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Characterization and Analysis of **Continuous Recycle Systems:**

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Part II. Cascade

A general analysis of a cascade of identical recycle units is presented by means of studying the fluid history inside the system. This is an extension of Part I (Mann et al., 1979) in which a single recycle unit was analyzed. The history of a fluid element is expressed in terms of the number of cycles it completes, the time it resides in the system and the total time it resides in a specific section of the system—all are random variables. Concepts from probability theory and stochastic processes are used to derive the number of cycles distribution (NCD), residence time distribution (RTD) and various total regional residence time distributions (TRRTDs) as well as their means and variances. Expressions for the joint distributions of pairs of these random variables are also derived as well as explicit expressions for their covariances and correlation coefficients. Two applications of the results are illustrated: one in analyzing a continuous spouted-bed coating unit and the second in using the cascade as a flexible, physically based multi-parameter flow model.

SCOPE

The history of a fluid element (or particle) in a continuous recycle system is characterized in terms of the number of cycles it completes, the time it resides in the system and the total time it resides in specific regions of the system. Detailed information on particle history can be quantitatively expressed by the joint distributions of pairs of these characteristics, by the number of cycle distribution (NCD), the residence time distribution (RTD), and the total regional residence time distribution (TRRTD). Joint distributions, their covariances and correlating coefficients are useful in analyzing processes in which particles undergo changes according to two different mechanisms (e.g., reaction and attrition of coal particles in a gasification

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The NCD is useful in analyzing processes in which the quality of the product is related on the number of cycles a particle completes (e.g., spouted bed coating). The RTD is useful when the quality of the product depends on the residence time in the system (e.g., chemical reaction) and in formulating flow models. The TRRTD is useful when particles undergo changes in certain sections of the system (e.g., reaction occurring in a high-temperature region).

In Part I (Mann et al., 1979) these concepts were introduced and a single recycle unit was studied. Explicit expressions for the joint distribution of the number of cycles and residence time, the NCD, RTD, TRRTD as well as the covariances and correlation coefficients of pairs of these characteristics were derived. In this article the analysis is expanded to a cascade of identical recycle units connected in series.

CONCLUSIONS AND SIGNIFICANCE

The history of a fluid element in a cascade of identical recycle units is described in terms of the number of cycles it completes, the time it resides in the system and the total time it resides in particular sections of the system—each of these is a random variable. Explicit expressions for the joint distribution of the number of cycles and the residence time, the number of cycle distribution (NCD), the residence time distribution (RTD), and total regional residence time distributions (TRRTDs) are derived.

The NCD is a negative binomial distribution which depends only on the recycle ratio, R, and the number of recycle units in the cascade, m. The mean of this distribution is m(R+1) and its variance is mR(1+R). It is shown that for a given value of the mean, the variance of the NCD decreases as m increases.

A general expression for the RTD is derived in terms of the recycle ratio, the number of units in the system and flow patterns, expressed by the one-pass RTDs in the two regions forming each unit. It is shown that for a given mean residence time, the density of the RTD converges to a dirac delta function as the number of units goes to infinity. In modeling, a flow model which is based on a cascade of recycle units provides two advantages over the continuous stirred tanks (CSTs) in-series model commonly used. First, each recycle unit may represent

more realistically actual sections of the system which are rarely well-mixed. Second, this model has three parameters (number of units, recycle ratio and unit configuration) which could be adjusted to fit a wide variety of measured RTD curves. The CSTs-in-series model has only one parameter (the number of tanks) and can be used to fit only RTD curves which belong to the class of gamma distribution.

Total regional residence time distributions are derived and it is shown that these distributions depend on m, R and the one-pass RTD through the respective region. The mean of the total regional residence time is equal to the product of the mean residence time in the system and volume fraction of that region.

The relationships among the number of cycles, residence time and total regional residence times are described by explicit expressions of the covariances and correlation coefficients of pairs of these variables. These could be used in estimating the value of one variable for a given value of the second. It is found that the correlation coefficients of the number of cycles and residence time, of the number of cycles and total regional residence time and of the total regional residence times in the two regions are independent of the number of units in the cascade.

BACKGROUND

In Part I (Mann et al., 1979), a general analysis of continuous recycle systems was given. The history of a fluid element inside the system was described in terms of several characteristics like the number of cycles a particle completes, the time it resides in the system and the total time it resides in a specified section of the system. Probabilistic methods were used to derive expressions for the number of cycles distribution (NCD), the residence time distribution (RTD) and the total regional residence time distribution (TRRTD) as well as their means and variances. The relationships among these characteristics were expressed in terms of joint distributions, covariances and correlation coefficients of pairs of these variables.

This paper expands the analysis of Part I to a cascade of m identical recycle units connected in series as shown schematically in Figure 1.

The interest in the cascade is two fold: first, numerous physical systems consist of identical recycle units. Two such systems are the continuous particle coating apparatus of Wurster and Lindlof (1966) and the granulation unit of Ormos et al. (1976). Second, many chemical engineering systems can be more realistically represented by a cascade of recycle units than by the commonly-used continuous stirred tanks (CSTs) in-series model (Levenspiel, 1972).

