Data inputs and interchange in integrated solution/design tower packages

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Literature Cited

Avriel, M.; Dembo, R. S.; Passy, U. *Int. J. Numer. Methods Eng.* **1975**, *9*, 149-168.
Barr, A. J.; Goodnight, J. H.; Sall, J. P.; Helwig, J. T. "A User's Guide to SAS", Sparks Press, 1976, p 76.
Blau, G. E.; Wilde, D. J. *Can. J. Chem. Eng.* **1969**, *47*, 317-326.
Book, N. L. Ph.D. Thesis, University of Colorado, 1976.
Dembo, R. S.; Avriel, M. *Math. Programming* **1978**, *15*, 12-15; Professor Dembo is now at Yale University, New Haven, CT.
Dembo, R. S. "User's Guide to GGP", McMaster University, Hamilton, Ont., Canada, 1975.
Dembo, R. S. *J. Opt. Theory Appl.* **1978**, *26*(2), 149-183.
Duffin, R. J.; Peterson, E. L.; Zener, C. M. "Geometric Programming", Wiley: New York, 1967.
Ecker, J. G.; Wiebking, R. D. *J. Opt. Theory Appl.* **1978**, *26*(2), 305-323.
Foust, S. F.; Wenzel, L. A.; Clump, C. W.; Maus, L.; Anderson, L. B. "Principles of Unit Operations", Wiley: New York, 1960.
Grant, L. G.; Ireson, W. G. "Principles of Engineering Economy", 5th ed.; Ronald Press: New York, 1970.
Guthrie, K. M. "Process Plant Estimating, Evaluation and Control", Craftsman Book Co., Solana Beach, CA, 1970.

Hoerner, D. R., Jr. M.S. Thesis University of Missouri-Rolla, Rolla, MO, Aug 1979.
McCabe, W. L.; Smith, J. C. "Unit Operations of Chemical Engineering", 3rd ed., McGraw-Hill: New York, 1976.
Passy, U.; Wilde, D. J. *SIAM J. Appl. Math.* **1967**, *15*, 1344-1356.
Perry, R. H.; Chilton, C. H. "Chemical Engineers' Handbook", 5th ed.; McGraw-Hill: New York, 1973.
Peters, M. S.; Timmerhaus, K. D. "Plant Design and Economics for Chemical Engineers"; 2nd ed.; McGraw-Hill: New York, 1968.
Rijckaert, M. J.; Martens, X. M. *AIChE J.* **1974**, *20*, 742-750.
Rijckaert, M. J.; Martens, X. M. *J. Opt. Theory Appl.* **1978a**, *26*(2), 205.
Rijckaert, M. J.; Martens, X. M. *J. Opt. Theory Appl.* **1978b**, *26*(2), 325-337.
Sawistowski, H. "Flooding Velocities in Packed Columns at Reduced Pressures", *Chem. Eng. Sci.* **1957**, *6*, 138-140.
Smith, B. D. "Design of Equilibrium Stage Processes", McGraw-Hill: New York, 1963.
Umeda, T. *Ind. Eng. Chem. Process Des. Dev.* **1969**, *8*, 308-316.

Data Inputs and Interchange in Integrated Solution/Design Tower Packages

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Generalized equations are derived allowing fast calculation of the degrees of freedom of single or multiple interconnected tower systems. Moreover, the external data requirements for the stage design correlations are identified and their significance on the overall column design is evaluated. In the process of analysis the phase property requirements for the stage-to-stage and stage design correlations are identified and the column operation and stage performance data to be interchanged are listed.

Introduction

A staged operations problem is completely defined when a unique solution of its stage-to-stage and stage design correlations exists. Such solution requires a number of process variables and design parameters to be externally fixed, phase property correlations to be provided, and finally, column operation and stage performance data to be interchanged (Economopoulos, 1978a).

Enumeration methods for calculating the degrees of freedom have been described by Dunstan (1938), Gilliland and Reed (1942), Kwauk (1956), and Howard (1967) and appear in textbooks, as in Smith (1963). The enumeration procedure, however, is cumbersome and for this reason is rarely used in practice. Moreover, the analysis has been limited to the stage-to-stage calculations, with the stage pressure drops and holdups reported as degrees of freedom and the stage efficiencies ignored.

