for instability. To do this we first need to derive an expression for the transfer-function $(\partial y_D / \partial L_w)_v(s)$. With V constant the total differential of y_D may be written:

$$dy_D(s) = \left(\frac{\partial y_D}{\partial L} \right)_v(s) dL(s) \quad (16)$$

We have $L = L_w/M$ and for binary mixtures M is a (static) function of y_D only. Thus, for binary separations Eq. 11 applies. Combining Eqs. 16 and 11 yields:

$$\left(\frac{\partial y_D}{\partial L_w} \right)_v(s) = \frac{\frac{1}{M} \left(\frac{\partial y_D}{\partial L} \right)_v(s)}{1 - \frac{L(M_2 - M_1)}{M} \left(\frac{\partial y_D}{\partial L} \right)_v(s)} \quad (17)$$

To use the Routh-Hurwitz criterion write the transfer-function from molar reflux L to top composition y_D on the following form:

$$\left(\frac{\partial y_D}{\partial L} \right)_v(s) = \frac{k_{y_D L}^{LV} (1 + b_1 s + b_2 s^2 + \dots + b_m s^m)}{1 + a_1 s + a_2 s^2 + \dots + a_n s^n} \quad (18)$$

where

$$k_{y_D L}^{LV} = (\partial y_D / \partial L)_v(0) \quad (19)$$

is the steady-state gain. The response is assumed to be strictly proper, that is, $n > m$. We also assume that this transfer function is stable, that is, we have "molar" stability. This always holds for the case of constant molar flows (Doherty and Perkins, 1982). From the Routh-Hurwitz stability criterion, which says that the system is stable only if all the coefficients in the pole polynomial (denominator of Eq. 18) have the same sign, we conclude that all a_i 's in Eq. 18 are positive. Inserting Eq. 18 into Eq. 17 yields:

$$\left(\frac{\partial y_D}{\partial L_w} \right)_v(s) = \frac{\frac{1}{M} k_{y_D L}^{LV} (1 + b_1 s + b_2 s^2 + \dots + b_m s^m)}{1 + a_1 s + a_2 s^2 + \dots + a_n s^n - \frac{k_{y_D L}^{LV} L (M_2 - M_1)}{M} (1 + b_1 s + b_2 s^2 + \dots + b_m s^m)} \quad (20)$$

Since $n > m$ we know that the highest-order term in the denominator is $a_n s^n$ where a_n is positive. From the Routh-Hurwitz condition we then find that Eq. 20 is unstable if the constant term in the denominator is negative, and we conclude that a sufficient criterion for instability with the $L_w V$ -configuration is:

$$\frac{k_{y_D L}^{LV} L (M_2 - M_1)}{M} > 1 \quad (21)$$

This is exactly the same criterion as given in Eq. 4 in order to have a negative slope between mass and molar reflux. Thus, we have proven for binary mixtures that a sufficient condition for instability with the $L_w V$ -configuration is that at steady state:

$$\left(\frac{\partial L}{\partial L_w} \right)_v < 0 \quad (22)$$

In conclusion, we have proved that solution branches with a negative slope between L_w and L represent unstable solutions with the $L_w V$ -configuration. This result is in accordance with numerical results (see, for example, Figure 2).

Remarks. (1) Our derivation has assumed binary mixture and "molar" stability, but otherwise no assumptions about the energy balance or VLE has been made.

(2) We assume perfect control of the pressure and of the condenser and reboiler levels. The assumption of perfect level control is *not* important for the $L_w V$ configuration since the composition responses are only weakly dependent on the level control in this case. In particular, the presence of instability we find will be independent of the level control. This is easily understood by considering the general form (Eq. 18) of $(\partial y_D / \partial L)_v(s)$. Changing the tuning of the level controllers will affect the size of the parameters a_i and b_i to some extent, but will not affect the size of the steady-state gain $k_{y_D L}^{LV}$ nor the stability of $(\partial y_D / \partial L)_v(s)$. Since our derivation of the sufficient condition for instability (Eq. 21) is independent of the size of the parameters a_i and b_i , the level control will not affect the instability we find. Similar arguments apply to the effect of flow dynamics, that is, the presence of flow dynamics in the dynamic model does not affect the instability we find.

(3) It is of interest to consider the instability conditions from a somewhat more general perspective. Write L as a total differential of L_w and y_D to get the following generalization of Eq. 11:

$$dL(s) = \left(\frac{\partial L}{\partial L_w} \right)_{y_D}(s) dL_w(s) + \left(\frac{\partial L}{\partial y_D} \right)_{L_w}(s) dy_D(s) \quad (23)$$

Combining Eqs. 16 and 23 then yields:

$$\left(\frac{\partial y_D}{\partial L_w} \right)_v(s) = \frac{\left(\frac{\partial L}{\partial L_w} \right)_{y_D}(s) \left(\frac{\partial y_D}{\partial L} \right)_v(s)}{1 - \left(\frac{\partial L}{\partial y_D} \right)_{L_w}(s) \left(\frac{\partial y_D}{\partial L} \right)_v(s)} \quad (24)$$

We see that the instability condition (Eq. 21) corresponds to having the static term in the denominator negative:

$$\left(\frac{\partial L}{\partial y_D}\right)_{L_w}(0)\left(\frac{\partial y_D}{\partial L}\right)_V(0) > 1 \quad (25)$$

Note that Eq. 25 corresponds to a positive feedback loop with the product of the loop gains being larger than unity at steady state. The transfer-function from L_w to y_D in Eq. 24 may alternatively be written:

$$\left(\frac{\partial y_D}{\partial L_w}\right)_V(s) = \frac{\left(\frac{\partial y_D}{\partial L}\right)_V(s)}{\left(\frac{\partial L}{\partial L_w}\right)_V(s)} \quad (26)$$

and we see that provided $(\partial L/\partial L_w)_{y_D}(0) > 0$ (it is equal to $1/M$ for binary mixtures), the instability condition (Eq. 25) is equivalent to:

$$\left(\frac{\partial L}{\partial L_w}\right)_V(0) < 0 \quad (27)$$

Effect of operating conditions on stability

Jacobsen and Skogestad (1991) provide analytical results on when a negative slope between mass and molar reflux, that is, instability according to the analysis above, is most likely. They show that a negative slope is most likely with large internal flows (that is, large L and V) and intermediate purities in the top (that is, intermediate L for given V). This corresponds to having L and k_{yD}^{LV} large, and according to Eq. 21 this is the case for which instability is most likely. Note that the analytical treatment in Jacobsen and Skogestad (1991) was based on ideal separation with constant relative volatility and constant molar flows.