As in Part I, the present analysis considers the history of a single fluid element in the system in terms of the number of cycles it completes, the time it resides in the system, the time it

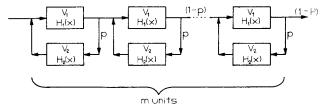


Figure 1. Schematic representation of the system.

resides in certain sections of the system and the relationships among these characteristics. In order to illustrate the use of some basic results from probability theory, the NCD and RTD are discussed first and then the joint distribution.

DISTRIBUTION OF NUMBER OF CYCLES

Consider a continuous system which consists of m identical recycle units shown schematically in Figure 1. Each unit consists of two flow regions: flow region "1" with volume V_1 and flow rate w_1 , located on the main flow line, and region "2" with volume V_2 and flow rate w_2 located on the recycle line. The net flow rate through each unit is $w_0 = w_1 - w_2$. In each unit, the one-path residence time distribution (RTD) in region "1" is $H_1(x)$ and the one-path RTD in region "2" is $H_2(y)$, as defined in Part I. It is assumed that the system is at steady state, and that the fluid consists of identical elements whose flow properties do not change within the system.

The number of cycles, N, is defined here as the number of times a fluid element passes through any one of flow regions "1" during its passage through the system. Thus, N is a discrete random variable which assumes the integral values m, m+1, Let

$$q_m(n) = P\{N = n\} \quad (n = m, m + 1...)$$

be the probability function of N. The corresponding distribution function

$$Q_m(n) = P\{N \le n\} = \sum_{i=m}^n q_m(i)$$

is the number of cycles distribution (NCD).

Let N_1 be the number of cycles which a fluid element completes in unit 1, N_2 the number of cycles it completes in unit 2, etc., then

$$N = N_1 + \ldots + N_m. \tag{1}$$

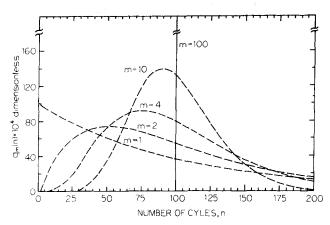


Figure 2. Number of cycles distribution.

Now, N_1, N_2, \ldots, N_m are independent random variables and from Part I we know that each follows a geometric distribution with parameter p, i.e.,

$$q(n) = P\{N_i = n\} = (1 - p)p^{n-1} \quad (n = 1, 2, ...)$$

where $p = w_2/w_1$ is the recycle fraction in each unit. In other words, p is the probability that a fluid element recycles in a unit after leaving its region "1."

It is well known that the sum of m independent random variables each with a geometric distribution follows a negative binomial distribution (Feller, 1968, p. 269), i.e.,

$$q_m(m+k) = {m+k-1 \choose k} p^k (1-p)^m$$
. $k=0, 1, 2...$ (2)

Here, k denotes the number of cycles above the minimum number, m, completed during a passage through the system. The mean and variance of this distribution are

$$\mu_N = E[N] = \frac{m}{1 - p} = (R + 1)m$$
 (3)

and

$$\sigma_N^2 = Var[N] = \frac{mp}{(1-p)^2} = R(R+1)m$$
 (4)

Note that, as expected, both the mean and the variance of the cascade are the sum of the mean and the variance, respectively, of the individual units.

The NCD depends on only the number of units and the recycle fraction (or recycle ratio) in each unit and does not depend on the internal configuration of the units, fluid properties or flow patterns in each unit. Figure 2 shows the probability function for systems with different numbers of units. For comparison purposes, we take the mean of the distribution as a constant, $\mu_N=100$. Then for each value of m, the recycle fraction, p, is determined by Eq. 3. Note that the probability function of N varies from a geometric distribution when m=1 to a dirac delta function when $m=\mu_N$. In the latter case p=0, and each fluid element passes only once through each unit and completes exactly m cycles. This can be seen by examining the ratio of the standard deviation and the mean of N for different number of units. From Eqs. 3 and 4:

$$\frac{\sigma_N}{\mu_N} = \sqrt{\frac{p}{m}} \,, \tag{5}$$

but for a given value of μ_N , $p = (\mu_N - m)/\mu_N$ and Eq. 5 becomes

$$\frac{\sigma_N}{\mu_N} = \sqrt{\frac{\mu_N - m}{\mu_N m}} \quad . \tag{6}$$

Since $\mu_N \ge m$, the ratio becomes smaller as m increases and is

zero when $m = \mu_N$ in which case p = 0 by Eq. 3. Note that when $\mu_N >> m$, Eq. 6 is reduced to $\sigma_N/\mu_N \cong \sqrt{1/m}$, thus the relative standard deviation is inversely proportional to \sqrt{m} .

RESIDENCE TIME DISTRIBUTION

The residence time in the system is defined as the time a fluid element spends in the system. Let T denote this random variable and let

$$F_m(t) = P\{T \le t\} \quad (t > 0)$$

stand for its distribution function. $F_m(t)$ is by definition the RTD of the system.

