Structural Stability of Chemical Process Plants

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The problem of analyzing the stability of chemical process plants is considered. The idea of exchange plants has been extended with more general binary transfer terms and binary equilibrium, resulting in the idea of binary exchange-equilibrium plants. Their structural stability is analyzed using results on conservation matrices. It was found that binary exchange-equilibrium plants with no azeotropes are stable irrespective of the connections of their operating units. The general results are illustrated in examples of practical importance: heat-exchanger networks and binary distillation columns with constant molar flows.

Introduction

The general problem of the stability of process plants has been addressed by many authors for many years, and various special cases have been solved. Applying matrix mathematics as early as in 1955, Acrivos and Amundson (1955) investigated the stability of some simple process systems. The stability problems of different models of homogeneous continuous distillation was treated by Doherty and Perkins (1982). An analysis of the effect of recycles on the dynamics, including stability, was reported in Morud and Skogestad (1993) for the simplest one-variable, one-unit case.

The present article can be regarded as an extension of an earlier article on the stability of exchange plants (Hangos and Varga, 1995). There the stability of the exchange plants was guaranteed as a consequence of the conservation of mass or energy forming the basis of their dynamic state-space model. In other words, the first law of thermodynamics (Reid, 1960) (or its mass analog) ensures the stability of exchange plants. The same engineering concept, that is, to apply basic laws of thermodynamics to derive conditions for the stability of process plants, is extended to a more general class of process systems in the present article. The second law of thermodynamics provides constraints for equilibrium relations that are used here to prove stability of binary exchange equilibrium process plants.

Basic Assumptions and Tools

Process plants are usually described by lumped-cascade models for the purpose of their dynamic analysis. This results in a lumped-parameter model of a process plant suitable for analysis. We restrict ourselves to process plants for which the following assumptions hold.

- The models of the operating units are approximated by cascade models, that is, by networks of regions. A region can be regarded as an elementary balancing volume for which lumped dynamic conservation balances are set up. Therefore only one perfectly mixed phase or a collection of phases in physical equilibrium are present in a region. The regions are the elementary dynamic units of the whole processing plant.
- No dynamic effect (for example, delay) caused by the connections (i.e., piping between operating units) is taken into account.
- Submodels of the regions are constructed from first engineering principles, that is, from dynamic balance equations for two extensive quantities: component masses and energy.
 - Constant holdup is assumed in each region.
- All phases involved are assumed to be incompressible with constant pressure. Moreover the temperature and pressure dependence of their physical properties, such as density (ρ) and specific heat (c_P) , are assumed to be negligible.
- Constant molar flow rates are assumed for all convective flows.

The last three assumptions are used mainly for technical reasons: with a constant holdup $H^{(j)}$ in region j a simple

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relation can be established between the extensive conserved quantities $\phi^{(j)}$ and their intensive counterpart $\varphi^{(j)}$ (energy-temperature, component mass-concentration) in the form of

$$\phi^{(j)} = CH^{(j)}\varphi^{(j)},$$

where C=1 for the component mass-concentration and $C=c_P$ for the energy-temperature pair. Moreover, no balance equation is needed for the overall mass in any region. Therefore the dynamic balance equations can be easily transformed into their *potential form* containing only intensive quantities but conserving the structure and physical meaning of the original balance equations.

Engineering model of the elementary dynamic units

The engineering model of the *j*th region consists of K+1 dynamic balance equations for its conserved extensive quantities, rewritten in potential form containing the corresponding intensive quantities denoted by $\varphi^{(j)}$, augmented with a set of algebraic equations describing, for example, equilibrium relations. From the basic assumptions just listed, the dynamic balance equations are in the following general form:

$$\gamma_1^{(j)} \frac{d\varphi^{(j)}}{dt} = -\gamma_2^{(j)} v^{(j)} \varphi^{(j)} + \Psi_{i\text{conv}}^{(j)} + \Psi_{\text{transfer}}^{(j)} + \Psi_{\text{source}}^{(j)}, \quad (1)$$

with

$$\varphi^{(j)} \in X^{(j)} = \{T^{(j)}, c_k^{(j)}, k = 1, ..., K\},$$
 (2)

where t is time, $\gamma_1^{(j)}$ and $\gamma_2^{(j)}$ are constants, $v^{(j)}$ is the flow rate, $T^{(j)}$ is the temperature, and $c_k^{(j)}$ is the concentration of the kth component in the jth region. The variables $(X^{(j)})$ are termed state variables.

