DETERMINING RESIDENCE TIME DISTRIBUTIONS IN COMPLEX PROCESS SYSTEMS

A Simple Method

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In many chemical processing plants, liquid products pass through multiple operations. These processes may include purification, reaction, filtering, blending, and other unit operations. The operations may be batch, continuous, or semicontinuous, and the overall processing may involve batch and continuous units with intermediate storage. In mixed batch and continuous units, batches may be dropped at various times and average residence time calculated from steady state conditions in the continuous component of the system may be invalid. If component batches react in the continuous part of the process, these properties will also be affected by the RTD (residence time distribution) of the product in the process. In addition, the approximation of complex flow patterns in industrial reactors by combinations of simple ideal reactors (*e.g.,* tanks in series, etc.) can be accomplished using the concepts of RTD. The reader can doubtless find other applications where calculating the RTD of a complex system becomes important.

Unless the system consists exclusively of well-mixed, constant-volume tanks and plug-flow elements operating at steady state, a rigorous solution to calculation of the RTD of the systems can be formidable. The problem must be set up in code (FORTRAN, etc.) for each specific flowsheet and is greatly complicated if there are parallel units in the flow system, or time-dependent flows. Alternatively, Nauman and Buffham[1] suggest taking the Laplace transform of the RTD of each vessel. Using the properties of Laplace transforms, the RTD for two vessels in series is then the inverse Laplace transform of the product of the transforms for the individual vessels. Anderssen and White[2] have proposed solution of the RTD problem via Laguerre Functions.

The experimental determination of the RTD of an arbitrary flow is the subject of a great deal of study. Bischoff[3] has used pulse inputs, while Kramers and Alberda[4] apply frequency response techniques, and Woodburn[5] and Gibilaro, *et al.,*[6] propose the use of pseudorandom binary inputs. Nor is the concept of the moments of the RTD new. Nauman[7] gives the equation for the unsteady-state evolution of the first moment of the RTD. Nauman and Buffham[1] introduce the moments of the steady-state RTD and their relationship to the mean and variance of the distribution, and Nauman[8] gives the expression for the first three moments of the RTD of the steady-state CSTR. The use of the moments of an experimentally determined RTD to fit the parameters of a flow model is the subject of a great number of papers, the

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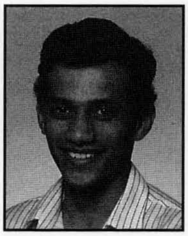

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more recent among them being Nauman and Buffham,[1] Simandi, *et al.*,[9] van Gelder and Westerterp,[10] and Thijert, *et al.*[11]

This paper extends the work of Nauman[7] by deriving the ordinary differential equations describing the evolution of the higher moments of the RTD of the unsteady-state CSTR. In addition, it derives the expressions for the moments of the RTD of a plug flow reactor. This allows the calculation of the overall RTD of a processing system using the leading moments of the RTD to characterize the RTD in much the same way as the leading moments of the molecular weight distribution of a polymer are used to characterize the molecular weight distribution of that polymer. Finally, the resulting equations are implemented in MATLAB®/SIMULINK® in a way that allows each process flow system to be defined graphically.

MOMENT DESCRIPTION OF THE RESIDENCE TIME DISTRIBUTION

We will define two basic types of vessels with which we will construct a flow system by connecting vessels of these two types in some combination of series and parallel. The first type will be a **stirred tank.** While we will assume that the contents of the tank are well mixed, we will not require that the tank contents have constant volume. That is, flow out does not have to equal flow in. Thus, the level in the tank can vary between zero and a maximum volume (V_{max}). Flow out may be constant, may vary with liquid level in an open-loop manner, or may be controlled by some type of level controller. The second type of vessel will be the **plug flow vessel.** This vessel will have perfect radial mixing, no axial mixing, and constant volume. Flow in will necessarily equal flow out. Figure 1 shows the two types of vessels. Any other vessel (such as a tank with dead volume) may be approximated as combinations of these two vessels, as in Levenspiel.[12]

