Characterization and Analysis of Continuous Recycle Systems

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A general analysis of continuous recycle systems and models is presented by considering the fluid history inside the systems. The history of a fluid element is expressed in terms of the number of cycles it completes, its residence time in the system and the total time it resides in a specific section of the systems. Concepts from probability theory are used to derive expressions for the number of cycles distribution (NCD), the residence time distribution (RTD), and the total regional residence time distribution (TRRTD) and their means and variances. Applications of these distributions in various processes are considered. Relationships between the number of cycles, the residence time, and the total regional residence time are expressed in terms of the covariances and correlation coefficient of pairs of these variables. These relationships allow the estimation of one variable for a given value of the other, and thus provide a mathematical means to completely describe the history of fluid in a recycle system.

Part I. Single Unit

SCOPE

This article presents a general analysis of continuous recycle systems and recycle flow models. The history of the fluid in a recycle system is considered in terms of the time a fluid element resides there, the number of cycles it completes, and the total time it spends in specific sections of the system. Three new concepts are introduced: the joint distribution of number of cycles and residence time, the number of cycle distribution (NCD), and total regional time distribution (TRRTD). These, together with the residence time distribution (RTD), the joint parameters of cycle time, number of cycles, and regional residence time derived here provide a mathematical means to characterize and analyze recycle processes and models.

Continuous recycle operations are commonplace in the chemical industry. Many processes such as catalytic cracking, particle coating, granulation and crystallization are based on recirculation. In many large reactors, a recycle stream is used to enhance mixing or improve selectivity. In addition, recycle flow models are commonly used to represent flow in large vessels and describe deviations from ideal mixing (Gibilaro 1971).

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At present, only the residence time distribution (RTD) is used to characterize recycle systems and construct recycle models.While the RTD is a useful tool in building and verifying recycle models and in characterizing certain recycle processes, there are many systems where the RTD is not an important process parameter. For example, in particle coating (Mann 1972, Mathur and Epstein 1974) the number of cycles a particle completes will determine the amount of coating accumulated. The number of cycles distribution (NCD) provides a means to express the coating uniformity. In systems where different regions are maintained at different conditions, the system performance may be more accurately related to the total time a fluid element resides in a certain region, rather than its residence time in the system as a whole. This is the case, for example, in continuous spouted-bed drying (Mathur and Epstein 1974), where different sections are maintained at different temperatures, and in reactors, where different sections exhibit different activities. These systems are characterized by the total regional residence time distribution (TRRTD). In general, detailed information on particle history should be very useful in interpreting data of complex particulate systems such as coal gasification and liquefaction, where several mechanisms occur simultaneously. This article provides the mathematical tools to characterize and analyze such systems.

CONCLUSIONS AND SIGNIFICANCE

The history of a fluid element in a continuous recycle system 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 a particular section of the system—each of these is a random variable. Concepts from probability theory are used to derive an expression for the joint distribution of number of cycles and residence time, in terms of the recycle ratio, the system geometry, and flow configurations. The NCD and the RTD are then derived as special cases of the joint distribution. It is shown that the NCD depends only on the recycle ratio, R, and is independent of the system geometry and the flow conditions. The mean of the NCD is $(R + 1)$ and its variance is $(R + 1)R$.

A general expression for the RTD is derived in terms of the recycle ratio and the flow parameters. Explicit expressions for some practical configurations whose RTDs have not been available previously are provided. Examples are given for systems whose two-flow regions are represented by either a plug-flow zone, a series of equal-size stirred tanks, or a combination of the two. Oscillation of the RTD, characteristic of recycle systems, is illustrated.

The total regional residence time distributions (TRRTD), in each of the system's two regions, are defined and derived. Expressions for their means and variances are provided. It is shown that each TRRTD depends on the recycle ratio and the flow configuration in the respective region. The mean of each TRRTD is equal to the product of the volume fraction of the respective region and the mean residence time in the system. Expressions for the variances of the TRRTDs are also provided.