In Eq. 1 the term $\Psi_{o\,\text{conv}}^{(j)} = -\gamma_2^{(j)} v^{(j)} \varphi^{(j)}$ stands for the output convective term; $\Psi_{i\,\text{conv}}^{(j)}$ is the input convective transport term; $\Psi_{t\,\text{ransfer}}^{(j)}$ is the transfer rate between regions involving the same type of extensive quantity; C, K are the number of units and components, respectively; and $\Psi_{\text{source}}^{(j)}$ is the source term for the quantity that may depend on all other quantities within and in some cases outside the unit in a nonlinear way (e.g., chemical reaction). Possible forms of these terms are described by additional algebraic equations, and are discussed in the following sections.

Convective transport, connections

In the case of connections representing convective transport between the dynamic elementary units, the single-unit input convective transport term

$$\Psi_{i\text{conv}}^{(j)} = \gamma_2^{(j)} v^{(j)} \varphi_R^{(j)},$$

with φ_B being the inlet value of the quantity, is replaced by

$$\Psi_{i\,\text{conv}}^{(j)} = \gamma_2^{(j)} v^{(j)} \left(\sum_{l=1}^{C} \left[\alpha_l^{(j)} \varphi^{(l)} \right] + \alpha_0^{(j)} \varphi_B^{(j)} \right), \tag{3}$$

with

$$\sum_{l=0}^{C} \alpha_l^{(j)} = 1,$$
 (4)

where the input of the *j*th unit is connected to the mixed stream from the outputs of the other units and from an external input stream $\varphi_B^{(j)}$. Note that the new input convective term in Eqs. 3 and 4 is a convex linear combination of the output convective terms of all the units including the *j*th one, that is, the factors in the linear combination in Eq. 3 fulfill the relation, Eq. 4.

Two special cases of the general connection just given are of great practical importance. In the case of the mixing of the output of the zth and rth unit, the input convective transport term is

$$\Psi_{i,\text{conv}}^{(j)} = \gamma_2^{(j)} v^{(j)} (\alpha_z^{(j)} \varphi^{(z)} + \alpha_r^{(j)} \varphi^{(r)}), \quad \alpha_z^{(j)} + \alpha_r^{(j)} = 1, \quad (5)$$

while in the case of the recycle of the output of the jth unit to its input with recycle ratio $\alpha^{(j)}$

$$\Psi_{i \text{ conv}}^{(j)} = \gamma_2^{(j)} v^{(j)} (\alpha^{(j)} \varphi^{(j)} + (1 - \alpha^{(j)}) \varphi_B^{(j)}). \tag{6}$$

Binary equilibrium relations

Besides dynamic balance equations describing processes taking place in the system, equilibrium relationships between the state variables should also be taken into account. Only process plants with binary equilibrium relations, that is, equilibrium between two phases, will be considered in this article. However, if one has an equilibrium between multiple phases, it can usually be described as a set of pairwise binary equilibrium relations.

This can be the case when more than two phases are present in a region. One of them is assumed to be dynamic, that is, the subject of dynamic balance equations, and all the others are related to it through static algebraic binary equilibrium relations.

Moreover, it is assumed that the equilibrium relates two components at most in the two phases, that is, *only binary mixtures are considered*.

Let us denote the equilibrium value of an intensive quantity belonging to a conserved-extensive quantity by $\varphi_*^{(j)}$ for a phase in the *j*th region; then the binary equilibrium relationship between the *j*th and *l*th phase in the same or two different regions is in the following general form:

$$\varphi_*^{(j)} = \mathbf{N}^{(j,l)}(\varphi_*^{(l)}), \quad \frac{\partial \mathbf{N}^{(j,l)}}{\partial \varphi_*^{(l)}} > 0,$$
 (7)

where $V^{(j,l)}$ is a given nonlinear function.

Transfer-rate expressions

According to the second law of thermodynamics and the expressions and laws on the internal energy and chemical potential (Smith and Van Ness, 1987), the transfer rate of conserved extensive quantity (component mass or energy) between two phases depends on the difference of the related intensive quantities between two phases in two regions con-

nected by a common boundary. Note that multiphase transport processes are usually decomposed into transport processes between two phases, assuming additivity of the transport phenomena.