Figure 5 shows the stable and unstable regions in terms of boilup V and distillate flow D for the methanol-propanol column with the L_wV -configuration. The regions were determined using the full nonlinear dynamic model. Note that the assumption of liquid feed and constant molar flows implies that $L = V - D$. Thus, for a given V , $D = 0$ corresponds to maximum reflux L_{\max} and $D = F$ to minimum reflux L_{\min} . From the figure we see that at low internal flows, that is, low values of V , there is no unstable region and hence all operating points are unique and stable. However, for values of V above 1.32 kmol/min there exists a region of unstable operating points which, as expected from the analysis, expands with increasing V . We also see from the figure that instability, as expected, is most likely for intermediate values of D , that is, intermediate values of L for a given V . Furthermore, from Figure 5 we see that instability is unlikely for values of the distillate flow D significantly less than 0.5 ($=Fz_F$). This is explained by the fact that the top product becomes very pure with D significantly less than Fz_F , and that k_{yD}^{LV} in Eq. 21 becomes correspondingly small.

The borders between the stable and unstable regions in Figure 5 correspond to limit-points with the L_wV configuration. Locally, two steady-state solutions exist around a limit-point

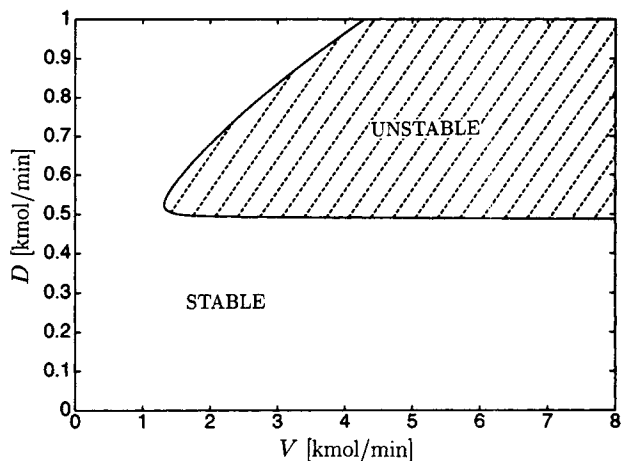


Figure 5. Regions of stable and unstable operating points in terms of distillate flow D and boilup V for methanol-propanol column with L_wV configuration.

Constant molar flows.

(see, for example, Golubitsky and Schaeffer, 1985). One might expect that for values of V for which there are two singular points there exist three steady-state solutions for some value of L_w , for example, as in Figure 2. However, this is not necessarily the case since the upper and lower solution branches (resulting from the two limit-points) may not overlap. (They will always overlap if we allow for negative product flows.)

We finally note that although the instability criterions (Eqs. 21 and 22) derived above only are sufficient, they give exact limits for instability in this example, for example, the border between the stable and unstable region in Figure 5 correspond to operating points with $(\partial L_w/\partial L)_V(0) = 0$. This is also the case for all examples with constant molar flows that we have studied. However, as we shall see below, when the energy balance is included in the model there may exist borders between stable and unstable regions which are not predicted by Eqs. 21 and 22.

Instability Caused by the Energy Balance

To this point we have assumed "molar stability" which always holds for the binary case with constant molar flows. However, Jacobsen and Skogestad (1991) show that when the energy balance is included in the model, even molar specifications may yield multiplicity in distillation. The multiplicity is in this case caused by interactions between the flows and compositions inside the column. The flows will affect the compositions through the material balance while the compositions will affect the flows through the energy balance.

Methanol-propanol example

Figure 6 and Table 3 show steady-state solutions for the methanol-propanol column with the LV configuration and the energy-balance included in the model. The heats of vaporization of the two pure components at 1 atm are approximately $\Delta H_1^{\text{vap}} = 35$ kJ/mol and $\Delta H_2^{\text{vap}} = 41$ kJ/mol (the exact enthalpy data used are given in Table 4). Note that the lightest component in terms of volatility (methanol) also has the smallest

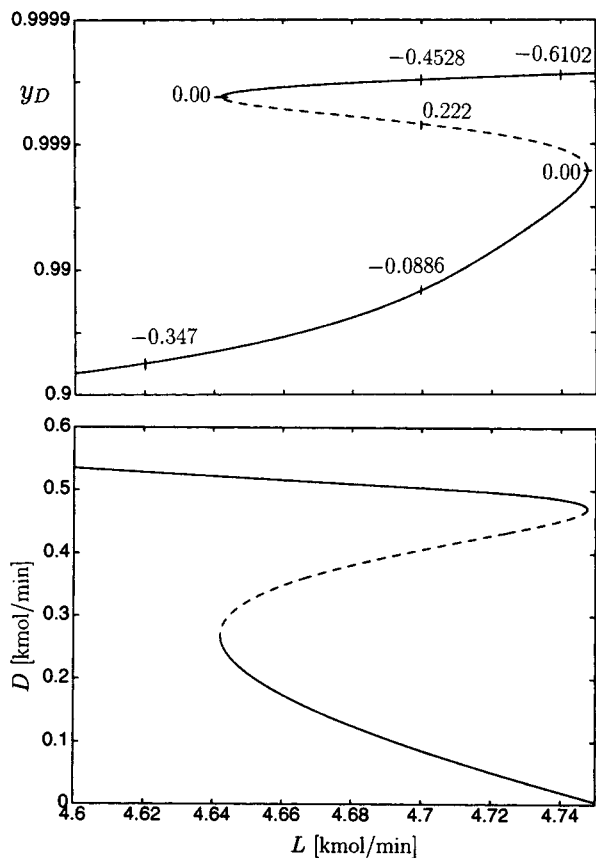


Figure 6. Steady-state solutions as function of molar reflux L for methanol-propanol column with energy balance included.

Maximum eigenvalue is shown in selected operating points. Eigenvalues were computed assuming a static energy balance. Boilup $V = 4.5$ kmol/min.

heat of vaporization. The maximum eigenvalue at selected operating points is also shown in Figure 6, and we see that the solution branch with a negative slope between molar reflux L and top composition y_D corresponds to unstable solutions. The eigenvalues were obtained using the same dynamic model as was used for the L_wV configuration above apart from the inclusion of the energy balance. For simplicity we used a static energy balance, that is, the term dU^L/dt was neglected in the dynamic model.