In the present study, generalized equations are developed allowing direct calculation of the degrees of freedom of individual towers with any number of material and heat leaks, as well as multiple interconnected tower systems. In the process of analysis the internal system relationships (equations of the mathematical model), as well as the required physical properties and stage performance characteristics, are identified and listed. The parameters to be externally fixed for the stage design correlations are identified and their significance to the overall column design evaluated. The required column operation and physical property data are also listed.

Through the above process, the required external specifications for and data interchanges among the stage-to-stage and stage design correlations are established.

Process Variables and Relationships

A staged operations problem is completely defined when the total number of variables in the process is equal to the total number of independent relationships which can be written for the process. The latter is the sum of the internal system relationships (equations of the mathematical model) and the externally imposed specifications. Thus

$$Nv = Ni + Nf \tag{1}$$

The total number of variables involved in a process composed of U units is given by equation

$$(Nv)_p = (Nr)_p + \sum_{u=1}^{U}(Nv)_u \tag{2}$$

where $(Nr)_p$ accounts for the decisions to be made regarding the number of units in the process and the location of the incoming and outgoing material and heat leaks.

The number of internal relationships which can be written for the above process is

$$(Ni)_p = (Ni)_{com} + \sum_{u=1}^{U}(Ni)_u \tag{3}$$

where $(Ni)_{com}$ is the number of independent relationships, which are generated when the U units are combined to form the process: (i) one temperature, $(M-1)$ component

Table I. Internal Relationships for Tower Stages (Figure 1)

	no. of relations
component material balances around the stage	M
component distribution relations between phases	M
enthalpy balance around the stage	1
temperature identity between L_s and V_s	1
pressure identities between L_{s+1} and V_s, L_s and V_{s-1}	2
pressure correlation between V_{s-1} and V_s	1
pressure identity between each feed and L_s or V_s	$(IF)_s$
composition, pressure, and temperature identity between each side product and L_s or V_s	$(ISP)_s(M + 1)$

$$(Ni)_s = 2M + 5 + (IF)_s + (ISP)_s(M + 1)$$

Figure 1. Diagram of a generalized tower stage.

Table II. Internal Relationships for Total Condensers (Figure 2)

	no. of relations
component material balances around the condenser	M
enthalpy balance around the condenser	1
composition, temperature and pressure identities between L_{N+1} and D	$M + 1$
pressure correlation between V_N and D	1
total	$(Ni)_c = 2M + 3$

Figure 2. Diagram of a total condenser.

Table III. Internal Relationships for Total Condenser/Decanter Systems (Figure 3)

	no. of relations
component balances around the system	M
enthalpy balance around the system	1
composition identity between L^1_{N+1} or L^u_{N+1} and D	$M - 1$
component distribution relationships between layers	M
pressure and temperature identities between L^u_{N+1}, L^1_{N+1} and D	4
pressure relationship between L_e and D	1
pressure relationship between V_N, V'_N, and D	2
total	$(Ni)_{c/d} = 3M + 7$

Figure 3. Diagram of a total condenser/decanter system.

fractions and one flow rate identities for each interstream between two units; assuming that the U units are interconnected by a total of θ such interstreams, we have a total of $\theta(M + 1)$ identities; (ii) a total of $(U - 1)$ independent pressure correlations between the U interconnected units. Indeed, as we will see, the pressures of all material streams in each unit are related through the internal system relationships listed in Tables I to III, leaving only one independent pressure to be specified for each unit. Thus, having U units in the process, we can only establish $(U - 1)$ independent pressure correlations between all interstreams. Thus

$$(Ni)_{com} = \theta(M + 1) + U - 1 \tag{4}$$

Introducing eq 2, 3, and 4 into eq 1 and solving for Nf we get

$$(Nf)_p = (Nr)_p + \sum_{u=1}^{U} [(Nv)_u - (Ni)_u] - \theta(M + 1) - U + 1 \tag{5}$$