There are two alternative ways of expressing the transfer term $\Psi^{(j)}_{\text{transfer}}$ in Eq. 1. In the first case the transfer rate is assumed to depend on the difference between the state variables $\varphi^{(j)}$ and $\varphi^{(l)}$ in the jth and lth regions:

$$\Psi_{\text{transfer}}^{(j)} = \sum_{l=1}^{C} K_{\text{transfer}}^{(j,l)}(\varphi^{(l)} - \varphi^{(j)}). \tag{8}$$

The other way of describing the transfer rate is as a function of the difference between the actual $\varphi^{(j)}$ and the equilibrium value $\varphi_*^{(j)}$ of an appropriate intensive quantity in a region, that is,

$$\Psi_{\text{transfer}}^{(j)} = \sum_{l=1}^{C} k_{\text{transfer}}^{(j,l)} (\varphi_{\star}^{(j)} - \varphi^{(j)}). \tag{9}$$

These two cases can be generalized to a unified form of binary transfer rate as follows

$$\Psi_{\text{transfer}}^{(j)} = \sum_{l=1}^{C} K_{\text{transfer}}^{(j,l)}(\varphi^{(l)}, \varphi^{(j)}), \qquad (10)$$

with

$$\frac{\partial \mathbf{K}_{\text{transfer}}^{(j,l)}}{\partial \boldsymbol{\varphi}^{(j)}} < 0, \qquad \frac{\partial \mathbf{K}_{\text{transfer}}^{(j,l)}}{\partial \boldsymbol{\varphi}^{(l)}} > 0, \quad j \neq l$$
 (11)

and

$$\Phi_l^{(j,l)} = \frac{\partial \mathbf{K}_{\text{transfer}}^{(j,l)}}{\partial \boldsymbol{\varphi}^{(l)}}$$
 (12)

Here the nonlinear function $K_{\rm transfer}^{(j,l)}$ is either a linear function as in Eq. 8 or involves the linear relationship with $k_{\rm transfer}^{(j,l)}$ and the nonlinear equilibrium function $V^{(j,l)}$. It is important to note that because of the construction the following relation exists between the transport-rate term of two connected regions:

$$\mathbf{K}_{\text{transfer}}^{(j,l)} = -\mathbf{K}_{\text{transfer}}^{(l,j)};\tag{13}$$

therefore

$$\Phi_l^{(j,l)} = \Phi_i^{(l,j)}, \quad \Phi_i^{(j,l)} < 0, \quad \Phi_l^{(j,l)} > 0, \quad j \neq l.$$
(14)

The case of no binary azeotropes

From the viewpoint of stability analysis, the case of binary mixtures with no binary azeotropes is of great theoretical interest. With no binary azeotropes one can always rearrange Eqs. 7 such that the linearized form of the equation satisfies

$$\varphi_{\star}^{(j)} = \Gamma_{l}^{(j,l)} \varphi_{\star}^{(l)}, \qquad \Gamma_{l}^{(j,l)} \le 1, \tag{15}$$

that is, the (j) index corresponds to the more condensed (liquid) phase.

Let us now investigate the transfer terms connecting the jth and lth regions in this case. The special form of the rate expressions, Eqs. 9-14, is as follows:

$$\Psi_{\text{transfer}}^{(j,l)} = k_{\text{transfer}}^{(j,l)} (\varphi_*^{(j)} - \varphi^{(j)}) = -k_{\text{transfer}}^{(l,j)} (\varphi_*^{(l)} - \varphi^{(l)}). \quad (16)$$

Substituting Eq. 15 into Eq. 16, we obtain

$$\varphi_{*}^{(l)} = \frac{k_{\text{transfer}}^{(j,l)}}{(k_{\text{transfer}}^{(l,j)} + \Gamma_{l}^{(j,l)}k_{\text{transfer}}^{(j,l)})} \varphi^{(j)} + \frac{k_{\text{transfer}}^{(l,j)}}{(k_{\text{transfer}}^{(l,j)} + \Gamma_{l}^{(j,l)}k_{\text{transfer}}^{(j,l)})} \varphi^{(l)}.$$
(17)