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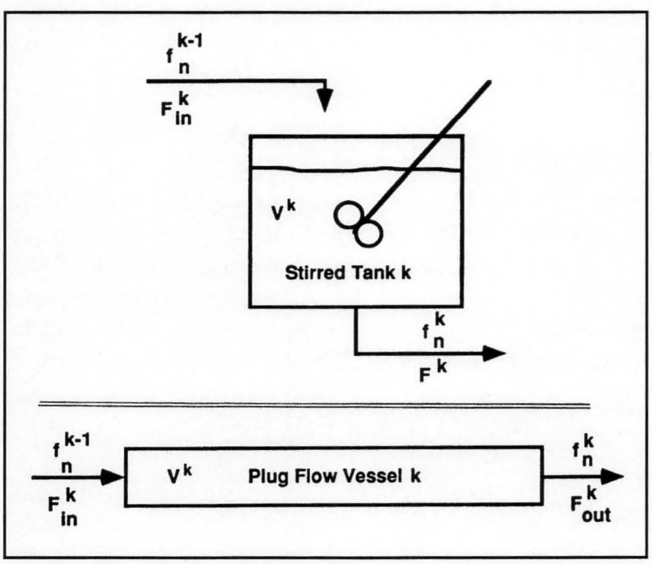

Figure 1. Stirred tank *(shown at the top) for the RTD development. Note that liquid volume need not be constant since inflow is not restricted to equal outflow. This allows the analysis of the RTD during tank filling or draining. In the* ***plug flow vessel*** *(shown at the bottom) volume is constant and inflow and outflow must be equal.*

Consider the k-th vessel in a flow system to be a stirred tank as shown in Figure 1. Define $f_n^k(t)$ to be the volume fraction at time t of the flow leaving vessel k which has a total age (since entering the flow system) of τ, where $n\Delta\tau \le \tau < (n+1)\Delta\tau$ and $\Delta\tau$ is a fixed increment of residence time. A balance over $f_n^k(t)$ for the stirred tank in Figure 1 yields

$$\frac{d\left[V^k f_n^k(t)\right]}{dt} = F_{in}^k f_n^{k-1}(t) - F_{out}^k f_n^k(t) - V\left[f_n^k(t) - f_{n-1}^k(t)\right] \tag{1}$$

The generation term (last term on the right-hand side) occurs because the fluid ages as it passes through the vessel. When Eq. (1) is multiplied by $(n)^i$ and summed over all values of n, the result is

$$\frac{d\left[V^k \sum_{n=0}^{\infty}(n)^i f_n^k(t)\right]}{dt} = F_{in}^k \sum_{n=0}^{\infty}(n)^i f_n^{k-1}(t) - F_{out}^k \sum_{n=0}^{\infty}(n)^i f_n^k(t) - V^k \sum_{n=0}^{\infty}(n)^i\left[f_n^k(t) - f_{n-1}^k(t)\right] \tag{2}$$

Defining $\lambda_i^k(t)$ as the i-th moment of the RTD, at time t, leaving the k-th vessel,

$$\lambda_i^k(t) = \sum_{n=0}^{\infty}(n)^i f_n^k(t) \tag{3}$$

Eq. (2) then becomes

$$\frac{d\left[V^k \lambda_i^k(t)\right]}{dt} = F_{in}^k \lambda_i^{k-1}(t) - F_{out}^k \lambda_i^k(t) - V\left[\lambda_i^k(t) - \sum_{n=2}^{\infty}(n)^i f_{n-1}^k(t)\right] \tag{4}$$

Algebraic manipulation will yield the following for the final term in Eq. (4):

$$\sum_{n=2}^{\infty}(n)^i f_{n-1}^k(t)=\begin{cases}\lambda_0^k(t)+\lambda_1^k(t) & \text{for } i=1\\ \lambda_0^k(t)+2\lambda_1^k(t)+\lambda_2^k(t) & \text{for } i=2\end{cases} \quad (5)$$

Thus, the RTD moment equations for the stirred tank become

$$\frac{d\left[V^k\lambda_1^k(t)\right]}{dt}=F_{in}^k\lambda_1^{k-1}(t)-F_{out}^k\lambda_1^k(t)+V^k\lambda_0^k(t)$$
$$\lambda_1^k(0)=0 \quad (6)$$

$$\frac{d\left[V^k\lambda_2^k(t)\right]}{dt}=F_{in}^k\lambda_2^{k-1}(t)-F_{out}^k\lambda_2^k(t)+V^k\left[\lambda_0^k(t)+2\lambda_1^k(t)\right]$$
$$\lambda_2^k(0)=0 \quad (7)$$