Conditions for instability with LV -configuration

To consider the stability properties for the LV configuration with the energy balance included we utilize the fact that the DV configuration in all known cases yields unique solutions (Sridhar and Lucia, 1989; Jacobsen and Skogestad, 1991) which are stable under the assumption of perfect level control (see

the section on instability with D_wV -configuration). We will again employ the Routh-Hurwitz criterion to derive a sufficient condition for instability and hence need an expression for $(\partial y_D / \partial L)_V(s)$.

Table 3. Steady-State Solutions for Methanol-Propanol Column with Boilup $V = 4.5$ kmol/min*

	L kmol/min	D kmol/min	y_D	x_B
I	4.60	0.535	0.9324	0.002474
II	4.70	0.505	0.9845	0.006344
III	4.70	0.406	0.9993	0.1587
IV	4.70	0.0866	0.9997	0.4526

*The energy balance is included in the model (enthalpy data are given in Table 4); see also Figure 6.

The transfer function from molar reflux L to top composition y_D may be written:

$$\left(\frac{\partial y_D}{\partial L}\right)_V(s) = \left(\frac{\partial y_D}{\partial D}\right)_V(s) \left(\frac{\partial L}{\partial D}\right)_V^{-1}(s) \quad (28)$$

Write the transfer-function from D to y_D on the form:

$$\left(\frac{\partial y_D}{\partial D}\right)_V(s) = k_{y_D D}^{DV} \frac{n_1(s)}{d(s)} = k_{y_D D}^{DV} \frac{1 + b_1s + \dots + b_n s^n}{1 + a_1s + \dots + a_m s^m} \quad (29)$$

Note that the constant terms in the polynomials $n_1(s)$ and $d(s)$ are equal to 1. Stability implies that all a_i 's in Eq. 29 are positive. A material balance around the condenser yields (assuming perfect level control):

$$L = V_T - D \quad (30)$$

where V_T denotes vapor flow to the condenser. Next consider the energy balance, and let $\tilde{H}^V(y_i)$ denote the molar enthalpy of the vapor with each component as pure saturated liquid at column pressure as reference. For simplicity we will here assume saturated liquid feed and neglect changes in the liquid enthalpy with composition. An overall energy balance then yields (see Appendix for details):

$$V_T = \frac{\tilde{H}_B^V}{\tilde{H}_T^V} V \quad (31)$$

where

$$\tilde{H}_B^V = \tilde{H}^V(y_B); \quad \tilde{H}_T^V = \tilde{H}^V(y_T) \quad (32)$$

y_B is the composition of the vapor leaving the reboiler and is assumed to be in equilibrium with x_B . y_T is the composition of the vapor on the top stage, which at steady state is equal to y_D . Inserting Eq. 31 in the material balance (Eq. 30) and differentiating yields for a binary mixture:

$$\left(\frac{\partial L}{\partial D}\right)_V(s) = -1 + V \frac{\left(\frac{\partial \tilde{H}^V}{\partial y}\right)_{y=y_B} \left(\frac{\partial y_B}{\partial D}\right)_V(s) \tilde{H}_T^V - \left(\frac{\partial \tilde{H}^V}{\partial y}\right)_{y=y_D} \left(\frac{\partial y_T}{\partial D}\right)_V(s) \tilde{H}_B^V}{(\tilde{H}_T^V)^2} \quad (33)$$

where the transfer functions from D to y_B and y_T may be written:

Table 4. Saturated Molar Enthalpies (kJ/mol) for Methanol-Propanol System at a Pressure of 1 atm*

$$\begin{aligned} H_i^L &= 16.67e^{-1.087x_i} \\ H_i^V &= 13.49e^{-3.98x_i} + 43.97e^{-0.068x_i} \end{aligned}$$

*Reference state: Pure components as liquid at 0°C. x_i denotes mole fraction methanol in liquid phase.

$$\left(\frac{\partial y_B}{\partial D}\right)_V(s) = k_{y_B D}^{DV} \frac{n_2(s)}{d(s)}; \quad \left(\frac{\partial y_T}{\partial D}\right)_V(s) = k_{y_D D}^{DV} \frac{n_3(s)}{d(s)} \quad (34)$$

since all the transfer functions share the pole-polynomial $d(s)$. Finally, inserting Eqs. 29, 33, and 34 into Eq. 28 yields

$$(\partial y_D / \partial L)_V(s) = \frac{k_{y_D D}^{DV} (\tilde{H}_T^V)^2 n_1(s)}{V \left[\left(\frac{d\tilde{H}^V}{dy}\right)_{y=y_B} k_{y_B D}^{DV} \tilde{H}_T^V n_2(s) - \left(\frac{d\tilde{H}^V}{dy}\right)_{y=y_D} k_{y_D D}^{DV} \tilde{H}_B^V n_3(s) \right] - (\tilde{H}_T^V)^2 d(s)} \quad (35)$$

Applying Routh-Hurwitz stability criterion to the pole-polynomial in Eq. 35 yields:

$$V \frac{\left(\frac{d\tilde{H}^V}{dy}\right)_{y=y_B} k_{y_B D}^{DV} \tilde{H}_T^V - \left(\frac{d\tilde{H}^V}{dy}\right)_{y=y_D} k_{y_D D}^{DV} \tilde{H}_B^V}{(\tilde{H}_T^V)^2} > 1 \quad (36)$$

as a sufficient condition for instability with the LV configuration. Comparing Eqs. 36 and 33 finally yields that instability will occur if $(\partial L / \partial D)_V > 0$ at steady state.

Remarks. (1) In the above derivation we have assumed a binary mixture such that the saturated vapor enthalpy, \tilde{H}^V , is a function of one composition only (pressure is assumed constant) as shown in Eq. 32. We have also selected the reference state for energy such that it is reasonable to set the liquid enthalpy equal to zero at all stages, that is, $H_i^L = 0$ (see Appendix for details). This assumption is very good for many mixtures.

(2) From the exact steady-state balances $D = V_T - L = F + V - L_B$ we derive for F constant the following equivalent steady-state conditions for instability:

$$\left(\frac{\partial L}{\partial D}\right)_V > 0 \Leftrightarrow \left(\frac{\partial V_T}{\partial L}\right)_V > 1 \Leftrightarrow \left(\frac{\partial L_B}{\partial L}\right)_V < 0 \quad (37)$$

Thus, we find that an unstable operating point corresponds to a situation where at steady state an increase in liquid flow, L , in the top of the column, yields a decrease in liquid flow, L_B , at the bottom of the column (to observe this in practice one would need feedback to stabilize the operating point).