With this equation the following relation can be obtained for the terms $\Phi_i^{(j,l)}$ and $\Phi_i^{(j,l)}$:

$$\Phi_{j}^{(j,l)} = -k_{\text{transfer}}^{(l,j)} \frac{k_{\text{transfer}}^{(j,l)}}{(k_{\text{transfer}}^{(l,j)} + \Gamma_{l}^{(j,l)} k_{\text{transfer}}^{(j,l)})} < 0$$
 (18)

$$\Phi_{l}^{(j,l)} = k_{\text{transfer}}^{(j,l)} \frac{k_{\text{transfer}}^{(j,l)} \Gamma_{l}^{(j,l)}}{(k_{\text{transfer}}^{(j,l)} + \Gamma_{l}^{(j,l)} k_{\text{transfer}}^{(l,l)})} > 0.$$
 (19)

Moreover, the relation

$$\Phi_{l}^{(j,l)} + \Phi_{j}^{(j,l)} = k_{\text{transfer}}^{(l,j)} \frac{k_{\text{transfer}}^{(j,l)} (\Gamma_{l}^{(j,l)} - 1)}{(k_{\text{transfer}}^{(j,l)} + \Gamma_{l}^{(j,l)} k_{\text{transfer}}^{(l,j)})} \le 0 \quad (20)$$

also holds in this case.

Conservation matrices

For the readers' convenience some facts related to conservation matrices are briefly summarized here (Hearon, 1963).

Definition 1. A real square matrix $F = \{f_{ij}\}_{i,j=1}^n$ of order n is said to be a column conservation matrix (or a row conservation matrix) if it is a matrix with dominant main diagonal with respect to columns (or rows), that is,

$$|f_{ii}| \ge \sum_{i \ne i} |f_{ij}| = R_i, \quad i = 1, 2, ..., n$$
 (21)

or

$$|f_{ii}| \ge \sum_{j \ne i} |f_{ji}| = C_i, \quad i = 1, 2, ..., n,$$
 (22)

and its elements have the following sign pattern

$$f_{ii} \le 0, \quad f_{ij} \ge 0, \quad i \ne j.$$
 (23)

In the case of proper inequality for every inequality in either Eq. 21 or 22, F is said to be a strict column conservation matrix or a strict row conservation matrix.

It is easy to see from this definition that the sum of two or more column (or row) conservation matrices is also a column (or row) conservation matrix. Lemma 1. Conservation matrices are nonsingular. Furthermore, all the eigenvalues of a conservation matrix have nonpositive (i.e., zero or negative) real parts, while there is no purely imaginary eigenvalue. In other words, conservation matrices are stable matrices, that is, the real parts of all the eigenvalues are negative.

It is important to note that this Lemma states more than the well-known Gershgorin theorem:

Lemma 2. Every eigenvalue of a square matrix F lies in at least one of the discs (Gershgorin discs) with centers f_{ii} and radii:

$$\sum_{i \neq i} |f_{ij}|. \tag{24}$$

Because conservation matrices are nonsingular and have no purely imaginary eigenvalue, they have no zero eigenvalue either.

Binary Exchange-Equilibrium Plants

The concept of exchange plants (Hangos and Varga, 1995) is extended in this section to construct a more general class of process plants: the binary exchange-equilibrium plants. We then analyze their stability.

Definition 2. A process plant satisfying basic assumptions is called a binary exchange-equilibrium plant if

- There is no source term $(\Psi_{\text{source}}^{(j)})$ in any of Eqs. 1,
- There are only binary exchange and equilibrium relations taking place in the system, that is, the transfer term in the form of Eqs. 10 and 11 extended with the equilibrium relationships (Eq. 7), for every $\varphi^{(j)} \in X^{(j)}$, $j = 1, \ldots, C$.

In other words, only convective transport and binary transfer processes with binary equilibrium take place in a binary exchange-equilibrium plant.

Lemma 3. Exchange plants defined in Hangos and Varga (1995) are special cases of binary exchange-equilibrium plants.

This statement is a simple consequence of the definitions: exchange plants contain no source term in Eq. 1, and the transfer term is assumed to be in the simple linear form of Eq. 8.

Linearized state-space model of the elementary dynamic

In order to investigate the stability of binary exchange-equilibrium plants, the properties of their state matrix A in the linearized state equation

$$\frac{dx}{dt} = Ax + Bu \tag{25}$$

should be determined, where x is the state and u is the input vector of the linearized system, while A and B are constant matrices, the state matrix and the input matrix, respectively (Kailath, 1980). A linear system, Eq. 25, is (asymptotically) stable if all eigenvalues of the state matrix A have negative real parts.