The relationships between the number of cycles, residence time, and total regional residence times are expressed in terms of covariances and correlation coefficients of pairs of these variables. These allow the estimation of one variable for a given value of the second, and thus provide a mathematical means to describe completely the history of the fluid in a recycle system.

Continuous recycle systems such as the one shown schematically in Figure 1 are frequently encountered in the chemical industry. In many reactors a recycle stream is used to enhance mixing or improve selectivity. Numerous particulate processes, such as catalytic cracking, particle coating, granulation, crystallization and certain coal gasification processes are based on recirculation of particles. In addition, recycle flow models which are also represented by Figure 1 have received considerable attention in the chemical engineering literature. These models are used to describe mixing in large vessels and to express deviations from ideal mixing. Gibilaro (1971) provides a comprehensive review of the various recycle flow models and their applications.

At present, only the residence time distribution (RTD) is used to characterize recycle systems and to construct recycle models. While the RTD is an important tool in building and verifying flow models and characterizing certain recycle processes, there are many systems where the RTD is not an important parameter of the process. For example, in particle coating (Mann, 1972; Mathur and Epstein, 1974) the number of cycles a particle completes determines the amount of coating accumulated. In this case the number of cycles distribution (NCD) provides a means to express the coating uniformity in terms of system and operating parameters. In systems where different regions are maintained at different conditions, the performance may be more accurately related to the total time a fluid element resides in a certain region rather than to its residence time in the system as a whole. This is the case in continuous spouted-bed drying (Mathur and Epstein, 1974) where different sections are maintained at different temperatures. This system as well as reactors in which different sections exhibit different activity are characterized by the total regional residence time distribution (TRRTD). In general, detail information on particle history should be useful in interpreting data of complex particulate processes such as coal gasification and liquefaction where several mechanisms occur simultaneously.

This article provides a general analysis of continuous recycle systems. The history of a fluid element inside the system 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 a particular system section. Concepts from probability theory (see, for example, the survey of Seinfeld and Lapidus 1974) are used to derive expressions for the NCD, the RTD, the TRRTD, their means and variances. Relationships between the number of cycles, residence time and total regional residence times are expressed in terms of covariances and correlation coefficients of pairs of these variables. These relationships allow the estimation of one variable for a given value of the second and thus provide a mathematical means to describe completely the history of the fluid in the system.

JOINT DISTRIBUTION OF RESIDENCE TIME AND NUMBER OF CYCLES

Consider the general continuous recycle system shown in Figure 1. The system consists of two flow regions; region "1" with volume V_1 and flow rate w_1, located in the main flowline, and region "2" with volume V_2 and

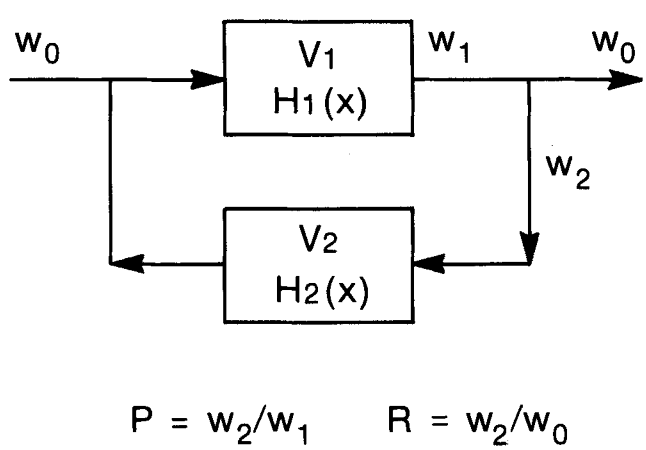

Figure 1. Schematic representation of a continuous recycle system.

flow rate w_2, located in the recycle line. Net flow rate through the system is $w_0 = w_1 - w_2$. It is assumed that the system is in steady-state, and that the fluid consists of identical elements whose mixing properties do not change within the system.