or

$$(Nf)_p = (Nr)_p + \sum_{u=1}^{U} (Nf)_u - \theta(M + 1) - U + 1 \tag{6}$$

Equation 1 can be used for calculating the degrees of freedom of simple units, such as stages and total condensers. Equation 6 is most suitable for calculating the degrees of freedom of systems composed of interconnected units.
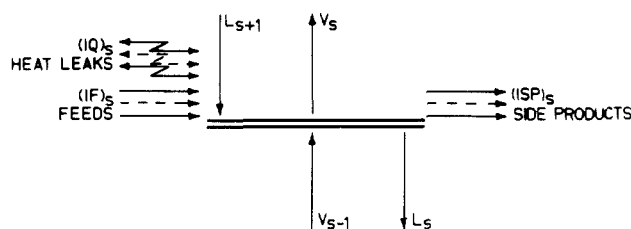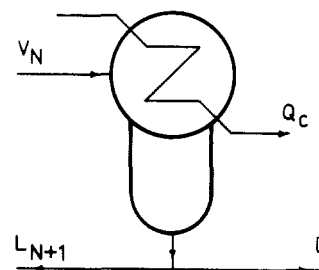
Generalized Diagrams

Figures 1, 2, and 3 present the generalized diagram of a stage, condenser, and condenser/decanter system, respectively. The material and heat leaks account for all external factors affecting the material and enthalpy balances around these units.

It can be observed that multiple feed, side product, and heat streams are attached into each stage. This introduces the parameters $(IF)_s$, $(ISP)_s$, and $(IQ)_s$ into the analysis, and these, through eq 19 to 21 give rise as we will see to the variables $(IF)_t$, $(ISP)_t$, and $(IQ)_t$. This allows the derivation of equations which apply directly to staged operations with any number of material and heat leaks.

The diagram of Figure 4 presents a generalized tower comprising N stages, each of which is identical with that illustrated by Figure 1. Reboilers and partial condensers, if any, are counted as normal stages. This diagram can be used to represent any countercurrent absorption and extraction unit as well as distillation columns with partial condensers.

Adding the condenser of Figure 2 or the condenser/decanter of Figure 3 into the column of Figure 4, we obtain
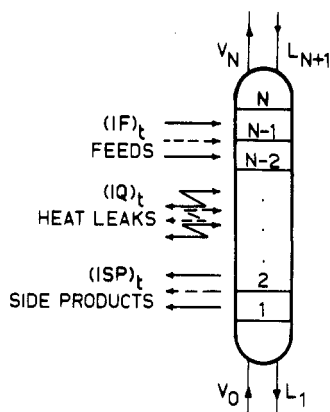
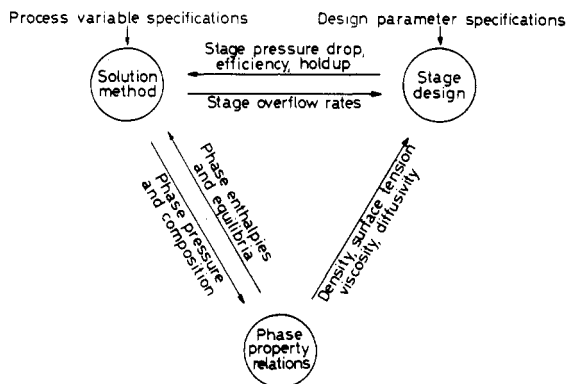**Figure 4.** Diagram of a generalized tower.

Figure 5. Diagram of data inputs and interchanges.

generalized diagrams for distillation columns equipped with total condenser or condenser/decanter systems.

Degrees of Freedom of Simple Units

A material stream is completely defined at a given point when its flow rate, pressure, temperature, and composition are fixed. Hence, the number of associated process variables are

$$(Nv)_m = M + 2 \qquad (7)$$

Rate alone specifies completely a heat stream; thus

$$(Nv)_q = 1 \qquad (8)$$

The units illustrated in Figures 1, 2, and 3 are completely defined when all process variables associated with their material and heat leaks are defined. Thus

$$(Nv)_s =$$
$$4(M + 2) + (IQ)_s + (IF)_s(M + 2) + (ISP)_s(M + 2) \qquad (9)$$

$$(Nv)_c = 3(M + 2) + 1 \qquad (10)$$

$$(Nv)_{c/d} = 6(M + 2) + 1 \qquad (11)$$

When the phase properties and column performance characteristics listed in Figure 5 are given, the independent relationships for the generalized stage, condenser and condenser/decanter systems listed in Tables I, II, and III, respectively, can be formulated. Hence