The linearized state-space model of the *j*th region can be obtained from the K+1 dynamic balance Eq. 1 by linearizing it around a nominal steady state $\varphi^{(j)} = \varphi_S^{(j)}$ for every $\varphi^{(j)} \in$

 $X^{(j)}$. The entries in the state vector $x^{(j)}$ of the region are in the form of

$$x_i^{(j)} = \varphi^{(j)} - \varphi_{\mathcal{S}}^{(j)}, \tag{26}$$

where the index *i* identifies the state variable $\varphi^{(j)}$. Moreover the state matrix $A^{(j)}$ can be decomposed into two parts

$$A^{(j)} = A^{\operatorname{conv}(j)} + A^{\operatorname{transfer}(j)}, \tag{27}$$

where the matrix $A^{\text{conv}(j)}$ describes the effect of the convective terms and the matrix $A^{\text{transfer}(j)}$ corresponds to the transfer term in Eq. 1. Both of these matrices are diagonal with diagonal elements

$$a_{ii}^{\text{conv}(j)} = -\gamma_2^{(j)} v^{(j)} \left(1 - \alpha_j^{(j)}\right) < 0,$$

$$a_{ii}^{\text{transfer}(j)} = \sum_{l=1}^{C} \Phi_j^{(j,l)} \chi_i^{(j,l)} \le 0,$$
(28)

where $\chi_i^{j,l}$ is an indicator function indicating if there is a transfer of the state variable φ between the phases j and l. Note that the first inequality is obtained by considering the terms containing $\varphi^{(j)}$ in the sum $\Psi_{l\,\text{conv}}^{(j)} + \Psi_{o\,\text{conv}}^{(j)}$ (see Eq. 3) while the latter inequality is just another form of Eq. 18.

Stability of binary exchange-equilibrium plants

The state-space model of the overall binary exchange-equilibrium plant is made from the state-space models of the regions, modified by the effect of their connections. The set of state variables of the overall plant X is the union of all state variables of the elementary dynamic units, that is,

$$X = \bigcup_{j \in C} X^{(j)}. \tag{29}$$

The linearized model of an overall binary exchange-equilibrium plant will be linear because it is created from the linearized state-space models of the regions. The overall state matrix A^E can be decomposed in the same way as the state matrices of the regions

$$A^E = A^{E \text{ conv}} + A^{E \text{ transfer}}$$

A row in the state matrix A^E of the plant corresponds to a linearized dynamic balance equation arising from Eq. 1 with the following matrix elements:

$$a_{kk}^{E \text{ conv}} = -\gamma_2^{(j)} v^{(j)} \left(1 - \alpha_j^{(j)}\right) < 0,$$

$$a_{kk}^{E \text{ transfer}} = \sum_{l=1}^{C} \Phi_j^{(j,l)} \chi_k^{(j,l)} \le 0$$
(30)

$$a_{kr}^{E\, {
m conv}} = \gamma_2^{(j)} v^{(j)} \alpha_l^{(j)} \ge 0, \quad a_{kr}^{E\, {
m transfer}} = \Phi_l^{(j,l)} \chi_k^{(j,l)} \ge 0, \quad (31)$$

where k identifies a state variable $\varphi^{(j)}$ of the jth region and r corresponds to the same type of variable in another, the lth region.

Proposition 1. The state matrix A^E of a binary exchange-equilibrium plant with no azeotropes is a sum of two row conservation matrices.

Proof. We shall show that both $A^{E \text{ conv}}$ and $A^{E \text{ transfer}}$ are row conservation matrices.

It is easy to see from the elements of the matrices (Eqs. 30 and 31) that the convective term $A^{E \text{ conv}}$ is a row conservation matrix, taking into account Eqs. 3 and 4.

Furthermore it follows from Eqs. 30 and 31 and the properties of the transfer rate term (Eqs. 12, 13, and 14) that the transfer term $A^{E \text{ transfer}}$ is a column conservation matrix.

Moreover, the relation 20 in the no azeotrope case assures that the transfer term $A^{E \text{ transfer}}$ is a row conservation matrix, as well.

An immediate consequence of Proposition 1 and Lemma 3 gives the main result for stability analysis of binary exchange-equilibrium plants.