The RTD in region "1," $H_1(x)$, is defined as the probability that in a single passage, a fluid element resides there no longer than time x. Thus $H_1(x) = P\{X \leq x\}$ for $x \geq 0$, where X is a random variable, the time a fluid element resides in region "1" during a single passage. Since the fluid consists of identical elements, $H_1(x)$ may be interpreted as the fraction of fluid which resides in region "1" no longer than time x. The density of $H_1(x)$, when it exists, is $h_1(x) = dH_1(x)/dx$. Similarly, $H_2(y) = P\{Y \leq y\}$ is the RTD in region "2," where Y is a random variable which represents the time a fluid element resides in region "2" in a single passage. The density of $H_2(y)$, when it exists, is $h_2(y) = dH_2(y)/dy$. It is assumed that all fluid elements reside in each flow region a finite period of time and therefore $H_1(x)$ and $H_2(y)$ are proper distributions, i.e. $H_1(\infty) = H_2(\infty) = 1$.

In the analysis below, distribution functions rather than density functions are considered. This is done deliberately, because to impose the requirement that density functions exist narrows the scope of the analysis. There are systems in which the flow configurations are determined by switching mechanisms, in turn, controlled by the recycle ratio (see, for example, Rubinovitch and Mann 1979). Then, density functions do not exist, but useful results may still be obtained using distribution functions. The only condition imposed on $H_1(x)$ and $H_2(y)$ in the analysis is that they are proper distributions. This condition is fulfilled in all conceivable physical systems.

The history of a fluid element in a system is characterized by the number of cycles it completes and by the time it resides there. The number of cycles N, is defined as the number of times a fluid element passes through flow region "1." N is a discrete random variable which assumes integral values $(1, 2, \ldots)$. Let

$$q(n) = P\{N = n\} \qquad n = 1, 2, \ldots \tag{1}$$

be the probability function of N. When the fluid consists of identical elements, $q(n)$ can be interpreted as the fraction of fluid which completes exactly n cycles during its passage through the system. The distribution function of N is

$$Q(n) = P\{N \leq n\} = \sum_{j=1}^{n} q(j) \tag{2}$$

which is by definition the number of cycles distribution (NCD).

Since the mixing properties of the fluid do not change, the time a fluid element stays in each flow region on successive visits are mutually independent and independent of the number of cycles the element completes. Consequently, the residence time of an element in the system, T, is also a random variable and its distribution function is defined as

$$F(t) = P\{T \leq t\} \qquad (t > 0) \tag{3}$$

This is the RTD for the system. The density of the RTD, when it exists, is $f(t) = dF(t)/dt$.

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.,

$$G(n, t) = P\{N = n, T \leq t\} \tag{4}$$

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

$$F(t) = P\{T \leq t\} = P\{N < \infty, T \leq t\} = \sum_{n=1}^{\infty} G(n, t) \tag{5}$$

and

$$q(n) = P\{N = n\} = P\{N = n, T < \infty\} = \lim_{t \to \infty} G(n, t) \tag{6}$$

Let $p = w_2/w_1$ be the recycle fraction, then p is the probability that a fluid element recycles after leaving region "1." The term "recycle fraction" is used for p to distinguish it from the "recycle ratio," R, commonly used in the literature and defined as $R = w_2/w_0$. Clearly, $p = R/(R + 1)$.