(3) Since $(\partial y_D / \partial D)_V$ usually is negative at steady-state (see Appendix 1 in Jacobsen and Skogestad, 1991; counter examples are given by Jacobsen, 1993), we see from Eq. 28 that the instability condition $(\partial L / \partial D)_V(0) > 0$ corresponds to $k_{y_D L}^{LV} = (\partial y_D / \partial L)_V(0) < 0$. That is, at the unstable operating points we have the unexpected situation where the separation gets worse with increasing reflux. This is in accordance with the numerical results in Figure 6.

(4) In industrial practice it is more common to specify the heat input to the reboiler Q_B rather than the molar boilup $V \approx Q_B / \Delta H^{vap}$. In many cases the molar heat of vaporization ΔH^{vap} is nearly constant and the LQ_B -configuration is very similar to the LV -configuration. However, the instability we find with the LV configuration is related to changes in the heats of vaporization and we therefore need to consider whether the instability persists with the LQ_B configuration. Assuming $(\partial y_D / \partial D)_{Q_B}(s)$ stable and neglecting changes in liquid enthalpy with composition yields:

$$-Q_B \frac{\left(\frac{\partial \tilde{H}^V}{\partial y}\right)_{y=y_T} k_{y_T D}^{DQ_B}}{(\tilde{H}_T^V)^2} > 1 \quad (38)$$

as a sufficient criterion for instability with the LQ_B configuration. According to Eq. 38 instability is unlikely with the LQ_B configuration in the usual case where $d\tilde{H}^V / dy < 0$ and $k_{y_T D}^{DQ_B} < 0$. Thus, while the energy balance may cause instability with the LV configuration it is unlikely to cause instability with the LQ_B configuration. The exception is for cases where $d\tilde{H}^V / dy > 0$ for which Eq. 38 predicts that instability may occur. Note that we have in deriving Eq. 38 neglected changes in liquid enthalpy with composition and that instability with the LQ_B configuration is possible also in the normal case when this assumption is removed. However, we do not include any proof here since instability in this case only is predicted in regions of operation where the internal flows are unrealistically high.

The fact that the energy balance is unlikely to yield instability with the LQ_B configuration does not render the results for the LV -configuration of no interest from a practical viewpoint. First, some industrial columns are effectively operated with molar boilup V as an independent input. This is usually achieved by inferring the boilup rate V from the differential pressure across a column section and manipulating Q_B to keep the boilup constant (Kister, 1990). Secondly, many simulation models use the molar boilup V as a specification rather than the heat input Q_B . Our results show that it may be crucial to choose the correct specification in simulations in order to correctly predict the behavior of the real column.

Effect of operating conditions on stability

From Eq. 36 we see that the probability of instability with the LV configuration will increase with internal flows (that is, V). This is similar to what was found for the instability caused by a singularity in the input transformation with the $L_w V$ configuration.

If we assume ideal vapor phase and neglect the small contribution from vapor heat capacity, we have for a binary mixture:

$$\tilde{H}^V(y) = y\Delta H_1^{vap} + (1-y)\Delta H_2^{vap} \quad (39)$$

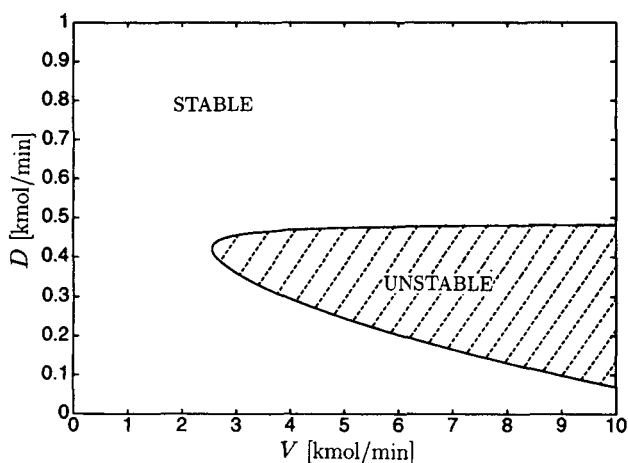


Figure 7. Regions of stable and unstable operating points in terms of distillate flow D and boilup V for methanol-propanol column with LV -configuration.

Energy-balance included.

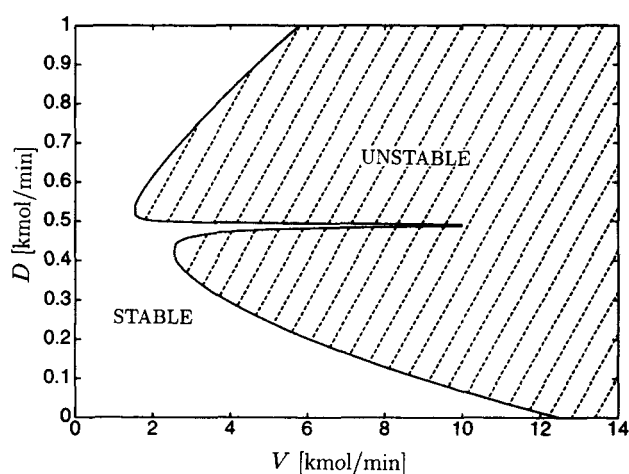


Figure 8. Regions of stable and unstable operating points in terms of distillate flow D and boilup V for methanol-propanol column with L_wV -configuration.

Energy-balance included.

and we derive $d\bar{H}^V/dy = \Delta H_1^{\text{vap}} - \Delta H_2^{\text{vap}}$ which is the difference in heats of vaporization for the light and heavy component at their boiling point. Usually, the most volatile component has the smallest heat of vaporization and we get $d\bar{H}^V/dy < 0$. We also usually have $k_{y_D}^{DV} < 0$ and $k_{y_D}^{DV} < 0$ (Jacobsen and Skogestad, 1991), and we see from Eq. 36 that in this case instability is most likely when $|k_{y_D}^{DV}|$ is large relative to $|k_{y_D}^{DV}|$, which corresponds to having high purity in the top relative to the bottom. Note that this is different from what was found for the instability caused by the transformation between molar and mass reflux for the L_wV configuration where instability was found to be most likely with a relatively impure top product.