The initial conditions reflect the fact that at time equal to zero, all fluid in all vessels is assumed to have zero age. The values of the moments of the RTD of the inlet stream to vessel $k\left[\lambda_i^{k-1}(t)\right]$ are the moments of the outlet stream of vessel k-1. A stream entering the flow system will have the following moments:

$$\lambda_0^{k=0}(t)=1 \quad (8)$$

$$\lambda_i^{k=0}(t)=0 \qquad i>1 \quad (9)$$

his simply means that all material entering the system has an age of zero. In addition, since $f_n^k(t)$ is defined to be a volume fraction, it is normalized and the sum of all fractions must equal unity

$$\lambda_0^k(t)=\sum_{n=0}^{\infty}(n)^0 f_n^k(t)=1 \quad (10)$$

Thus, it is not necessary to track $\lambda_0^k(t)$. Equations (6)-(10) define the first three moments of the RTD at the outlet of the k-th vessel (if vessel k is a stirred tank).

Consider now the second type of vessel, the **plug flow vessel.** It corresponds to a simple pipe, or other processing unit in which there is no backmixing, as shown at the bottom of Figure 1. Proceeding as before, it is possible to write a balance on $f_n^k(t)$ over the plug flow vessel:

$$f_n^k(t)=f_{n-m}^{k-1}(t-\theta) \quad (11)$$

where θ is the residence time of the vessel, (V/F), and m is an integer representing the number of RTD increments, $\Delta\tau$, in θ. (If $\Delta\tau$ arbitrarily is set to one time unit, then $m=\theta$.) As before, Eq. (13) can be multiplied by $(n)^i$ and summed over all n

$$\sum_{n=0}^{\infty}(n)^i f_n^k(t)=\sum_{n=0}^{\infty}(n)^i f_{n-m}^{k-1}(t-\theta) \quad (12)$$

The left-hand side of Eq. (12) is $\lambda_i^k(t)$ (the i-th moment of the material leaving vessel k). Algebraic manipulation of the right-hand side results in

$$\sum_{n=0}^{\infty}(n)^i f_{n-m}^{k-1}(t-\theta)=\begin{cases}\lambda_1^{k-1}(t-\theta)+\theta\lambda_0^{k-1}(t-\theta) & \text{for } i=1\\ \lambda_2^{k-1}(t-\theta)+2\theta\lambda_1^{k-1}(t-\theta)+\theta^2\lambda_0^{k-1}(t-\theta) & \text{for } i=2\end{cases} \quad (13)$$

Thus, the moments of the RTD for the plug flow vessel become

$$\lambda_1^k(t)=\lambda_1^{k-1}(t-\theta)+\theta\lambda_0^{k-1}(t-\theta) \quad (14)$$

$$\lambda_2^k(t)=\lambda_2^{k-1}(t-\theta)+2\theta\lambda_1^{k-1}(t-\theta)+\theta^2\lambda_0^{k-1}(t-\theta) \quad (15)$$

Equations (14) and (15), along with Eqs. (8) through (10) where appropriate, make up the moment characterization of the RTD of the plug flow vessel. Notice once again that it is not necessary to track the zeroth moment since this is always unity for a normalized distribution.

Once a set of moments is available, the RTD of the material leaving the k-th vessel can be reconstructed in a number of ways.[13] The average residence time is the ratio of the first to zeroth moments

$$\theta^k(t)=\frac{\lambda_1^k(t)}{\lambda_0^k(t)} \quad (16)$$

The variance of the RTD can be written as

$$\sigma^k(t)^2=\frac{\lambda_2^k(t)}{\lambda_0^k(t)}-\left(\frac{\lambda_1^k(t)}{\lambda_0^k(t)}\right)^2 \quad (17)$$

In many cases, measures of the mean (θ) and variance (σ) may be enough to characterize the RTD. If desired, a log normal distribution may be presumed[14]

$$f(\tau,t)=\frac{a(t)^{0.5}}{\tau\pi^{0.5}}\exp\left[-a(t)\left(\ell n\,\tau-\ell n\,b(t)\right)^2\right]$$

$$a(t)=2\,\ell n\left(\frac{\lambda_0(t)\lambda_2(t)}{\lambda_1(t)^2}\right) \qquad b(t)=\frac{\lambda_1(t)}{\lambda_0(t)\exp\left(\frac{a(t)}{2}\right)} \quad (18)$$

IMPLEMENTATION IN MATLAB®

Equations (6), (7), (14), and (15) form a mathematical description of the RTD. As such, the set of equations, along with the proper initial conditions, could be solved with any numerical integrator in any standard language. We have chosen instead to exploit the graphical programming capabilities of MATLAB®, a mathematical analysis program marketed by The MathWorks of Natick, Massachusetts, which runs under DOS Windows or Macintosh System 7, as well as on various Unix workstations. A unique feature of MATLAB is the SIMULINK® subsystem. This is a block-diagram oriented environment in which various dynamic blocks are chosen from a library of dynamical elements and interconnected on the screen to form a dynamical system. The system can then be simulated directly from the diagram with no additional programming.