The probability that an element completes exactly one cycle, i.e., passes only once through region "1," while residing in the system no longer than time t is given by

$$G(1, t) = P\{N = 1, T \leq t\} = (1 - p)H_1(t) \tag{7}$$

Similarly, the probability that an element completes exactly two cycles while residing in the system no longer than time t is given by

$$G(2, t) = P\{N = 2, T \leq t\} = (1 - p)p[H_1^{*2} * H_2](t) \tag{8}$$

where the asterisk denotes the convolution operator. The term $[H_1^{*2} * H_2](t)$ is the distribution function of the duration of two passages through region "1" and one passage through region "2" (see Appendix A). In general, the probability that a particle completes exactly n cycles $(n \geq 2)$ while residing in the system no longer than time t is given by

$$G(n, t) = P\{N = n, T \leq t\} = (1 - p)p^{n-1}[H_1^{*n} * H_2^{*(n-1)}](t) \tag{9}$$

It follows that the general expression for the joint distribution of N and T is

$$G(n, t) = \begin{cases} (1 - p)H_1(t) & n = 1 \\ (1 - p)p^{n-1}[H_1^{*n} * H_2^{*(n-1)}](t) & n \geq 2 \end{cases} \tag{10}$$

In principle, whenever p, $H_1(x)$, and $H_2(y)$ are known, $G(n, t)$ can be evaluated for any value of n and t. When $p = 0$, i.e., when the system has no recycle, $G(n, t) = H_1(t)$ which is then, the RTD of the system. When $p = 1$, i.e., for a closed circulating system, $G(n, t) = 0$ for all $n < \infty$ and $t < \infty$ and $G(\infty, \infty) = 1$. This means that all fluid elements complete infinite number of cycles and their residence time is unbounded.

In many cases, it is technically difficult to evaluate $G(n, t)$ explicitly. It is then useful to consider the joint transform

$$\hat{G}(z, s) = \sum_{n=1}^{\infty} z^n \int_0^{\infty} e^{-st} d_t G(n, t), \quad (|z| < 1, s > 0) \tag{11}$$

from which moments can be easily calculated. Substituting (10) into (11) and using properties of Laplace transform (Appendix B), one obtains

$$\hat{G}(z, s) = (1 - p)z\hat{H}_1(s) +$$

$$(1-p)\sum_{n=2}^{\infty} z^n p^{n-1}[\hat{H}_1(s)]^n[\hat{H}_2(s)]^{n-1} \quad (12)$$

where $\hat{H}_1(s)$ and $\hat{H}_2(s)$ are the Laplace transforms of $H_1(x)$ and $H_2(y)$, respectively. For each fixed value of s, $pz\hat{H}_1(s)\hat{H}_2(s) < 1$ and (12) is reduced to

$$\hat{G}(z,s) = \frac{(1-p)z\hat{H}_1(s)}{1 - zp\hat{H}_1(s)\hat{H}_2(s)} \quad (13)$$

Equations (10) and (13) are two key relations which completely characterize continuous recycle systems. It will be shown below how the NCD, the RTD, their moments, as well as useful expressions for the relationship between N and T, can be derived from them.

THE DISTRIBUTION OF THE NUMBER OF CYCLES

The probability function of N can be derived directly or obtained from $G(n, t)$ using (6). Noting that $H_1(\infty) = H_2(\infty) = [H_1^{*n} * H_2^{*(n-1)}](\infty) = 1$ (see Appendix A), (10) becomes

$$q(n) = P\{N = n\} = (1-p)p^{n-1} \quad (n \geq 1) \quad (14)$$

The NCD, $Q(n)$, is thus given by

$$Q(n) = P\{N \leq n\} = \sum_{j=1}^{n} q(n) = 1 - p^n \quad (15)$$

The mean and the variance of the NCD are then

$$\mu_N = E[N] = \sum_{n=1}^{\infty} nq(n) = \frac{1}{1-p} = R + 1 \quad (16)$$

and

$$\sigma_N^2 = Var[N] = \sum_{n=1}^{\infty} (n - \mu_N)^2 q(n) = \frac{p}{(1-p)^2} = R(R+1) \quad (17)$$

The probability function of N, given by (14), is the geometric distribution with parameters p and $(1 - p)$. Hence, the NCD is a function of the recycle fraction (or recycle ratio) only and does not depend on the system geometry, fluid properties or the flow patterns.