Figure 7 shows the stable and unstable regions in terms of boilup V and distillate flow D for the methanol-propanol column with the LV configuration. For low internal flows, that is, low values of V , we see that there is no unstable region and thus a unique stable operating point for all values of L . For values of V above 2.56 kmol/min there exist a region of unstable operating points and, as expected from the analysis above, the unstable region expands with increasing V , that is, increasing internal flows. We also see from Figure 7 that for high values of D , corresponding to low purity in the top relative to the bottom, there are no unstable operating points. This is true even at high values of the internal flows, and is also as expected from the above analysis.

The borders between the stable and unstable regions in Figure 7 correspond to limit-points with the LV configuration and locally there are thus two steady-state solutions around these points.

Note that we in our derivation of Eq. 36 as a sufficient criterion for instability neglected changes in the liquid enthalpy with composition while the region of unstable operating points in Figure 7 was computed using a model with composition dependent liquid enthalpy. Because of this the unstable region is not predicted exactly by the criterion (Eq. 36). However, we note that the instability criterion $(\partial L/\partial D)_V(0) = 0$ predicts the unstable region in Figure 7 exactly. This is not surprising as

the border between the stable and unstable regions corresponds to $(\partial y_D/\partial L)_V(s)$ being singular, that is, it has a single zero eigenvalue at the border, and from Eq. 28 we see that this corresponds to having $(\partial L/\partial D)_V^{-1}(s)$ singular or $(\partial D/\partial L)_V(0) = 0$.

Combining mass flows and energy balance

Jacobsen and Skogestad (1991) show that both types of multiplicities may be present in the same region of operation. They show that a column operating with the L_wV -configuration may have up to five different steady-state solutions, two of which they find to be unstable.

Figure 8 shows the stable and unstable operating regions in terms of boilup V and distillate flow D for the methanol-propanol column with the L_wV configuration and the energy balance included in the model. The unstable region appearing in the upper part of the figure is caused by singularities in the transformation from L_w to L while the unstable region in the lower part is due to the presence of the energy balance. As seen from Figure 8, the two regions coincide at high values of V , that is, high internal flows, and all solutions are therefore unstable for $V > 12.2$ kmol/min.

Note that all the unstable operating points have only a single RHP pole. Some readers might believe that both types of instability may be present in some operating points thereby possibly giving rise to two unstable poles. However, instability with the LV -configuration, due to energy balance effects, corresponds to $(\partial y_D/\partial L)_V(0) < 0$ which implies that instability due to the input transformation is not possible according to Eq. 21 (when $M_1 < M_2$).

Instability with D_wV -Configuration

We have so far only considered using reflux and boilup as independent variables. However, as discussed in the introduction, there are many possible configurations. For instance, changing condenser level control from using distillate D_w to using reflux L_w results in the D_wV -configuration. Jacobsen and

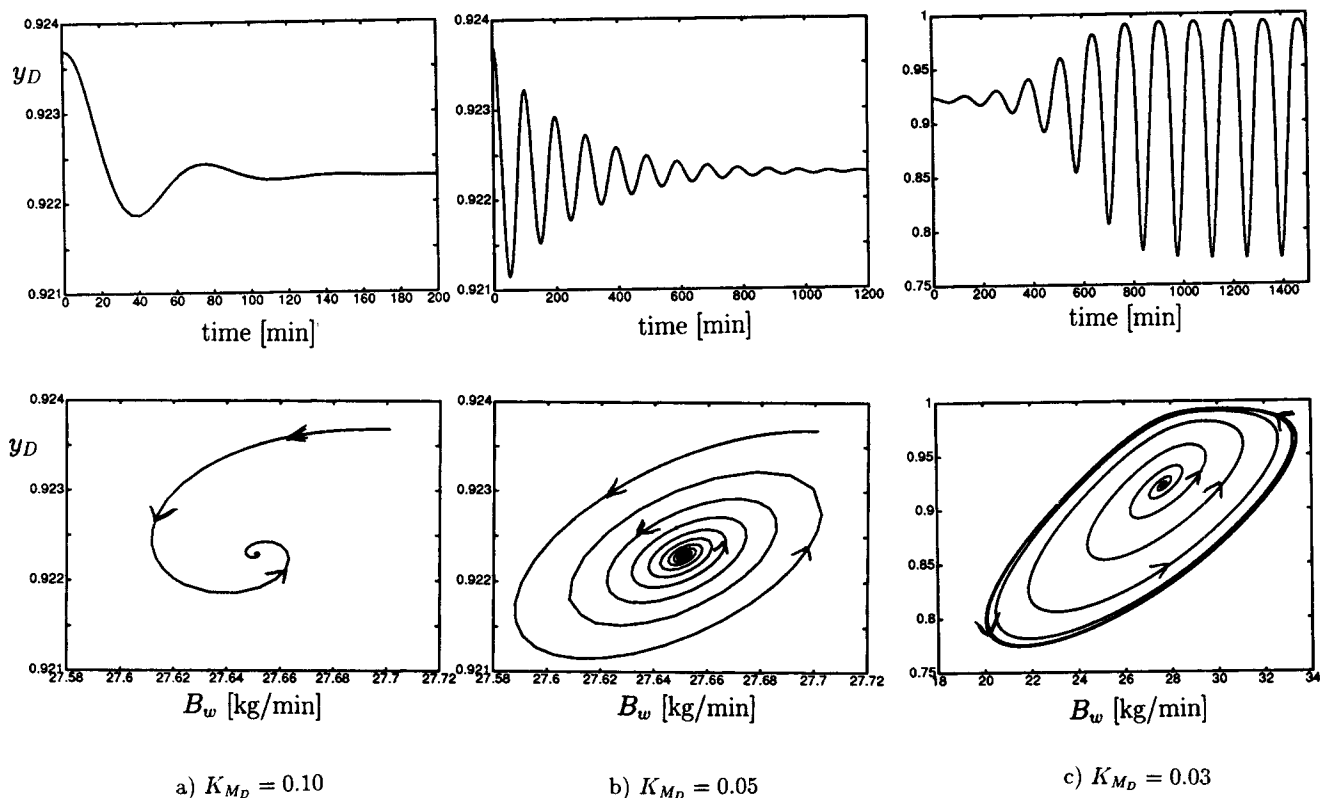


Figure 9. Nonlinear open-loop simulation of methanol-propanol column with D_wV configuration.