The residence time in the system is clearly the sum of the residence time in each of its units. Thus, if T_1 denotes the time a fluid element spends in unit "1," T_2 the time it spends in unit "2," etc., then

$$T = T_1 + T_2 + \ldots + T_m . (7)$$

The random variables T_1, T_2, \ldots, T_m are all independent and all follow the same distribution, F(t), which is the RTD of a single unit derived in Part I. Hence, the RTD of the cascade is the m-fold convolution of F(t), i.e.,

$$F_m(t) = [F(t)]^{*m}. \tag{8}$$

It follows immediately (see Appendix B of Part I) that the Laplace transform of $F_m(t)$ is:

$$\hat{F}_m(s) = \int_a^\infty e^{-st} dF_m(t) = [\hat{F}(s)]^m,$$

which upon substitution according to Eq. 20 of Part I, becomes

$$\hat{F}_{m}(s) = \left[\frac{(1-p) \, \hat{H}_{1}(s)}{1-p \, \hat{H}_{1}(s) \, \hat{H}_{2}(s)} \right]^{m}. \tag{9}$$

This is the transfer function of the system.

The mean and variance of the RTD can be derived from Eq. 9 using the methods described in Appendix B of Part I, or alternatively, directly from Eq. 7 noting that the expected value of a sum of random variables is the sum of the expected values of the individual random variables. Thus,

$$\mu_T = E[T] = E[T_1 + \dots + T_m]$$

$$= E[T_1] + \dots + E[T_m] = m E[T_1].$$

But, $E[T_1]$ is the mean residence time in a single unit which is given in Eq. 21 of Part I. Hence,

$$\mu_T = \frac{m}{1 - p} (\mu_1 + p\mu_2) . \tag{10}$$

Similarly, using the fact that the variance of the sum of independent random variables is the sum of the variances of the individual random variables we have

$$\sigma_T^2 = Var[T] = Var[T_1 + \dots + T_m]$$

$$= Var[T_1] + \dots + Var[T_m] = m \ Var[T_1].$$

Substituting the expression for the variance of the RTD in a single unit given in Eq. 22 of Part I we obtain,

$$\sigma_T^2 = \frac{mp}{(1-p)^2} (\mu_1 + \mu_2)^2 + \frac{m}{1-p} (\sigma_1^2 + p\sigma_2^2).$$
 (11)

Here, μ_1 , μ_2 , σ_1^2 , σ_2^2 are, respectively, the means and variances of $H_1(x)$ and $H_2(y)$. Since $\mu_1 = V_1/w_1$, $\mu_2 = V_2/w_2$, $V_n = V_1 + V_2$, $w_2 = pw_1$ and $w_2 = w_1 - w_n$, it follows from Eq. 10, that the mean residence time in the system is mV_n/w_n , which, as expected, is equal to the total volume of the system divided by the volumetric flow rate.

A more explicit expression for the RTD of the system than Eq.

8, may be obtained either by performing the formal convolution operation of Eq. 8, or alternatively, as follows. First write

$$F_m(t) = P\{T \le t\} =$$

$$\sum_{k=0}^{\infty} P\{T \le t \mid N = m + k\} P\{N = m + k\}. \quad (12)$$

This is the so-called formula of total probability where the first term under the sum is the conditional probability that $T \le t$ given that N = m + k. To evaluate this term we note that once it is known that a fluid element completed exactly m + k cycles in the system it then immediately follows that it visited m + k times in regions "1", and k times in regions "2". Thus,

$$P\{T \le t \mid N = m + k\} = [H_1^{*(m+k)} * H_2^{*k}](t) \cdot$$

Substituting this in Eq. 12 together with the expression for $P\{N = m + k\}$ according to Eq. 2 we obtain

$$F_m(t) = (1-p)^m \sum_{k=0}^{\infty} {m+k-1 \choose k} \cdot p^k [H_1^{*(m+k)} H_2^{*k}](t) . \quad (13)$$

When the densities of $H_1(x)$ and $H_2(y)$ exist, one can obtain an explicit expression for the density of the RTD, $f_m(t)$, by rewriting Eq. 13 as:

$$f_m(t) = (1 - p)^m \sum_{k=0}^{\infty} {m + k - 1 \choose k}$$
$$p^k \int_0^t h_1^{*(m+k)}(x) h_2^{*k}(t - x) dx. \quad (14)$$

In principle, whenever m, p, $h_1(x)$ and $h_2(y)$ are known, $f_m(t)$ can be calculated directly from Eq. 14. However, the computations may be quite difficult since they involve evaluations of n-th order convolutions and an infinite series of integrals. Fortunately, for many commonly-used system configurations useful RTD expressions can be derived. Using the same technique described in Part I, we can write explicit RTD expressions when the two flow regions are represented by either a plug-flow zone, a series of equal-size stirred tanks, or the combination of the two. For convenience, we divide these into the four cases shown schematically in Figure 2 of Part I.

Case A: In each unit, both flow regions are represented by a plug flow zone, i.e., $h_1(x) = \delta(x - \tau_1)$, $h_2(y) = \delta(y - \tau_2)$, where $\tau_1 = V_1/w_1$ and $\tau_2 = V_2/w_2$. For this case Eq. 14 reduces to

$$f_m(t) = (1 - p)^m \sum_{k=0}^{\infty} {m + k - 1 \choose k} \cdot v^k \delta(t - (m + k)\tau_1 - k\tau_2).$$
 (15)