Note that when $p = 0$ (no recycle), $q(1) = 1$ and $q(n) = 0$ for $n \neq 1$. Thus, the fluid visits flow region "1" only once. When $p = 1$ (closed circulating system), $Q(n) = 0$ for $n < \infty$, i.e., each fluid element completes an infinite number of cycles.

The generating function of the NCD is

$$\hat{q}(z) = \sum_{n=1}^{\infty} z^n q(n) = \frac{(1-p)z}{1-zp} \quad (18)$$

This can be obtained either directly from (14) or by setting $s = 0$ in (13).

RESIDENCE TIME DISTRIBUTION

The RTD of the system is obtained from the joint distribution by substituting (10) into (5). This gives

$$F(t) = (1-p)H_1(t) + (1-p)\sum_{n=2}^{\infty} p^{n-1}[H_1^{*n} * H_2^{*(n-1)}](t) \quad (19)$$

which is the general expression of the RTD in terms of the recycle fraction and the individual single-path RTDs of the two flow regions. Useful expressions can be obtained for certain systems whose RTD expressions have not been available (Clegg and Coates 1967, Gibilaro 1971) by expressing the convolution terms explicity. Before such expressions are discussed, we first examine $F(t)$ in the limiting cases, when $p = 0$ and $p = 1$, and derive expressions for its means and variances.

When $p = 0$ (no recycle), $F(t) = H_1(t)$, whereas when $p = 1$ (closed circulating system), $F(t) = 0$. The results of both limiting cases are in agreement with the physical situation. In the former case only flow region "1" is in operation whereas in the latter case the residence time in the system is infinite as $F(t) = 0$ for any $t < \infty$.

The Laplace transform of the RTD is obtained either directly from (19) or from (13). It is given by

$$\hat{F}(s) = \hat{G}(1,s) = \frac{(1-p)\hat{H}_1(s)}{1 - p\hat{H}_1(s)\hat{H}_2(s)} \quad (20)$$

where $\hat{F}(s)$ is the transfer function of the system as defined, for example, by Gibilaro (1971). It depends on the recycle fraction, p, and the Laplace transforms of $H_1(x)$ and $H_2(y)$. The mean, μ_T, and the variance, σ_T^2, of the RTD are derived from (20) using methods described in Appendix B. The results are

$$\mu_T = E[T] = \frac{1}{1-p}(\mu_1 + p\mu_2) \quad (21)$$

$$\sigma_T^2 = \text{Var}[T] = \frac{p}{(1-p)^2}(\mu_1 + \mu_2)^2 + \frac{1}{1-p}(\sigma_1^2 + p\sigma_2^2) \quad (22)$$

where 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 $\mu_1 = V_1/w_1$, $\mu_2 = V_2/w_2$, $V_0 = V_1 + V_2$, $w_2 = pw_1$ and $w_2 = w_1 - w_0$, it follows from (21), that the mean residence time in the system is V_0/w_0. Also, when $p = 0$, (21) and (22) are reduced to $\mu_T = \mu_1$ and $\sigma_T^2 = \sigma_1^2$, respectively. When $p = 1$, μ_T is infinite.

The last two relations are especially useful in determining the parameters of the RTD, without the need to derive an expression for the RTD itself. They are also useful in estimating the recycle ratio and the configuration of a system from RTD measurements. This point is discussed at the end of this section.

When the densities of $H_1(x)$ and $H_2(y)$ exist, one can obtain a useful expression for the density of RTD by rewriting (19) as (See Appendix A),

$$f(t) = (1-p)h_1(t) + (1-p)\sum_{n=2}^{\infty} p^{n-1}\int_0^t h_1^{*n}(x)\, h_2^{*(n-1)}(t-x)dx \quad (23)$$

$$[h_1^{*n}(x) * h_2^{*(n-1)}](t-x)dx \quad (23)$$

In principle, whenever, p, $h_1(x)$ and $h_2(y)$ are known, the RTD can be calculated directly from (23). However, computations may be quite difficult since they involve evaluations of n-th order convolutions and infinite series of integrals. Fortunately, for many commonly-used recycle flow models simplified expressions can be derived. These are illustrated here for systems whose flow regions are represented by either, a series of equal-size stirred tanks, a plug flow region, or a combination of the two.