Corollary 1. A binary exchange-equilibrium plant is always asymptotically stable.

where $K_{\text{transfer}}^{(j,l)} > 0$ is a constant transfer coefficient. Notice that only convective transport and a difference-based transfer term (see Eq. 8) is present in the equation.

State Matrix of the Heat-Exchanger Network. The set of state variables of the overall heat-exchanger network is the set of temperatures in the cells, that is,

$$X = \bigcup_{j \in C} T^{(j)},\tag{33}$$

where the number of the regions C = 2N, and N is the total number of heat-exchanger cells in the heat-exchanger network.

Let us rearrange the indices of the regions forming the overall heat-exchanger network so that the pair forming a heat-exchanger cell is assigned neighboring indices, that is, l=j+1. Moreover, let us assume general connections between the cells described by Eqs. 3 and 4. Then the convective transport $A^{E\, {\rm conv}}$ and transfer $A^{E\, {\rm transfer}}$ state matrices are in the following form.

$$A^{E \text{ transfer}} = \begin{bmatrix} -K_{\text{transfer}}^{(1,2)} & K_{\text{transfer}}^{(1,2)} & \cdots & 0 & 0 \\ K_{\text{transfer}}^{(1,2)} & -K_{\text{transfer}}^{(1,2)} & 0 & \cdots & 0 \\ \cdots & \cdots & \cdots & \cdots & \cdots \\ 0 & \cdots & 0 & -K_{\text{transfer}}^{(2N-1,2N)} & K_{\text{transfer}}^{(2N-1,2N)} \\ 0 & \cdots & 0 & K_{\text{transfer}}^{(2N-1,2N)} & -K_{\text{transfer}}^{(2N-1,2N)} \end{bmatrix}$$
(34)

Examples

The notion and stability results for binary exchange-equilibrium plants are illustrated in two practically important cases:

- Heat-exchanger networks;
- Binary distillation columns in the case of constant molar flows, both with the equilibrium and the rate expression form for the transfer term therein.

Heat-exchanger network

Heat-exchanger networks can be regarded as the simplest process systems satisfying our basic assumptions where only the energy transport equations form the engineering model. Moreover, heat-exchanger networks are shown to be exchange plants, so they are binary exchange-equilibrium plants as well.

Engineering Model of the Heat-Exchanger Cell. A heat-exchanger cell is a pair of regions connected by a heat-conducting wall. By our basic assumptions, the engineering model of the jth region connected to the lth one forming jointly a heat-exchanger cell is the potential form of the energy transport equation for the temperature $T^{(j)}$ of the region in the form of

$$\gamma_1^{(j)} \frac{dT^{(j)}}{dt} = -\gamma_2^{(j)} v^{(j)} T^{(j)} + \gamma_2^{(j)} v^{(j)} T_B^{(j)} + K_{transfer}^{(j,l)} (T^{(l)} - T^{(j)}), \quad (32)$$

$$A^{E \text{ conv}} = \begin{bmatrix} A_D^{(1)} & A_{OD}^{(1,2)} & \cdots & A_{OD}^{(1,N)} \\ A_{OD}^{(2,1)} & A_D^{(2)} & \cdots & A_{OD}^{(2,N)} \\ \cdots & \cdots & \cdots & \cdots \\ A_{OD}^{(N,1)} & A_{OD}^{(N,2)} & \cdots & A_D^{(N)} \end{bmatrix},$$
(35)

where the diagonal blocks in the the convective matrix term are

$$A_{D}^{(j)} = \begin{bmatrix} -\gamma_{2}^{(j)} v^{(j)} \left(1 - \alpha_{j}^{(j)}\right) & \gamma_{2}^{(j)} v^{(j)} \alpha_{j+1}^{(j)} \\ \gamma_{2}^{(j+1)} v^{(j+1)} \alpha_{j}^{(j+1)} & -\gamma_{2}^{(j+1)} v^{(j+1)} \left(1 - \alpha_{j}^{(j+1)}\right) \end{bmatrix}.$$

Note that the diagonal blocks just given are usually diagonal matrices because the convective connection between the cold and hot side of the same heat-exchanger cell is a rare exception. Furthermore the off-diagonal blocks $A_{OD}^{(i,j)}$ have elements that are equal either to 0 or to a connection term $\gamma_2^{(j)}v^{(j)}\alpha_l^{(j)}$, where l=i or l=i+1.