Responses to increase in D_w of 0.05 kg/min with different gains K_{M_D} in condenser level controller. a) = $K_{M_D} = 0.10$; b) = $K_{M_D} = 0.05$; c) = $K_{M_D} = 0.03$. Upper plot: time as independent variable, lower plot: phase-plane plot. Boilup $V = 2.0$ kmol/min. Constant molar flows.

Skogestad (1991) show that multiplicity is unlikely with this configuration. Dynamically, with perfect level control, the operating points are found to be asymptotically stable in all examples we have considered. However, here we show that without the assumption of perfect level control an operating point may become unstable also with the D_wV -configuration. We start by considering an example and will then explain the results thereof using analytical results.

Methanol-propanol example

Consider the methanol-propanol column in Table 1. The holdups in the reboiler and condenser are increased to $M_D/F = M_B/F = 5.0$ min. We consider the case with constant molar flows, and use distillate flow D_w and boilup V as independent inputs, that is, D_wV -configuration. With this configuration the condenser level is controlled by reflux L_w and the reboiler level is controlled by bottoms flow B_w . The nominal operating point we consider has $D_w = 18.36$ kg/min and $V = 2.0$ kmol/min. For these specifications we obtain $y_D = 0.9237$ and $x_B = 0.0078$, and the steady state is unique. Note that the operating point corresponds to solution III in Table 2 and Figure 2, that is, the operating point is unstable with the L_wV -configuration.

We now consider the response for different gains K_{M_D} in the condenser level controller. A pure proportional controller is used, that is, $dL_w(s) = K_{M_D} dM_{D_w}(s)$. We assume perfect level control in the reboiler. The upper part of Figure 9a shows the response in top composition y_D to a small increase in D_w , keeping V constant, with the level controller gain $K_{M_D} = 0.10$. We see that the response is stable and slightly oscillatory. The

lower plot in Figure 9a shows the corresponding phase plot for y_D and B_w , and we see that the steady state is a stable spiral attractor (sink). Figure 9b shows the corresponding response with K_{M_D} reduced to 0.05. The response is now more oscillatory, but the steady state is still a stable spiral. With K_{M_D} reduced to 0.03 the operating point becomes an unstable spiral (source) as seen from Figure 9c. However, the response settles into a stable periodic behavior. This implies that there, in addition to the unstable steady state, is a solution corresponding to a stable limit cycle.

The fact that the steady state changes from a stable spiral to an unstable spiral as the level control gain is reduced implies that a pair of complex conjugate eigenvalues cross the imaginary axis. This may be seen from Figure 10a which shows the largest eigenvalues as a function of level control gain K_{M_D} , that is, the root locus. We see that as the gain is reduced below a value of 0.043, the eigenvalues cross the imaginary axis, and the operating point becomes unstable. The fact that a pair of complex conjugate eigenvalues crosses the imaginary axis as K_{M_D} is reduced below a certain value implies that the column undergoes a Hopf bifurcation and a limit cycle results as the steady state becomes stable. The bifurcation is supercritical in this case as the resulting limit cycle is stable (see for example, Guckenheimer and Holmes, 1983).

Conditions for instability

To understand why the steady state for the D_wV configuration becomes unstable, consider the transfer function $(\partial y_D / \partial D_w)_V(s)$ which may be written:

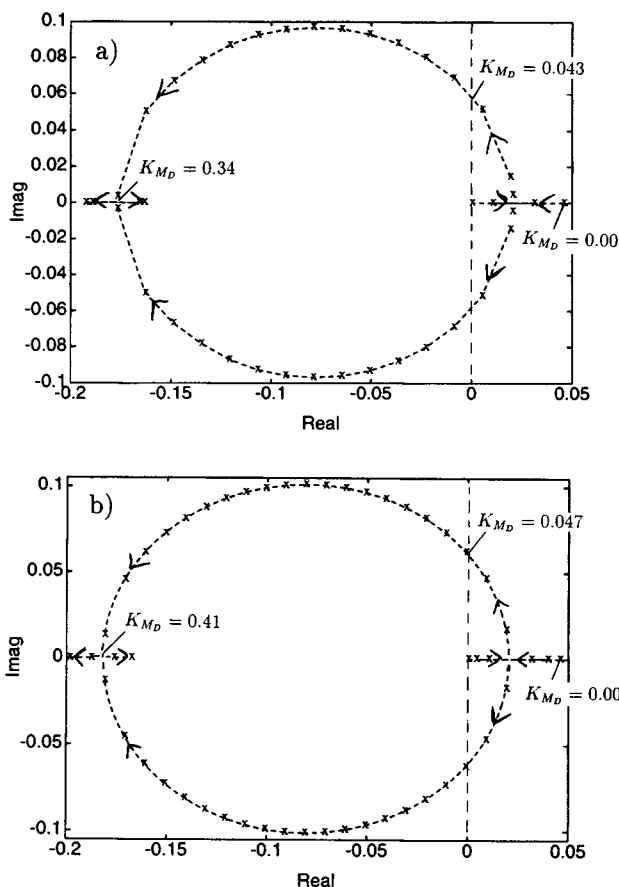


Figure 10. Root locus for methanol-propanol column with constant molar flows and D_wV configuration.

$D_w = 18.36$ kg/min and $V = 2.0$ kmol/min. Plot shows maximum eigenvalues as function of gain K_{M_D} in condenser level controller (arrows indicate increasing gain). a. Eigenvalues computed from full model; b. eigenvalues computed from simple expression (Eq. 45).

$$\left(\frac{\partial y_D}{\partial D_w}\right)_V(s) = \left(\frac{\partial y_D}{\partial L_w}\right)_V(s) \left(\frac{\partial L_w}{\partial D_w}\right)_V(s) \quad (40)$$

Here the transfer function $(\partial y_D/\partial L_w)_V(s)$ expresses the effect of reflux on top composition with the L_wV -configuration. For simplicity we consider only the largest pole in the transfer function:

$$\left(\frac{\partial y_D}{\partial L_w}\right)_V(s) = k_{yD L_w}^{L_w V} \frac{-\lambda_w}{s - \lambda_w} \quad (41)$$

Here λ_w denotes the largest eigenvalue with the L_wV -configuration, and as we have seen it may become positive, either because of the input transformation or because of the energy-balance effects. Here we consider the first case only, that is, we assume constant molar flows. However, similar results are obtained for cases where the instability is caused by the energy balance.