Case B: In each unit, region "1" is represented by α_1 equalsize stirred tanks connected in series to a plug flow zone and region "2" is represented by a plug flow zone, i.e.,

$$h_{1}(x) = \frac{1}{\Gamma(\alpha_{1})} \frac{1}{\beta_{1}} \left(\frac{x - \tau_{1}}{\beta_{1}} \right)^{\alpha_{1} - 1} e^{-(x - \tau_{1})/\beta_{1}} \quad (x \geq \tau_{1})$$

$$h_{2}(y) = \delta(y - \tau_{2}),$$

where $\tau_1=V_{1p}/w_1$, $\beta_1=V_{1m}/\alpha_1w_1$ and $\tau_2=V_2/w_2$. For this case Eq. 14 reduces to

$$f_{m}(t) = (1 - p)^{m} \sum_{k=0}^{\infty} {m + k - 1 \choose k}.$$

$$\frac{p^{k}}{\Gamma((m+k)\alpha_{1})} \frac{1}{\beta_{1}} \left(\frac{t - (m+k)\tau_{1} - k\tau_{2}}{\beta_{1}} \right)^{(m+k)\alpha_{1}-1} \cdot e^{-(t-(m+k)\tau_{1}-k\tau_{2})/\beta_{1}} \quad (t \geq (m+k)\tau_{1} + k\tau_{2}) \quad (16)$$

Case C: In each unit, region "1" is represented by a plug-flow

zone and region "2" by a plug flow zone connected to a series of α_2 equal-size stirred tanks, i.e.,

$$h_1(x) = \delta(x - \tau_1)$$

$$h_2(y) = \frac{1}{\Gamma(\alpha_2)} \; \frac{1}{\beta_2} \; \left(\frac{y - \tau_2}{\beta_2} \right)^{\alpha_2 - 1} e^{-(y - \tau_2)/\beta_2} \; , \quad (y \geq \tau_2)$$

where $\tau_1=V_1/w_1$, $\tau_2=V_{2p}/w_2$ and $\beta_2=V_{2m}/\alpha_2w_2$. For this case Eq. 14 reduces to

$$f_m(t) = (1 - p)^m \delta(t - m\tau_1)$$

$$+ (1 - p)^{m} \sum_{k=1}^{\infty} {m+k-1 \choose k}.$$

$$\frac{p^{k}}{\Gamma(k\alpha_{2})} \frac{1}{\beta_{2}} \left(\frac{t - (m+k)\tau_{1} - k\tau_{2}}{\beta_{2}} \right)^{k\alpha_{2}-1}$$

$$\cdot e^{-(t-(m+k)\tau_{1}-k\tau_{2})/\beta_{2}} \qquad (t > (m+k)\tau_{1} + k\tau_{2}) \quad (17)$$

Case D: In each unit, both region "1" and region "2" are represented by a plug flow zone connected to a series of equal-size stirred tanks (α_1 in region "1" and α_2 in region "2"). Here

$$h_1(x) = \frac{1}{\Gamma(\alpha_1)} \frac{1}{\beta_1} \left(\frac{x - \tau_1}{\beta_1} \right)^{\alpha_1 - 1} e^{-(x - \tau_1)/\beta_1} \quad (x \ge \tau_1)$$

$$h_2(y) = \frac{1}{\Gamma(\alpha_2)} \frac{1}{\beta_2} \left(\frac{y - \tau_2}{\beta_2} \right)^{\alpha_2 - 1} e^{-(y - \tau_2)/\beta_2}, \quad (y \ge \tau_2)$$

where $\tau_1 = V_{1p}/w_1$, $\beta_1 = V_{1m}/\alpha_2w_1$, $\tau_2 = V_{2p}/w_2$ and $\beta_2 = V_{2m}/\alpha_2w_2$. In this case, Eq. 14 reduces to

$$f_{m}(t) = (1 - p)^{m} \sum_{k=0}^{\infty} {m + k - 1 \choose k} \cdot p^{k} \frac{1}{\Gamma((m+k)\alpha_{1})} \frac{1}{\Gamma(k\alpha_{2})} \frac{1}{\beta_{1}} \frac{1}{\beta_{2}} \cdot \int_{(m+k)\tau_{1}}^{t-k\tau_{2}} \frac{\left(x - (m+k)\tau_{1}}{\beta_{1}}\right)^{(m+k)\alpha_{1}-1} e^{-\frac{x-(m+k)\tau_{1}}{\beta_{1}}} \cdot \left(\frac{t - x - k\tau_{2}}{\beta_{2}}\right)^{k\alpha_{2}-1} e^{-\frac{t - x - k\tau_{2}}{\beta_{2}}} dx \quad (18)$$

for $t \ge (m + k)\tau_1 + k\tau_2$ and zero otherwise.

The foregoing RTD expressions can be used to formulate and verify various flow models. Figures 3 and 4 show the $f_m(t)$ curves for several illustrative systems at various values of m. The versatility of a flow model based on a cascade of recycle units to represent systems whose RTD curves have wide range of shapes is evident.

JOINT DISTRIBUTION OF THE NUMBER OF CYCLES AND RESIDENCE TIME

The joint distribution of N and T is defined as the probability that a fluid element completes exactly n cycles while residing in the system no longer than time t, viz.,

$$G_m(n, t) = P\{N = n, T \le t\}, (n = m, m + 1, ...; t > 0)$$

Both the RTD and NCD are special cases of the joint distribution and are related to $G_m(n, t)$ by:

$$F_m(t) = P\{T \le t\} = P\{N < \infty, \ T \le t\} = \sum_{n=m}^{\infty} G_m(n, \ t), \quad (19)$$

and

$$q_m(n) = P\{N = n\} = P\{N = n, T < \infty\} = \lim_{n \to \infty} G_m(n, t)$$
 (20)

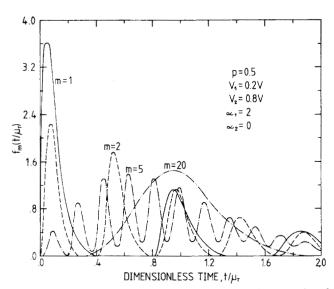


Figure 3. Illustration of residence time distribution for different number of units ($\rho = 0.5$; $V_1 = 0.2V_0$; $V_2 = 0.8V_0$; $\alpha_1 = 2$; $\alpha_2 = 0$).