$$(Ni)_s = 2M + 5 + (IF)_s + (ISP)_s(M + 1) \qquad (12)$$

$$(Ni)_c = 2M + 3 \qquad (13)$$

$$(Ni)_{c/d} = 3M + 7 \qquad (14)$$

Substituting eq 9 and 12, 10 and 13, 11 and 14 into eq 1, we get

$$(Nf)_s = 2M + 3 + (IQ)_s + (IF)_s(M + 1) + (ISP)_s \qquad (15)$$

$$(Nf)_c = M + 4 \qquad (16)$$

$$(Nf)_{c/d} = 3M + 6 \qquad (17)$$

Degrees of Freedom of Selected Tower Systems

The generalized tower systems illustrated in Figure 4 comprises $U = N$ units (stages) interconnected through a total of $\theta = 2(N - 1)$ interstreams. Using eq 15 to calculate the degrees of freedom of each unit, eq 6 yields

$$(Nf)_t = 2M + 3 + (Nr)_t + (IQ)_t + (ISP)_t + (IF)_t(M + 1) \qquad (18)$$

where

$$(IQ)_t = \sum_{s=1}^{U} (IQ)_s \qquad (19)$$

$$(ISP)_t = \sum_{s=1}^{U} (ISP)_s \qquad (20)$$

$$(IF)_t = \sum_{s+1}^{U} (IF)_s \qquad (21)$$

The variable $(Nr)_t$ in this case accounts for all decisions to be taken with regard to the number of stages in the tower and the location of feed, side product and heat streams. Thus

$$(Nr)_t = LN + LQ + LSP + LF \qquad (22)$$

A generalized tower/total condenser system is formed by two simpler units ($U = 2$), a tower and a condenser, sharing a total of two interstreams, V_N and L_{N+1}. Equation 18 yields the degrees of freedom of the tower and eq 16 those of the condenser. Equation 6 can be used with $(Nr)_{t/c} = 0$ to yield

$$(Nf)_{t/c} =$$
$$M + 4 + (Nr)_t + (IQ)_t + (ISP)_t + (IF)_t(M + 1) \qquad (23)$$

For the case of tower/condenser/decanter systems encountered in azeotropic distillation, a similar procedure, in which eq 17 is used instead of 16, yields

$$(Nf)_{t/c/d} =$$
$$3M + 6 + (Nr)_t + (IQ)_t + (ISP)_t + (IF)_t(M + 1) \qquad (24)$$

Equations 18, 23, and 24 show that each material stream leaving the system gives rise to 1 degree of freedom. This is because each such stream generates $M + 2$ new process variables, while according to Tables I, II, and III its pressure, temperature, and composition are related to those of other process streams, thus contributing $M + 1$ internal system relationships. Similarly, each process stream entering the system gives rise to $M + 1$ degrees of freedom since only its pressure has been related to that of other process streams according to Tables I, II, and III. Based on this, eq 18, 23, and 24 can be written in a somewhat more general form as follows

$$(Nf)_t = -1 + (Nr)_t + (IQ)_t + (IMO)_t + (IMI)_t(M + 1) \qquad (25)$$

$$(Nf)_{t/c} =$$
$$1 + (Nr)_t + (IQ)_t + (IMO)_{t/c} + (IMI)_{t/c}(M + 1) \qquad (26)$$

$$(Nf)_{t/c/d} =$$
$$1 + (Nr)_t + (IQ)_t + (IMO)_{t/c/d} + (IMI)_{t/c/d}(M + 1) \qquad (27)$$

Degrees of Freedom of Interconnected Tower Systems

In the case of interconnected tower systems, we first calculate the degrees of freedom of each individual tower through eq 25, 26, or 27 with $(Nr)_t$ given by eq 22. Then,
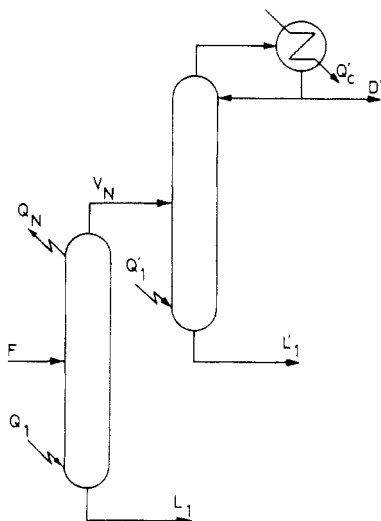
Figure 6. Distillation process with two interconnected towers.