When a flow region is represented by a series of α equal-size stirred tanks having total volume V_m, connected in series to a plug flow zone with volume, V_p, the RTD of the region is a shifted gamma distribution whose density is

$$h(x) = \begin{cases} 0 & (x < \tau) \\ \dfrac{1}{\Gamma(\alpha)} \dfrac{1}{\beta} \left(\dfrac{x-\tau}{\beta} \right)^{\alpha-1} e^{-(x-\tau)/\beta} & (x \geq \tau) \end{cases} \tag{24}$$

Here, $\tau = V_p/w$ is the retention time in the plug-flow zone, w is the volumetric flow rate in the region and $\beta = V_m/\alpha w$ is the mean residence time in each stirred tank. The mean of this distribution is $\tau + \alpha\beta = (V_m + V_p)/w$ and its variance is $\alpha\beta^2$.

Noting that the n-th order convolution of a gamma distribution with parameters α and β is a gamma distribution with parameters $n\alpha$ and β (Feller 1971), it can be easily shown that the n-th order convolution of a shifted gamma distribution is

$$h^{*n}(x) = \begin{cases} 0 & (x < n\tau) \\ \dfrac{1}{\Gamma(n\alpha)} \dfrac{1}{\beta} \left(\dfrac{x-n\tau}{\beta} \right)^{n\alpha-1} e^{-(x-n\tau)/\beta} & (x \geq n\tau) \end{cases} \tag{25}$$

Note that when $V_p = 0$ (no plug flow zone) $\tau = 0$, (24) reduces to a gamma density with parameters α and β and (25) reduces to a gamma density with parameters $n\alpha$ and β. When $V_m = 0$ (the region consists of a plug flow zone only), $\beta = 0$, (24) reduces to $h(x) = \delta(x - \tau)$ and (25) becomes $h^{*n}(x) = \delta(x - n\tau)$.

These results can be substituted into (23) to derive explicit RTD expressions for many commonly-used recycle models. For convenience, these models are divided into four cases shown schematically in Figure 2, each leading to a simplified analytical expression which can be easily calculated:

Case A: Both flow regions are represented by plug flow zones. Here, $h_1(x) = \delta(x - \tau_1)$ and $h_2(y) = \delta(y - \tau_2)$, where $\tau_1 = V_1/w_1$ and $\tau_2 = V_2/w_2$, and

$$f(t) = (1-p)\delta(t-\tau_1) + (1-p) \sum_{n=2}^{\infty} p^{n-1}\delta(t - n\tau_1 - (n-1)\tau_2) \tag{26}$$

This is a discrete distribution of spikes with height $(1-p)p^{n-1}$ at times $n\tau_1 + (n-1)\tau_2$, $n = 1, 2, \ldots$. The mean and variance of this RTD are

$$\mu_T = \frac{1}{1-p} (\tau_1 + p\tau_2) \tag{27}$$

and

$$\sigma_T^2 = \frac{p}{(1-p)^2} (\tau_1 + \tau_2)^2 \tag{28}$$

For the special case when $\tau_2 = 0$, i.e., an instantaneous recycle, (26) reduces to

$$f(t) = (1-p)\delta(t-\tau_1) + (1-p) \sum_{n=2}^{\infty} p^{n-1}\delta(t - n\tau_1) \tag{29}$$