This joint distribution can be evaluated using, once again, conditional probabilities,

$$G_m(m + k, t) = P\{N = m + k, T \le t\}$$

= $P\{T \le t \mid N = m + k\}P\{N = m + k\}.$ (21)

In the last expression of Eq. 21, the first term is given by Eq. 12 and the second term by Eq. 2. Hence,

$$G_m(m+k,t) = {m+k-1 \choose k} p^k (1-p)^m [H_1^{*(m+k)*} H_2^{*k}](t).$$
(22)

In principle, whenever m, p, $H_1(x)$, and $H_2(y)$ are known, $G_m(n,t)$ can be evaluated for any value of n and t. Note that for m=1, i.e., when the system consists of a single recycle unit, Eq. 22 is reduced to the joint distribution of a single unit as given in Part I. When p=0, i.e., when each unit has no recycle, $G_m(n,t)=H_1^{*m}(t)$ which is then the RTD of the system.

In many cases, it is technically difficult to evaluate $G_m(n, t)$ explicitly. It is then useful to consider the joint transform, defined in Part I, from which moments can be easily calculated. Applying the technique described in Part I, and using the properties of Laplace transform, the joint transform of a cascade of m recycle units is given by:

$$\hat{G}_{m}(z, s) = \left[\frac{(1 - p)z\hat{H}_{1}(s)}{1 - pz\hat{H}_{1}(s)\hat{H}_{2}(s)}\right]^{m}, \tag{23}$$

where |z| < 1 and s > 0 are the transform variables and $\hat{H}_1(s)$ and $\hat{H}_2(s)$ are the Laplace transforms of $H_1(x)$ and $H_2(y)$, respectively.

The expressions of Eqs. 22 and 23 are two key relations which characterize the behavior of the system. Both the NCD and the RTD can be derived from Eq. 22 using Eqs. 19 and 20 and their moments can be derived from Eq. 23. However, since these have already been obtained via different methods, we shall not pursue this here. Instead, we shall later use them to derive product moments and correlation coefficients of these variables. Before we do this, let us look at regional residence time distributions.

TOTAL REGIONAL RESIDENCE TIME DISTRIBUTIONS

Certain processes depend on the total time a fluid element stays in a specific section of the system. Such processes are conveniently characterized by the total regional residence time distributions (TRRTDs). In the cascade, several regions may be

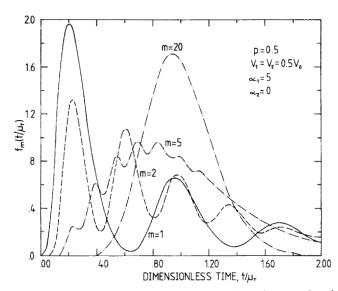


Figure 4. Illustration of residence time distribution for different number of units ($\rho=0.5$; $V_1=V_2=0.5V_0$; $\alpha_1=5$; $\alpha_2=0$).

of interest. When the residence time in a specific unit is important, the TRRTD is given by the RTD of a single recycle unit. When the total residence time in a flow region of a specific unit is important, the TRRTD is given by the TRRTD of either region "1" or region "2" derived in Part I. If the total residence time in all regions "1" or regions "2" is important, the TRRTD is obtained as follows.

Let W_1 be the total time a fluid element spends in region "1" of unit 1, W_2 be the time spent in region "1" of unit 2, etc. Then the total regional residence time in regions "1" of the cascade, $T_{m}^{(1)}$, is given by

$$T_m^{(1)} = W_1 + \ldots + W_m$$

The distribution of $T_m^{(1)}$ is

$$F_m^{(1)}(t) = P\{T_m^{(1)} \le t\}$$
,

which is by definition the TRRTD for all regions "1." An expression for $F_m^{(1)}(t)$ can be easily obtained directly from Eq. 13 by noting that the total residence time in regions "1" is equal to the residence time in the system if the residence time in all regions "2" were zero. Thus, by substituting a unit step function at the origin for $H_2(y)$ into Eq. 13 one obtains

$$F_m^{(1)}(t) = (1-p)^m \sum_{k=0}^{\infty} {m+k-1 \choose k} p^k [H_1^{*(m+k)}](t).$$
(24)

When the density of $H_1(x)$ exists the density of the TRRTD of regions "1" can be written as:

$$f_m^{(1)}(t) = (1-p)^m \sum_{k=0}^{\infty} {m+k-1 \choose k} p^k h_1^{*(m+k)}(t)$$
 (25)