It is easy to see from the preceding equations and matrices that the convective transport state matrix $A^{E\, {
m conv}}$ is a row conservation matrix, while the transfer state matrix $A^{E\, {
m transfer}}$ is a diagonal matrix with negative entries; therefore, the overall state matrix is a stable matrix. This implies that heat-exchanger networks satisfying our modeling assumptions are always asymptotically stable.

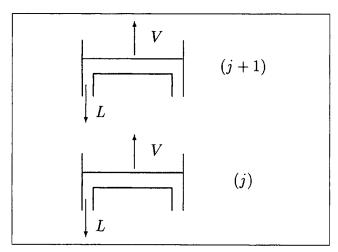


Figure 1. Distillation trays.

Binary distillation column with constant molar flows and vapor – liquid equilibrium

The elementary engineering unit of a distillation column is a real or theoretical distillation tray that is a two-phase perfectly mixed subsystem with vapor and liquid phases. In the case of binary distillation the composition of the binary mixture can be characterized by one mole fraction (for example, the mole fraction of the more volatile component) in both the liquid (ξ) and the vapor (η) phases. The distillation tray with constant molar flow is shown on Figure 1, where H_L is the liquid phase and H_V is the vapor phase molar holdup on the tray, L is the liquid, and V is the vapor molar flow.

Engineering Model of the Distillation Tray. If vapor-liquid equilibrium can be assumed on each tray and the vapor phase holdup is negligible compared to the liquid-phase holdup,

$$\eta^{(j)} = N(\xi^{(j)}),$$
(37)

where

$$\Gamma^{(j)} = \frac{\partial N(\xi^{(j)})}{\partial \xi^{(j)}} > 0 \tag{38}$$

for the jth tray, that is, for the jth region.

State Matrix of the Distillation Column. The set of state variables of the binary distillation column with constant molar flows is the set of liquid-phase mole fractions on the trays, that is.

$$X = \bigcup_{j \in C} \xi^{(j)},\tag{39}$$

where the number of the regions C = N, and N is the number of the trays.

Substituting Eq. 37 into the transport equation, Eq. 36, and linearizing the equations around the steady-state equilibrium points on the trays, the following state matrices result

$$A^{E \text{ conv}} = \begin{bmatrix} -\frac{L}{H_L} & \frac{L}{H_L} & 0 & \cdots & 0 \\ 0 & -\frac{L}{H_L} & \frac{L}{H_L} & \cdots & 0 \\ 0 & \cdots & \cdots & 0 & 0 \\ \cdots & \cdots & 0 & -\frac{L}{H_L} & \frac{L}{H_L} \\ 0 & \cdots & \cdots & 0 & -\frac{L}{H_L} \end{bmatrix}$$
(40)

$$A^{E \, \text{transfer}} = \begin{bmatrix} -\frac{V}{H_L} \Gamma^{(1)} & 0 & 0 & \cdots & 0 \\ \frac{V}{H_L} \Gamma^{(2)} & -\frac{V}{H_L} \Gamma^{(2)} & 0 & \cdots & 0 \\ 0 & \cdots & \cdots & \cdots & 0 \\ \cdots & \cdots & \frac{V}{H_L} \Gamma^{(N-1)} & -\frac{V}{H_L} \Gamma^{(N-1)} & 0 \\ 0 & \cdots & \cdots & \frac{V}{H_L} \Gamma^{(N)} & -\frac{V}{H_L} \Gamma^{(N)} \end{bmatrix}. \quad (41)$$

then the liquid and vapor phases of a tray form a region in the dynamic model of the column with the liquid phase as the dynamic one. Moreover, because of the molar flow conditions, only the component transport equation for the mole fraction of the mole volatile component need be considered together with the equilibrium relationship

$$H_L \frac{d\xi^{(j)}}{dt} = L(\xi^{(j+1)} - \xi^{(j)}) + V(\eta^{(j-1)} - \eta^{(j)})$$
 (36)

It can be seen from the preceding matrices that they both are row conservation matrices; therefore, the binary distillation column with vapor-liquid equilibrium is always asymptotically stable.

Binary distillation column with constant molar flows in the nonequilibrium case

If equilibrium on the trays cannot be assumed, then another dynamic equation describing the component transport

in the vapor phase should be considered instead of the equilibrium relationship, Eq. 37. Therefore two regions are needed to describe the behavior of a tray, one for the liquid, and one for the vapor phase. The transfer between the phases is described by transfer rate expressions.