Table IV. Typical Process Specifications for the System of Figure 6

	no. of relations
First Tower	
feedstock rate, temperature, and composition	4
overhead tower pressure	1
internal reflux ratio or Q_N	1
optimality of the feed location	1
two-component splits	2
Second Tower	
overhead tower pressure	1
reflux ratio	1
reflux temperature	1
optimality of the feed location	1
two-component splits	2
total	15

through eq 6 we calculate the degrees of freedom of the entire process with $(Nr)_p = 0$ since no decisions are required about the number of columns in the process.

Consider, for example, the two column distillation system of Figure 6 separating a feed with three components. For the first tower eq 25 is directly applicable with: $M = 3$ (there are three components in the system); $(Nr)_t = 2$ (the number of stages and the location of feed are to be decided); $(IQ)_t = 2$ (the tower has two heat leaks, Q_1 and Q_N); $(IMO)_t = 2$ (the tower has two outgoing material streams, L_1 and V_N); $(IMI)_t = 1$ (the tower has one incoming material stream, F). We thus obtain

$$(Nf)_t = 9 \qquad (28)$$

For the tower/total condenser system a similar procedure, in which eq 26 is used instead of eq 25, yields

$$(Nf)_{t/c} = 10 \qquad (29)$$

Introducing eq 28 and 29 into eq 6 with $\theta = 1$ (only one interstream is shared by the two towers) we get

$$(Nf)_p = 15 \qquad (30)$$

A typical set of design variables can be fixed to have the process completely defined is listed in Table IV.

Data for the Stage Design Correlations

In the design of the tower stages use is made of the column operation and physical property data listed in Figure 5. In addition, a number of design parameters, such as those listed in Table V for tray columns, must be ex-

Table V. Parameters to be Externally Fixed for the Stage Design Correlations

tray column	individual trays
hw	hw
td	td
dh	dh
SF	SF
$(Fw)_{md}$	Fw
$(ts)_{min}$	
$(ts)_{md}$	ts
$(Ff)_{md}$	Ff

ternally fixed (Economopoulos, 1978b).

The parameters ts, Ff, and Fw in the actual column change from tray to tray along with the operating conditions and the phase properties. For this reason, the column design is based on the maximum value these variables are allowed to obtain anywhere in the column, denoted by subscript md in Table V. Usually, ts and Ff obtain their maximum values in the top, while Fw does so in the bottom tray of each tray zone.

Among the eight parameters listed in Table V for tray columns, the first six are related to tray design practice and are not appreciably affected by the stage-to-stage calculations. The last two parameters, however, affect the tower shape and performance and for this reason their value should be fixed so that the most favorable overall design is achieved.

More specifically, hw, td, and dh are fixed on the basis of tray structural and hydraulic stability considerations, SF depends mainly on the foaming tendency of the system, Fw is more like a safety factor reflecting our confidence on the weep point correlations used, while $(ts)_{min}$ sets the minimum tray spacing requirements in the column for tray accessibility and maintenance reasons.

From the remaining two design parameters, $(ts)_{md}$ affects significantly the tower shape and performance. Generally, low values result in short thick columns with relatively low stage pressure drop and somewhat higher efficiency values. The most favorable value of $(ts)_{md}$ varies from problem to problem and depends also on the specified $(ts)_{min}$ value (Economopoulos, 1978b).

The maximum design flood factor, like the weep factor, reflects our confidence on the flood point correlations used. For low pressure operations however, where pressure drop becomes important, it has been shown that lower values of $(Ff)_{md}$ than the maximum permissible ones can yield the most favorable overall design (Economopoulos, papers to be published).

In packed columns the maximum design flood factor should also be fixed on the basis of overall process considerations. In addition, the packing type, size, and material may be also viewed as process variables to be fixed on the basis of overall process considerations since they affect significantly the tower shape and performance.