For the special case when region "1" is represented by a cascade of α_1 equal-size stirred tanks and a plug flow zone Eq. 25 reduces to

$$\begin{split} f_m^{(1)}(t) &= (1-p)^m \sum_{k=0}^{\infty} \binom{m+k-1}{k}, \\ p^k \frac{1}{\Gamma((m+k)\alpha_1)} \frac{1}{\beta_1} \left(\frac{t-(m+k)\tau_1}{\beta_1} \right)^{(m+k)\alpha_1-1} \\ &+ e^{-(t-(m+k)\tau_1)/\beta_1}. \end{split}$$

The Laplace transform of $F_m^{(1)}(t)$ is obtained by substituting $\hat{H}_2(s) = 1$ into Eq. 9, viz.,

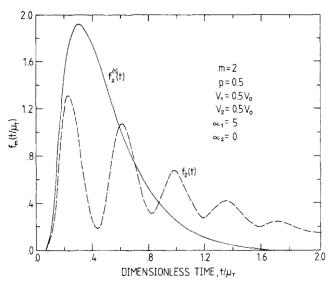


Figure 5. Illustration of residence time distribution and total regional residence time distribution (m=2; p=0.5; $V_1=V_2=0.5V_0$; $\alpha_1=5$; $\alpha_2=0$).

$$\hat{F}_{m}^{(1)}(s) = \left[\frac{(1-p)\hat{H}_{1}(s)}{1-p\hat{H}_{1}(s)} \right]^{m}. \tag{26}$$

The mean and the variance of $F_m^{(1)}(t)$ can be obtained directly from Eq. 26 using the relations given in Appendix B of Part I. The results are

$$E[T_m^{(1)}] = \frac{m}{1-p} \mu_1 \tag{27}$$

and

$$Var[T_m^{(1)}] = m \left[\frac{p}{(1-p)^2} \mu_1^2 + \frac{1}{1-p} \sigma_1^2 \right]$$
 (28)

Note that both $F_m^{(1)}(t)$ and its moments depend only on m, p, and $H_1(x)$, and not $H_2(y)$. Also note that the mean and the variance of the TRRTD for all regions "1" are, as expected, m times the mean and variance of the TRRTD in region "1" of a single unit. Also, when m=1, Eq. 25 is reduced to the single unit expression derived in Part I. Since, $\mu_1=V_1/w_1=(1-p)V_1/w_0$ and $\mu_T=m(V_1+V_2)/w_0$, Eq. 27 becomes

$$E[T_m^{(1)}] = \frac{V_1}{V_1 + V_2} \mu_T.$$
 (29)

Thus, the mean of the total regional residence time in regions "1" is proportional to the volume fraction of these regions and the mean residence time in the system. Also, upon substitution $\mu_T = (V_1 + V_2)m/w_n$ Eq. 29 becomes

$$E[T_m^{(1)}] = \frac{mV_1}{w_n},$$

which shows that the mean regional time in regions "1" is, as expected, the volume of regions "1" divided by the net flow rate through the system.

The TRRTD in regions "2" and its moments may be obtained in a similar way. The results are:

$$\begin{split} F_m^{(2)}(t) &= (1-p)^m \sum_{k=1}^\infty \binom{m+k-1}{k} p^k [H_2^{*k}](t), \\ \hat{F}_m^{(2)}(s) &= \left[\frac{(1-p)}{1-p \hat{H}_2(s)} \right]^m, \\ E[T_m^{(2)}] &= \frac{mp}{1-p} \mu_2 \ , \end{split}$$

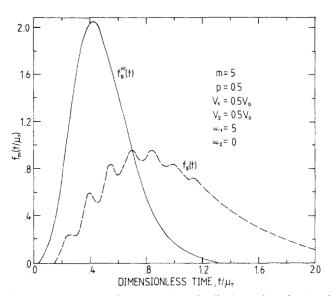


Figure 6. Illustration of residence time distribution and total regional residence time distribution (m = 5; p = 0.5; $V_1 = V_2 = 0.5V_0$; $\alpha_1 = 5$; $\alpha_2 = 0$).

$$Var[T_m^{(2)}] = m \left[\frac{p}{(1-p)^2} \mu_2^2 + \frac{p}{1-p} \sigma_2^2 \right].$$
 (30)

When region "2" is represented by a series of α_2 equal-size stirred tanks connected to a plug flow zone, the density of $F_m^{(2)}(t)$

$$f_m^{(2)}(t) = (1 - p)^m \sum_{k=1}^{\infty} {m + k - 1 \choose k} \cdot p^k \frac{1}{\Gamma(k\alpha_2)} \frac{1}{\beta_2} \left(\frac{t - k\tau_2}{\beta_2} \right)^{k\alpha_2 - 1} e^{-(t - k\tau_2)\beta_2}$$

Figures 5 and 6 show the densities of $f_m^{(1)}(t)$, and $f_m(t)$ for two illustrative recycle systems.