Engineering Model of the Distillation Tray. Because of the molar flow conditions, only the component transport equations for the mole fraction of the more volatile components in both phases have to be considered together with the equilibrium relationship

$$H_L \frac{d\xi^{(j)}}{dt} = L(\xi^{(j+1)} - \xi^{(j)}) + k_{L \text{transfer}}^{(j)}(\xi_*^{(j)} - \xi^{(j)})$$
 (42)

where the number of regions C = 2N, and N is the total number of trays in the column.

Let us rearrange the indices of the regions forming the distillation column so that the pair forming a tray is assigned neighboring indices that is, l = j + 1. Moreover, let us assume that the mixture forms no binary azeotrope; therefore, Eqs. 15-20 can be assumed with the simplified notation

$$k_{\text{transfer}}^{(j,l)} = k_{L\,\text{transfer}}^{(j)}, \quad k_{\text{transfer}}^{(l,j)} = k_{V\,\text{transfer}}^{(l)}, \quad \Gamma^{(j,l)} = \Gamma^{(j)}.$$
 (47)

Substituting Eq. 44 into the transport equations, Eqs. 42 and 43, and linearizing the equations around the steady-state equilibrium points on the trays, the following state matrices result

$$A^{E\text{conv}} = \begin{bmatrix} -\frac{L}{H_L} & 0 & \frac{L}{H_L} & 0 & \cdots & 0 & 0 \\ 0 & -\frac{V}{H_V} & 0 & 0 & \cdots & 0 & 0 \\ 0 & 0 & \cdots & \cdots & 0 & 0 & 0 \\ 0 & \cdots & \cdots & 0 & 0 & -\frac{L}{H_L} & 0 \\ 0 & \cdots & \cdots & 0 & \frac{V}{H_V} & 0 & -\frac{V}{H_V} \end{bmatrix}$$
(48)

$$A^{E \text{ transfer}} = \begin{bmatrix} \Phi_1^{(1,2)} & \Phi_2^{(1,2)} & 0 & \cdots & 0 \\ \Phi_1^{(2,1)} & \Phi_2^{(2,1)} & 0 & \cdots & 0 \\ 0 & \cdots & \cdots & 0 & 0 \\ \cdots & \cdots & 0 & \Phi_{2N-1}^{(2N-1,2N)} & \Phi_{2N}^{(2N-1,2N)} \\ 0 & \cdots & \cdots & \Phi_{2N-1}^{(2N,2N-1)} & \Phi_{2N}^{(2N,2N-1)} \end{bmatrix}.$$
(49)

 $H_{V} \frac{d\eta^{(l)}}{dt} = V(\eta^{(l-1)} - \eta^{(l)}) + k_{V \text{transfer}}^{(l)}(\eta_{*}^{(l)} - \eta^{(l)})$ (43)

$$\eta_{\star}^{(l)} = \mathcal{N}(\xi_{\star}^{(j)}),\tag{44}$$

with the usual nonnegativity condition, Eq. 38, for the jth liquid-phase and for the lth vapor-phase region on the tray and with

$$\Psi_{\text{transfer}}^{(j)} = k_{L \, \text{transfer}}^{(j)} (\xi_{*}^{(j)} - \xi_{*}^{(j)})
= -k_{L \, \text{transfer}}^{(l)} (\eta_{*}^{(l)} - \eta_{*}^{(l)}) = -\Psi_{\text{transfer}}^{(l)}.$$
(45)

State Matrix of the Distillation Column. The set of state variables of the distillation column is the set of mole fractions in the liquid phase and vapor phase of the trays, that is,

$$X = \left(\bigcup_{j \in N} \xi^{(j)}\right) \bigcup \left(\bigcup_{j \in N} \eta^{(j)}\right),\tag{46}$$

It can be seen from the preceding matrices that they both are row-conservation matrices, therefore the binary distillation column with no azeotrope is always asymptotically stable.

Conclusions

In this article, the stability of a certain class of process plants, exchange plants with binary transfer of mass or energy, has been established using the properties of conservation matrices. The generalization of previous results (Hangos and Varga, 1995) permits a broader range of plants to be considered, as illustrated by the distillation column examples presented in this article. Further generalization of the approach presented here to handle multicomponent transfer and chemical reaction is currently under investigation.

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