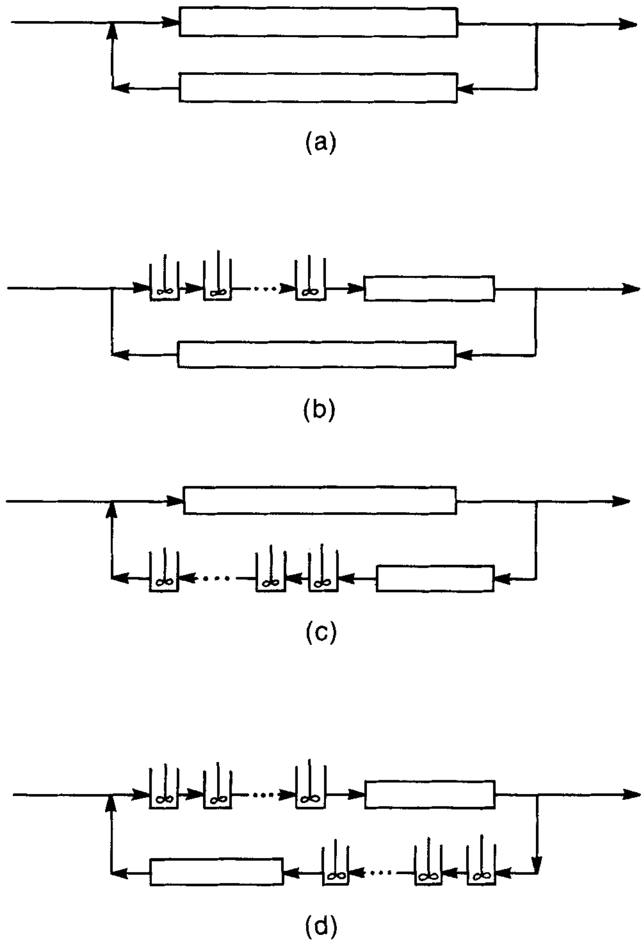

Figure 2. Four cases of RTD calculation.

which is identical to an expression derived by Rippin (1967).

Case B: Region "1" is represented by a combination of α_1 equal-size stirred tanks and a plug flow zone. Region "2" is represented by plug flow zone. Let V_{1p} and V_{1m} be the volumes of the plug-flow and mixing sections, respectively, in region "1" where $V_1 = V_{1p} + V_{1m}$ and let V_2 be the volume of region "2." For this case

$$h_1(x) = \frac{1}{\Gamma(\alpha_1)} \frac{1}{\beta_1} \left(\frac{x-\tau_1}{\beta_1} \right)^{\alpha_1-1} e^{-(x-\tau_1)/\beta_1} \quad (x \geq \tau_1) \tag{30}$$

$$h_2(y) = \delta(y - \tau_2) \tag{31}$$

where $\tau_1 = V_{1p}/w_1$, $\beta_1 = V_{1m}/\alpha_1 w_1$ and $\tau_2 = V_2/w_2$, and (23) reduces to

$$f(t) = (1-p) \frac{1}{\Gamma(\alpha_1)} \frac{1}{\beta_1} \left(\frac{t-\tau_1}{\beta_1} \right)^{\alpha_1-1} e^{-(t-\tau_1)/\beta_1} + (1-p) \sum_{n=2}^{\infty} p^{n-1} \frac{1}{\Gamma(n\alpha_1)} \frac{1}{\beta_1} \cdot \left(\frac{t - n\tau_1 - (n-1)\tau_2}{\beta_1} \right)^{n\alpha_1-1} e^{-(t-n\tau_1-(n-1)\tau_2)/\beta_1} \tag{32}$$

The mean and variance of this RTD are

$$\mu_T = \frac{1}{1-p} [(\tau_1 + \alpha_1\beta_1) + p\tau_2] \tag{33}$$

and

$$\sigma_T^2 = \frac{p}{(1-p)^2}(\tau_1 + \alpha_1\beta_1 + \tau_2)^2 + \frac{1}{1-p}(\alpha_1\beta_1)^2 \tag{34}$$