JOINT PARAMETERS

As indicated in Part I, useful concise information on fluid history in the system may be gathered from the joint parameters (covariances and correlation coefficients) of N and T, N and T_m⁽¹⁾, N and T_m⁽²⁾, T_m⁽¹⁾ and T_m⁽²⁾, etc. We start with the covariance of N and T. By definition,

$$COV(N, T) = E[(N - E[N])(T - E[T])].$$

From the definitions of $N_1, \ldots, N_m, T_1, \ldots, T_m$, and Eqs. 1 and 7 we obtain

$$COV(N, T) = E\left[\left(\sum_{i=1}^{m} N_{i} - E\left[\sum_{i=1}^{m} N_{i}\right]\right)\right] \cdot \left(\sum_{j=1}^{m} T_{j} - E\left[\sum_{j=1}^{m} T_{j}\right]\right) = E\left[\left(\sum_{i=1}^{m} (N_{i} - E[N_{i}])\right)\right] \cdot \left(\sum_{j=1}^{m} (T_{j} - E[T_{j}])\right) = E\left[\sum_{i=1}^{m} (N_{i} - E[N_{i}])(T_{i} - E[T_{i}])\right] + 2\sum_{i < j} (N_{i} - E[N_{i}])(T_{j} - E[T_{j}])\right] - \sum_{i < j} E\left[\left(N_{i} - E[N_{i} - E[N_{i}])(T_{j} - E[T_{j}])\right]\right] + 2\sum_{i < j} E\left[\left(N_{i} - E[N_{i} - E[N_{i}])(T_{j} - E[T_{j}])\right]\right].$$
(31)

For the *i*-th unit, $E[(N_i - E[N_i])(T_i - E[T_i])]$ is the covariance of N and T for a recycle unit and is given by Eq. 63 of Part I. Moreover, all the terms under the second sum vanish since each is $COV(N_i, T_j)$ and for i < j, N_i and T_j are independent. Thus Eq. 31 reduces to

$$COV(N, T) \approx \frac{mp}{(1-p)^2} (\mu_1 + \mu_2)$$
 (32)

The correlation coefficient of N and T

$$\rho(N, T) = \frac{COV(N, T)}{(Var[N])^{1/2}(Var[T])^{1/2}} ,$$

is obtained by substitution from Eqs. 4 and 11. It is:

$$\rho(N, T) = \left[1 + \frac{1 - p}{p} \frac{(\sigma_1^2 + p\sigma_2^2)}{(\mu_1 + \mu_2)^2}\right]^{-1/2}$$
(33)

Note that the correlation coefficient of N, and T in a cascade of m identical recycle units is independent of m and is the same as that of a single unit.

The covariance of N and $T_m^{(1)}$ can be obtained, like in Part I, by substituting $\mu_2 = 0$ and $\sigma_2 = 0$ in Eqs. 32 and 33. This gives:

$$COV(N, T_m^{(1)}) = \frac{mp}{(1-p)^2} \mu_1$$

and hence

$$\rho(N, T_m^{(1)}) = \frac{\mu_1}{\left(\mu_1^2 + \frac{1-p}{p} \sigma_1^2\right)^{1/2}}$$

Similarly the covariance and correlation coefficient of N and $T_m^{(2)}$ are:

$$COV(N, T_m^{(2)}) = \frac{mp}{(1-p)^2} \mu_2$$

and

$$\rho(N, T_m^{(2)}) = \frac{\mu_2}{\left[\mu_2^2 + (1-p)\sigma_2^2\right]^{1/2}}$$

Once again these are the same as for a single unit and are independent of m.

To obtain an expression for the covariance of $T_m^{(1)}$ and $T_m^{(2)}$ we note that $T = T_m^{(1)} + T_m^{(2)}$, use the relation

$$COV(T_m^{(1)}, T_m^{(2)}) = (Var[T] - Var[T_m^{(1)}] - Var[T_m^{(2)}])/2,$$

(see Eq. 70 of Part I) and substitute Eqs. 11, 28, and 30. This leads to:

$$COV(T_m^{(1)}, T_m^{(2)}) = \frac{mp\mu_1\mu_2}{(1-p)^2},$$

and hence

$$\rho(T_m^{(1)}, T_m^{(2)}) = \frac{p\mu_1\mu_2}{[p\mu_1^2 + (1-p)\sigma_1^2]^{1/2}[p\mu_2^2 + (1-p)p\sigma_2^2]^{1/2}}$$

Here again, the correlation coefficient is independent of m and is the same as that of a single unit.

APPLICATIONS

The foregoing analysis provides general expressions which can characterize almost any operation carried out in a process which can be represented by a cascade of *m* identical recycle units. To give an idea on the applicability of the results we shall discuss here two situations where they can be used. The first is an example of an operation in which the number of passages through regions "1" is the important system parameter. The second is a short discussion on the versatility of the cascade of recycle units in modeling.

Consider first the particle coating apparatus of Wurster and Lindorf (1966) which can be schematically described by Figure 1. Every time a particle passes through a region "1" it is being sprayed with a coating solution and the solvent evaporates when particles pass through regions "2." Thus, the total amount of coating on a particle depends on the number of cycles it completes and the amount of coating in each passage, which is also a random variable. A more complete analysis of the coating unit will be given elsewhere. Here, for simplicity, we assume that the amount of coating added on in each passage is constant. The problem we address is how many units should be used in order to satisfy given requirements on coating uniformity.