Most SIMULINK elements are transfer functions defined in the Laplace or Z-transform domains. Blocks are available to input systems of ordinary differential equations (ODEs) directly, but only if the ODEs are linear.

Equations (6), (7), (14), and (15) are nonlinear for varying liquid volumes or flowrates. To accommodate these nonlinear ODEs within the SIMULINK framework, we have developed SIMULINK modules for the stirred tank and plug flow vessel RTD. These are constructed of standard SIMULINK elements (integrators, etc.) and supplied to the user as dynamic blocks. The user then selects these elements from a menu much as he or she would select a transfer function block. The blocks are connected by arrows showing the information flow, and the entire block diagram is simulated by selecting the *Simulation* menu from SIMULINK. We have found that the nonlinear blocks are directly portable from the Macintosh to the Windows version of MATLAB. Plots of the time evolution of the moments can be created directly from SIMULINK or from MATLAB. The complete age distributions can be reconstructed from the moments by assuming a log normal distribution. This is easily done in MATLAB by means of a MATLAB function.

EXAMPLE

The use of the moment characterization and its implementation in MATLAB is best illustrated by an example. Consider the system shown in Figure 2 which is composed of two stirred tanks interconnected by a plug flow element. At time zero, the first stirred tank (CSTR 1) contains 500 L of material. The plug flow vessel (PFR) is initially empty, but has a volume of 100 L when full. The second stirred vessel (CSTR 2) has an initial liquid volume of 500 L.

At time zero, flow into CSTR 1 is begun at a constant 20 L/min. At the same time, flow out of CSTR 1 is begun, also at 20 L/min. PFR begins to fill at a rate of 20 L/min, and since its volume is 100 L, it is filled after five minutes, and flow into CSTR 2 begins (also at 20 L/min). When flow into CSTR 2 begins, flow out also begins at the same flowrate (20 L/min).

Figure 2 is not only the schematic for the RTD problem in question, it is also the SIMULINK "program" from which the simulation is to be carried out. Figure 2 was constructed by selecting the RTD stirred vessel and plug flow vessel modules from a menu and connecting them with arrows indicating information or material flows. The schematic also contains standard SIMULINK elements such as the clock and transport delay (used here to start outflow from PFR after it is filled).

The first three moments of the material entering CSTR 1 are set to zero, indicating zero age at the inlet. The initial volumes of CSTR 1 and 2 are set by double-clicking on the icon and entering a value when it is requested. The volume of the plug flow vessel (PFR) and its residence time are entered in a similar manner. Both the inflow and outflow of CSTR 1 are inputs. Their values are entered into the boxes at the left end of the respective arrows. The outflow of CSTR 1 becomes the inflow of PFR. Likewise, the outflow of PFR becomes the inflow of CSTR 2. This, however, is delayed (via the standard Transport Delay block) to account for the five minutes necessary to fill PFR. The RTD moments of the material leaving CSTR 1 become the moments of the inlet stream to PFR. Likewise, the moments of the outflow from PFR become the moments of the inflow to CSTR 2. Finally, the moments of the outflow from CSTR 2 characterize the RTD of the system under the conditions outlined above and can be sent to a SIMULINK workspace for plotting versus the time signal generated by the clock icon. The simulation is begun from the *Simulation* menu.

Figure 3 shows the time evolution of the average and variance of the RTD of the material leaving CSTR 2. The average residence time approaches a steady-state value of 55 min, which can be calculated as the sum of the nominal residence times of the two CSTRs (25 min each) and the residence time of the plug flow vessel (5 min). Figures 3, however, also shows the transient approach of the average residence time to its final value, as well as showing the variance of the RTD. The variance reaches a steady-state value of about 1,257, which gives a standard deviation (the square root of the variance) of approximately 35 min. Thus, although the average residence time at steady state is 55 min, the standard de-