The transfer function $(\partial L_w/\partial D_w)_V(s)$ may be computed from a material balance around the condenser:

$$dL_w(s) = \frac{K_{M_D}}{s} [dV_{T_w}(s) - dL_w(s) - dD_w(s)] \quad (42)$$

Rearranging and using $V_{T_w} = V_T M$ and assuming constant molar flows such that $dV_T = dV$ yields:

$$\left(\frac{\partial L_w}{\partial D_w}\right)_V(s) = \frac{K_{M_D}}{K_{M_D} + s} \left[V_T(M_1 - M_2) \left(\frac{\partial y_T}{\partial D_w}\right)_V(s) - 1 \right] \quad (43)$$

Here y_T denotes the composition of V_T . We consider only the dominant response and get $(\partial y_T/\partial D_w)_V(s) = (\partial y_D/\partial D_w)_V(s)$. Inserting Eqs. 41 and 43 into Eq. 40 yields:

$$\begin{aligned} \left(\frac{\partial y_D}{\partial D_w}\right)_V(s) &= \frac{-\lambda_w k_{yD L_w}^{L_w V} K_{M_D}}{s^2 + (K_{M_D} - \lambda_w)s + \lambda_w K_{M_D} (k_{yD L_w}^{L_w V} V_T(M_1 - M_2) - 1)} \quad (44) \end{aligned}$$

The two eigenvalues (poles) of the transfer function (Eq. 44) become:

$$\begin{aligned} \lambda &= -\frac{1}{2} (K_{M_D} - \lambda_w) \\ &\pm \frac{1}{2} \sqrt{(K_{M_D} - \lambda_w)^2 + 4\lambda_w K_{M_D} [1 - k_{yD L_w}^{L_w V} V_T(M_1 - M_2)]} \quad (45) \end{aligned}$$

Figure 10b shows the root locus for the methanol-propanol column computed using Eq. 45, and we see that the simple expression of this equation yields a reasonable prediction of the behavior of the full model in Figure 10a. The deviation is explained by the assumption of first-order response and negligible condenser holdup in the analytical treatment.

Let us now use Eq. 45 to consider the stability of the D_wV configuration for the two cases when the operating point is stable or unstable with the L_wV configuration.

(1) Stable L_wV -configuration ($\lambda_w < 0$ and $k_{yD L_w}^{L_w V} > 0$): In this case the first term in Eq. 45 is negative for all values of $K_{M_D} > 0$. Furthermore, the second term under the root in Eq. 45 is negative and the root will be real with a value less than $(K_{M_D} - \lambda_w)$, or it will be imaginary. This implies that both eigenvalues in Eq. 45 are in the LHP, that is, the D_wV -configuration is stable for all values of $K_{M_D} > 0$.

(2) Unstable L_wV -configuration ($\lambda_w > 0$ and $k_{yD L_w}^{L_w V} < 0$): In this case the first term in Eq. 45 is positive if $K_{M_D} < \lambda_w$, that is, at least one of the eigenvalues in Eq. 45 are in the RHP with $K_{M_D} < \lambda_w$. The size of K_{M_D} will determine whether the root in Eq. 45 is imaginary. For $K_{M_D} = \lambda_w$, that is, at the bifurcation point, the root is imaginary if $(-k_{yD L_w}^{L_w V} V_T(M_2 - M_1)) > 1$ (note that we also assume $M_2 > M_1$). From Eq. 17 we get with $s = 0$ that $k_{yD L_w}^{L_w V} = k_{yD L}^{L_w V} / [M - L(M_2 - M_1)k_{yD L}^{L_w V}]$, and we find that the root is imaginary if:

$$\frac{V_T(M_2 - M_1)k_{yD L}^{L_w V}}{L(M_2 - M_1)k_{yD L}^{L_w V} - M} > 1 \quad (46)$$

which is always satisfied since the denominator is positive and $V_T \geq L$. Thus, under the assumption of first-order response in

Eq. 41, the bifurcation point will always correspond to a Hopf bifurcation and a limit cycle is born as the steady state becomes unstable. In all examples that we have studied the bifurcation is supercritical, that is, the resulting limit cycle is stable.

We conclude from the above analysis that a prerequisite for instability with the D_wV configuration is that the operating point is unstable with the L_wV configuration. This is not surprising as the level control for the D_wV configuration may be viewed as a feedback effect on the L_wV configuration. If the feedback control is not sufficiently tight then we are not able to stabilize the column. With a gain $K_{M_D}=0$, that is, no condenser level control, we see from Eq. 45 that there will be a RHP pole at λ_w (in addition to a pole at 0), and we effectively have the stability properties of the L_wV -configuration. This may also be seen from the root locus in Figure 10a for $K_{M_D}=0$.

Also other configurations may display the type of instability found with the D_wV configuration above. For instance, it is easily shown that the L_wB_w -configuration becomes unstable if the L_wV configuration is unstable and the reboiler level control is not sufficiently tight. The instability is also in this case resulting from a Hopf bifurcation.

Discussion

Use of volumetric flows

In this article we have only discussed using inputs on a mass basis, for example, reflux L_w in kg/min. In many columns the flows are fixed on a volume basis [m³/min]. If this is the case, and we assume ideal mixing, the molecular weights M_1 and M_2 (for example, in Eq. 21) should be replaced by the molecular volumes $V_1 = M_1/\rho_1$ and $V_2 = M_2/\rho_2$ where ρ_i is the liquid density in kg/m³ of component i . For most mixtures the difference in liquid density between the components is small, and very similar results are obtained with volume flows as those obtained with mass flows.

Industrial columns

The methanol-propanol column that we have studied in this article is not optimally designed for the product compositions at operating point III in Table 2. In fact it is doubtful that the column would be unstable when optimally designed as the internal flows then would be significantly smaller. This is probably true for many separations, that is, an optimally designed column will be open-loop stable. However, few industrial columns are operated close to the optimal operating point. One reason is that the desired compositions will change after the column is built. In addition, many operators prefer to use high internal flows (overpurification) in order to assure that specifications are kept when disturbances enter the column. It is therefore likely that many industrial columns may have problems with open-loop instability. The fact that this has not been reported previously is probably due to the fact that open-loop instability has been believed to be impossible, and problems have therefore been explained by other means. In Jacobsen and Skogestad (1991b) we discuss in more detail the effect of instability on column operation and control.

Is the instability caused by feedback?