Since the amount of coating added on in each passage is fixed, the coating requirement immediately determines the mean number of cycles. Let us say that the mean is E[N] = M, where M is known. The requirement on coating uniformity among particles can be stated in several ways. In most cases the uniformity specification is that the fraction of particles carrying less than a specified amount of coating C_1 , or more than C_2 is less than γ . Since the amount of coating added in each cycle is constant, these limits can be expressed in terms of two integers L_1 and L_2 which denote the lowest and highest number of cycles acceptable. Then from Eq. 2 the uniformity specification can be written as:

$$(1-p)^m \sum_{k=L_1-m}^{L_2-m} {m+k-1 \choose k} p^k \ge 1-\gamma$$

From Eq. 3, p = (M - m)/M and the uniformity specification can be expressed as:

$$\left(\frac{m}{M}\right)^{m} \sum_{k=1,\dots,m}^{L_2-m} {m+k-1 \choose k} \left(\frac{M-m}{m}\right)^k \ge 1-\gamma$$

This relation can be solved numerically to determine the value of m. Once m is selected the recycle fraction (operating conditions) in each unit is determined by Eq. 3.

The second application concerns the use of a cascade of recycle units as a flow model. This model provides two advantages over the stirred tanks-in-series model, commonly used in chemical engineering. First, each recycle unit represents more realistically local deviations from ideal mixing. Second, this model consists of three parameters (m, p, and unit configuration) which could be conveniently adjusted to many practical situations. The configuration of each recycle unit is first hypothesized on the basis of available knowledge regarding the system structure and behavior. Then it is adjusted by trial and error until the shape of the RTD curve of the model corresponds to the measured RTD curve of the actual system. The diversity of RTD curves which can be represented by the cascade of recycle units model is illustrated in Figures 3 and 4.

Finally, we note that a well known result on the stirred tanks-in-series model also holds true for any cascade of recycle units irrespective of the configuration of each unit. Specifically, we wish to show that a cascade system with fixed mean residence time behaves, at the limit, when m goes to infinity like a plug flow region. To show this we assume that $\mu_T = mV_o/w_o$ is constant, that system configuration does not change, i.e., V_1/V_2 is constant, and that mixing patterns do not change, i.e., σ_1/μ_1 and σ_2/μ_2 are both constants. Then a straight forward argument using Eq. 10 will show that σ_T^2 goes to zero as m goes to infinity and the result follows.

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 $F_m(t)$, $F_m^{(1)}(t)$, $F_m^{(2)}(t)$ = total residence time distribution for the system, regions "1" and regions "2", respectively (dimensionless)

 $\boldsymbol{\hat{F}}_m(s),\,\boldsymbol{\hat{F}}_m^{\,(1)}\!(s),\,\boldsymbol{\hat{F}}_m^{\,(2)}\!(s)=$ Laplace transform of $\boldsymbol{F}_m(t),\,\boldsymbol{F}_m^{\,(1)}\!(t)$ and

 $F_m^{(2)}(t)$ (dimensionless) $f_m(t), f_m^{(1)}(t)f_m^{(2)}(t) = \text{densities of } F_m(t), F_m^{(1)}(t) \text{ and } F_m^{(2)}(t)$ $(time^{-1})$

 $G_m(n, t)$ = joint distribution of number of cycles and residence time (dimensionless).

 $H_1(x)$, $H_2(y)$ = one-pass residence time distribution in region "1" and region "2," respectively (dimensionless)

 $\hat{H}_1(s)$, $\hat{H}_2(s)$ = Laplace transform of $H_1(x)$ and $H_2(y)$, respectively (dimensionless)

 $h_1(x)$, $h_2(y) = \text{densities of } H_1(x) \text{ and } H_2(y)$, respectively (time⁻¹) = number of cycles in the system, a random variable

 N_i = number of cycles in the *i*-th unit, a random variable

= number of units in the cascade m

recycle fraction w_2/w_1 (dimensionless)

 $Q_m(n)$ = distribution function of N, NCD (dimensionless)

 $q_m(n)$ = probability function of N (dimensionless)

= recycle ratio w_2/w_0 (dimensionless)

= variable of Laplace transform

 T_i = residence time in the system and in the *i*-th unit, respectively, random variables

 $T_m^{(1)}$, $T_m^{(2)}$ = total residence time in regions "1" and "2," respectively, random variables

= time

 V_0 , V_1 , V_2 =volume of a unit, region "1" and region "2" of a unit, respectively

 V_{1m} , V_{2m} = volume of mixing zone in region "1" and region "2," respectively

 V_{1p} , V_{2p} = volume of plug flow zone in region "1" and region "2," respectively

 $w_0, w_1, w_2 = \text{net flow rate through each unit, through region "1"}$ and region "2," respectively (volume/time)

= total regional residence time in region "1" of the i-th W_{i} unit, a random variable

= time x, y

transform variable (dimensionless)

Greek Letters

 α_1, α_2 = number of equal-size stirred tanks in region "1" and region "2," respectively (dimensionless)

mean residence time of one passage through a stirred $\beta_1, \beta_2 =$ tank in region "1" and region "2," respectively (time)

 $\Gamma()$ = gamma function

 $\mu_1, \mu_2 =$ mean of $H_1(x)$ and $H_2(y)$, respectively (time)

mean of $Q_m(n)$ (dimensionless) μ_N

= mean of $F_m(t)$ (time) μ_T

= correlation coefficient (dimensionless) $\rho(,)$

 σ_1 , σ_2 = standard deviation of $H_1(x)$ and $H_2(y)$, respectively

= standard deviation of $Q_m(n)$ (dimensionless) σ_N

= standard deviation of $F_m(t)$ (time) σ_T

 $\tau_1, \tau_2 =$ residence time in plug flow zone of region "1" and "2," respectively (time)

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