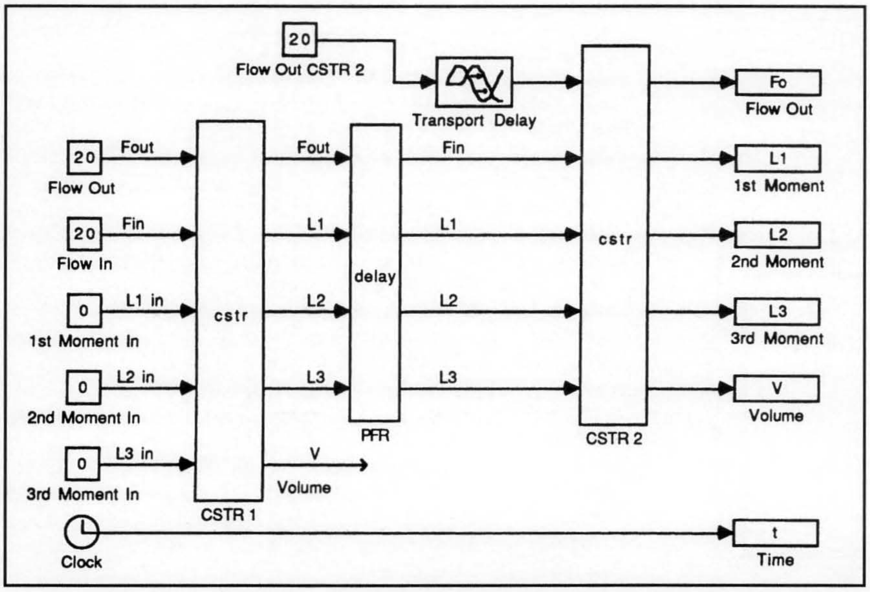

Figure 2. *SIMULINK diagram of two stirred tanks connected by a plug flow vessel. This is the diagram used to solve the Example. In it, Ln referes to the n-th moment of the RTD of the material leaving the vessel shown; Fin and Fout are the inlet and outlet flowrates, respectively, for a given vessel.*

viation of 35 min indicates quite a broad distribution. This is shown in Figure 4 where the steady-state moments from the example have been used to form a log-normal distribution. It may be seen that some elements of the fluid have residence times as short as 5 min, while others remain in the system for over 200 min.

The example was chosen for its simplicity to illustrate the use of the moment characterization and SIMULINK simulation. The level of complexity of problems which can be handled by this method is limited only by the patience of the user in developing the schematic and the computational speed of the computer.

CONCLUSIONS

The moment characterization of the RTD of a complex flow system allows the student to look at the effects of multiple vessels, nonsteady-state flow, and nonideal flow (where the nonideal vessel is approximated by a system of ideal vessels). When the moment description of the RTD is coupled with the graphical programming environment of the SIMULINK subsystem of the MATLAB mathematical programming package, the overall RTD for complex systems of vessels under the influence of transient flows can easily be analyzed. The simplicity of implementation allows the student to develop a conceptual understanding of RTD (in a visual, rather than equation, form), both for classroom demonstration and homework or project assignments.

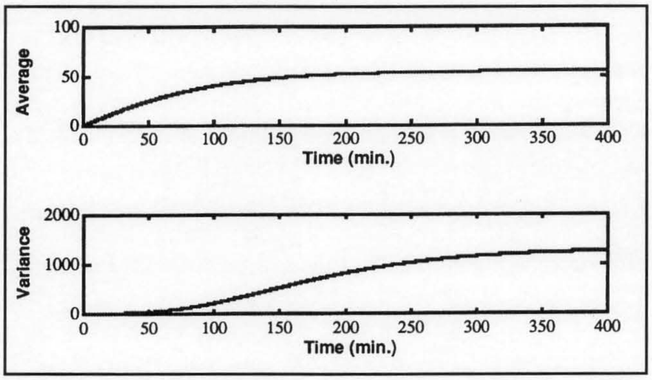

***Figure 3.** MATLAB plot of the evolution of the average residence time and the variance of the RTD as a function of time. The average and variance are derived from the moments of the RTD according to Eqs. (16) and (17), respectively. Note the approach to steady state as the system goes from startup to steady operation.*