All the results in this article are for a partially controlled

system where we assume feedback control of pressure and the two levels. One may ask whether the observed instability is a fundamental property or if it is an artifact caused by these feedbacks. We argue that it is indeed a fundamental property. Consider the L_wV or L_wQ_B configuration for which instability is most likely to occur in a practical situation. It is easily shown that the instability in this case is independent of the level control. Next consider the pressure. First, there are many cases where pressure may be kept constant without the need for feedback. As an example consider a typical lab-scale distillation column with an open vent to the surroundings, and where the pressure is "self-regulated" since the cooling adjusts itself to maintain atmospheric pressure at the top. Secondly, for the case of constant molar flows and constant relative volatility (independent of pressure), our analysis would be the same if we assumed constant cooling duty Q_D rather than constant pressure. This follows since $Q_D = V_T/H_T^V$ (Appendix 1) where H_T^V is independent of composition when we have constant molar flows. Thus, Q_D constant implies that V_T is constant such the vapor flow remains constant up the column. Also, if the relative volatility is independent of pressure, $(\partial y_D/\partial L)_V$ is the same both when p or V_T is constant. In conclusion, the instability for the L_wV -configuration is not caused by feedback control of level and pressure.

On the other hand, for the D_wV configuration the feedback control is critical for the observed results. Indeed, without any condenser level control the D_wV configuration behaves as the L_wV configuration. It is therefore unstable with sufficiently slow level control when the L_wV configuration is unstable, but the level control may stabilize the system, and yield, for example, the stable limit cycle as shown in Figure 9c.

Conclusions

(1) Two-product distillation columns operating with reflux and boilup as independent inputs may be open-loop unstable with a single right half plane pole. Two different effects may cause the instability:

- Singularities in the transformation between the actual input units (mass or volume) and the molar units which determine separation.
 - Singularities between molar flows and compositions due to interactions through the material and energy balance.
- In both cases the probability of instability is increased with increased internal flows.

(2) Distillation columns operating with distillate flow and boilup as independent variables may have unstable operating points if the condenser level control is not sufficiently tight. The instability will in this case correspond to a Hopf bifurcation, that is, a pair of complex conjugate eigenvalues cross the imaginary axis and a limit cycle (usually stable) is born as the steady state becomes unstable. A prerequisite for instability with this configuration is that the operating point is unstable with reflux and boilup as independent variables.

Acknowledgments

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Notation

- B = bottoms flow (kmol/min)
 $d(s)$ = $1 + a_1s + \dots$ - pole polynomial

D = distillate flow (kmol/min)
 F = feed rate (kmol/min)
 H^L = saturated liquid enthalpy (kJ/mol)
 H^V = saturated vapor enthalpy (kJ/mol)
 \tilde{H}^V = saturated vapor enthalpy (kJ/mol) with pure components as saturated liquids at column pressure as reference state
 ΔH^{vap} = heat of vaporization of pure component
 $K_{yu_1}^{u_1 u_2}$ = steady-state gain from input u_1 to output y with u_1, u_2 configuration
 K = slope of equilibrium line
 K_{M_D} = condenser level control gain
 L = reflux flow rate (kmol/min)
 LHP = left half plane
 M = molecular weight, usually of top product (kg/kmol)
 M_1 = molecular weight of most volatile component (kg/kmol)
 M_2 = molecular weight of least volatile component (kg/kmol)
 M_B = reboiler holdup (kmol)
 M_D = condenser holdup (kmol)
 M_i = stage liquid holdup (kmol)
 $n(s)$ = $1 + b_1 s + \dots$ - numerator polynomial
 N = number of theoretical stages in column
 N_F = feed stage location (1-reboiler)
 q_F = liquid fraction in feed
 Q_B = heat input to reboiler (kJ/min)
 Q_D = heat removal in condenser (kJ/min)
 RHP = right half plane
 s = Laplace variable
 V = boilup from reboiler (kmol/min) (determined indirectly by Q_B)
 V_T = vapor flow to condenser (kmol/min)
 x_B = mole fraction of most volatile component in bottom product
 x_i = liquid mole fraction of most volatile component on stage i
 y_B = vapor mole fraction of most volatile component in equilibrium with x_B
 y_D = mole fraction of most volatile component in distillate (top product)
 y_i = vapor mole fraction of most volatile component on stage i
 y_T = mole fraction of most volatile component in V_T
 z_F = mole fraction of most volatile component in feed

Greek letters

α = $(y_i/x_i)/[(1-y_i)/(1-x_i)]$ relative volatility (binary mixture)
 λ = eigenvalue (pole) of system
 λ_{\max} = eigenvalue with largest real part
 τ_L = hydraulic time constant (min)

Subscripts

0 = initial steady-state value
 1 = most volatile component
 2 = least volatile component
 i = stage no.
 w = flow rate in kg/min

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Appendix: Simplification of the Energy Balance

The energy balance on a stage without external heating or cooling is:

$$\frac{dU_i^L M_{Li}}{dt} = V_{i-1} H_{i-1}^V + L_{i+1} H_{i+1}^L - V_i H_i^V - L_i H_i^L + F_i H_i^F \quad (A1)$$

Here subscript i denotes stage-number (reboiler is stage 1), $H^V(y)$ and $H^L(x)$ denotes vapor and liquid enthalpies respectively and H^F feed enthalpy. Vapor holdup has been neglected. We also assume constant pressure p , and we then have $dU_i^L = dH_i^L$.

As the reference state for energy we select the pure components as saturated liquids at pressure p (the column pressure). Note that this means that the reference temperature for each component will be different, since their boiling points generally differ. Then, under the assumptions of no heat of mixing, equal heat capacities for the components and a linear boiling point curve we get $H_i^L = 0$ on all stages (this is a common assumption in distillation which yields "constant molar flows" if we in addition assume the same heat of vaporization for all components). Also assume that the feed is saturated liquid so that $H_i^F = 0$. With these assumptions the energy balance (Eq. 47) becomes:

$$V_{i-1} \tilde{H}_{i-1}^V - V_i \tilde{H}_i^V = 0 \quad (A2)$$

where we have used the notation \tilde{H} to explicitly show that this only holds when the saturated pure liquids are used as reference state. The corresponding energy balance for the entire column then becomes:

$$V_T \tilde{H}_T^V = V_B \tilde{H}_B^V \quad (A3)$$

where $V_B = V$ and V_T represent the vapor flows in the bottom and top of the column, respectively.

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