Summary and Discussion

Equations 25 to 27 along with eq 6 allow fast estimation of the number of process variables to be externally fixed for the stage-to-stage calculations. Similarly, Table V lists the parameters to be externally fixed for the stage design and performance correlations.

In the process of analysis all independent internal relationships for stages, condensers, and condenser/decanter systems were derived and are listed in Tables I, II, and III. Furthermore, all data interchanges among the stage-to-stage, stage design, and phase property correlations have been identified and appear in the diagram of Figure 5.

The data interchanges in Figure 5 suggest the possibility of a significant three-way interaction between the solution

method, the stage design, and performance correlations and the phase properties. This interaction has been largely ignored in practice, for it is common to have the column designed after the stage-to-stage calculations are completed. In two papers to be published, Economopoulos shows that significant design improvements are possible if the stage-to-stage and stage design correlations are simultaneously solved and if $(ts)_{md}$ and $(Ff)_{md}$ are treated as optimization variables along with the relaxed process variables of the solution method.

Nomenclature

D = distillate rate
dh = diameter of the tray perforations
Ff = flood factor
Fw = tray weep factor
hw = weir height
IF = number of feeds
IMI = total number of material streams entering the system
IMO = total number of material streams leaving the system
IQ = number of heat leaks
ISP = number of side products
L = liquid overflow rate
L_e = entrainer make-up rate
LF = number of feeds, the location of which is not specified
LN = variable equal to 1 if the number of tower stages is not specified; equal to 0 otherwise
LQ = number of side products the location of which is not fixed
M = number of components in the system
N = number of tower stages, including reboilers and partial, but not total, condensers
Nf = degrees of freedom
Ni = number of independent internal system relationships
$(Ni)_{com}$ = number of independent relationships arising when units are combined to form the system

Nr = number of decisions regarding the repetition of units and the location of the material and heat leaks
Nv = number of independent variables in the process
Q = rate of a heat leak
SF = system derating factor
td = tray deck thickness
ts = tray spacing
U = number of units in a system
V, V' = vapor overflow rate
θ = number of interstreams between processes

Subscripts
c = condenser
d = decanter
m = material stream
md = maximum design value
min = minimum design value
p = process
q = heat stream
s = stage
t = tower
u = unit

Superscripts
u = upper decanter layer
l = lower decanter layer

Literature Cited

Dunstan, A. W. "The Science of Petroleum", Oxford University Press, 1938.
Economopoulos, A. P. *Chem. Eng. (London)* **1978a**, *No. 330*, 199.
Economopoulos, A. P. *Chem. Eng. (NY.)* **1978b**, *85*, 27, 109.
Gilliland, E. R.; C. E. Reed *Ind. Eng. Chem.* **1942**, *34*, 551.
Howard, G. M. *Ind. Eng. Chem. Fundam.* **1967**, *6*, 86.
Kwauk, M. *AIChE J.* **1956**, *2*, 240.
Smith, B. D. "Design of Equilibrium Stage Processes", McGraw-Hill: New York, 1963.

NOₓ Removal with High-Capacity Metal Oxides in the Presence of Oxygen

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The reaction of nitric oxide with various coprecipitated transition metal oxides was studied in the temperature and concentration range of interest in the control of NO_x from power plants. Differential reaction rates and capacities were determined for alumina-supported cobalt oxide, chromium oxide, and manganese oxide. A high-capacity iron oxide–cobalt oxide sorbent containing sodium has been developed. The total capacity of our BS4 sorbent at 30 °C is greater than 10 g of NO_x/100 g of sorbent. The sorbents are thermally regenerable. Breakthrough curve data were correlated using a previously developed solid–gas reaction model. The model takes into account diffusion and structural changes caused by phase changes.

Introduction

The combustion of any fuel with air as the oxidant, even such fuels as hydrogen, results in the formation of oxides of nitrogen (NO_x). Electric power generation by means of

coal and natural gas accounts for the largest source of NO_x emissions from stationary sources (NAPCA, 1970). Other stationary sources are industrial combustion and gas plants (Bartok et al., 1971). Various methods of control of NO_x