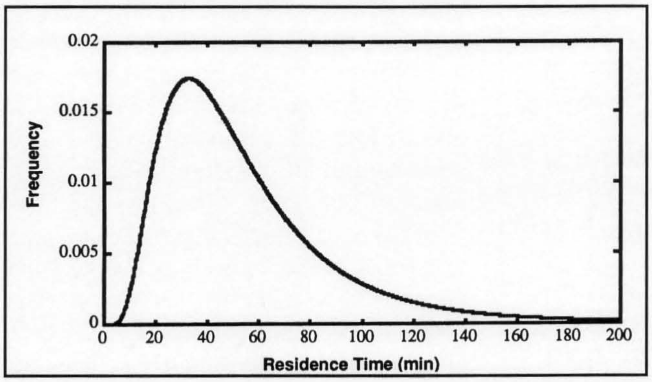

***Figure 4.** MATLAB plot of the steady-state RTD for the example. A log normal distribution has been used to recover the complete RTD from its first two moments.*

NOMENCLATURE

f_n^k fraction of material leaving vessel k which has a total residence time between $(n-1)\Delta\tau$ and $n\Delta\tau$

F_{in}^k flowrate into vessel k

F_{out}^k flowrate out of vessel k

t time

V^k volume of vessel k

Superscripts

k vessel number

Subscripts

i moment index

n residence time increment index

Greek Letters

$\Delta\tau$ increment in residence time

θ^k average residence time of the material leaving vessel k

λ_i^k i-th moment of the residence time of the material leaving vessel k

σ^k standard deviation of the RTD of the material leaving vessel k

τ residence time

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ChE book review

COULSON AND RICHARDSON'S CHEMICAL ENGINEERING

Volume 6 (Design), Second Edition

by R. K. Sinnott

Pergamon Press Ltd., Oxford, UK; 954 pages, $48.00 (1993)

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This book well serves the principal purpose of the author—to provide an undergraduate textbook for the chemical process design course. Expansions made to the first edition include an introductory presentation of process heat integration (pinch analysis), a discussion of safety and loss prevention, and a presentation of current (1992) costs related to process evaluation and step-counting techniques for fixed capital cost estimates.

The breadth of the treatment is impressive and includes mechanical design of process equipment. An extensive list of references, principally to sources in the UK and the USA, is available at the end of each chapter if additional detail is needed by the reader. The topics covered are discussed under the following chapter headings:

- Introduction to Design
- Fundamentals of Material Balances
- Fundamentals of Energy Balances
- Flow-Sheeting
- Piping and Instrumentation
- Costing and Project Evaluation
- Materials of Construction
- Design Information and Data
- Safety and Loss Prevention
- Equipment Selection, Specification, and Design
- Separation Columns (Distillation and Absorption)
- Heat-Transfer Equipment
- Mechanical Design of Process Equipment
- General Site Considerations

The book is of very high quality both in preparation and in presentation. A question that a chemical process design instructor might ask upon reading this text is, "Could this book be successfully used by students in my design course?" Since there are many ways to present a course in chemical process design, the answer to that question would not be an unambiguous yes or no. Some considerations include the following questions:

- *Is the volume self-sufficient?*
- *Are the topics covered current to the practice of process design?*
- *Would process costs that are presented in pounds sterling be accepted by the undergraduate audience in the USA?*
- *Does the library have resources to support access to the extensive list of references cited as publications of the IChemE (Institution of Chemical Engineers, London)?*
- *Is a sufficient supply of exercises provided for practice in the application of design techniques?*

The book is intended to be as self-sufficient as possible. The author often refers to the earlier books of the Coulson and Richardson series. For undergraduate programs that do not use that series in foundation courses, the instructor could prepare a list of equivalent references to alternate textbooks.

Current topics of design are covered in the revised edition. Topics mentioned, but not examined in detail, are batch processing and optimization. British standards and government management procedures for loss prevention are most frequently cited. Students in the USA will ask how those standards differ from USA standards and regulations, and the instructor should be prepared to answer.

The problem of converting costs from pounds sterling to American dollars is covered in detail, with examples that can be clearly understood by senior students.

A set of eight design projects is presented in Appendix G. A model answer is available in the literature for one project. There is no list of practice problems (exercises) at the end of each chapter. Fully-solved exercises are included, however, where appropriate to the presentation of topics in the book. An experienced design instructor should have no trouble in finding appropriate exercises and design projects from background. The lack of design exercises at the end of each chapter could be a more serious impediment to an instructor who wishes to rely entirely on the textbook as a source of design problems for the student.

The strengths of this book are the outstanding quality of writing, the consistent, successful effort to present technical material in the context of the process design requirement, and the breadth of coverage that results in a nearly self-sufficient textbook. It is recommended for serious consideration as a required textbook in an undergraduate process design course. ❐