Figures 3 and 4 show $f(t)$ of two illustrative systems for Case B. For the special case when $\tau_2 = 0$ (instantaneous recycle), (32) reduces to

$$f(t) = (1-p)\frac{1}{\Gamma(\alpha_1)}\frac{1}{\beta_1}\left(\frac{t-\tau_1}{\beta_1}\right)^{\alpha_1-1} e^{-(t-\tau_1)/\beta_1} + (1-p)\sum_{n=2}^{\infty} p^{n-1}\frac{1}{\Gamma(\alpha_1)}\frac{1}{\beta_1}\left(\frac{t-\tau_1}{\beta_1}\right)^{n\alpha_1-1} e^{-(t-n\tau_1)/\beta_1} \tag{35}$$

which is identical to an expression derived by Fu et al. (1971).

Case C: Region "1" is represented by a plug flow zone and region "2" by a combination of α_2 equal-size stirred tanks and a plug-flow zone. Let V_1 be the volume of region "1" and let V_{2p} and V_{2m} be, respectively, the volume of the plug-flow and the mixing sections in region "2", where $V_2 = V_{2p} + V_{2m}$. For this case

$$h_1(x) = \delta(x - \tau_1) \tag{36}$$

$$h_2(y) = \frac{1}{\Gamma(\alpha_2)}\frac{1}{\beta_2}\left(\frac{y-\tau_2}{\beta_2}\right)^{\alpha_2-1} e^{-(y-\tau_2)/\beta_2} \quad (y \geq \tau_2) \tag{37}$$

where $\tau_1 = V_1/w_1$, $\tau_2 = V_{2p}/w_2$ and $\beta_2 = V_{2m}/\alpha_2 w_2$, and (23) reduces to

$$f(t) = (1-p)\delta(t-\tau_1) + (1-p)\sum_{n=2}^{\infty} p^{n-1}\frac{1}{\Gamma((n-1)\alpha_2)}\frac{1}{\beta_1}\left(\frac{t-n\tau_1-(n-1)\tau_2}{\beta_2}\right)^{(n-1)\alpha_2-1} e^{-(t-n\tau_1-(n-1)\tau_2)/\beta_2} \tag{38}$$

The mean and the variance of this RTD are

$$\mu_T = \frac{1}{1-p}[\tau_1 + p(\tau_2 + \alpha_2\beta_2)] \tag{39}$$

and

$$\sigma_T^2 = \frac{p}{(1-p)^2}(\tau_1 + \tau_2 + \alpha_2\beta_2)^2 + \frac{1}{1-p}(p\alpha_2\beta_2)^2 \tag{40}$$

Figure 5 illustrates the RTD of one illustrative system of Case C.

Case D: Each region is represented by a combination of a series of equal-size stirred tanks and a plug-flow zone; region "1" by α_1 tanks and region "2" by α_2 tanks. Let V_{1p} and V_{1m} be, respectively, the volumes of the plug-flow and the mixing zones in region "1" and V_{2p} and V_{2m} be the volume of the plug-flow and the mixing zones in region "2," where $V_1 = V_{1p} + V_{1m}$, $V_2 = V_{2p} + V_{2m}$ and $V_0 = V_1 + V_2$. For this case

$$h_1(x) = \frac{1}{\Gamma(\alpha_1)}\frac{1}{\beta_1}\left(\frac{x-\tau_1}{\beta_1}\right)^{\alpha_1-1} e^{-(x-\tau_1)/\beta_1} \quad (x \geq \tau_1) \tag{41}$$

$$h_2(y) = \frac{1}{\Gamma(\alpha_2)}\frac{1}{\beta_2}\left(\frac{y-\tau_2}{\beta_2}\right)^{\alpha_2-1} e^{-(y-\tau_2)/\beta_2} \quad (y \geq \tau_2) \tag{42}$$

where $\tau_1 = V_{1p}/w_1$, $\beta_1 = V_{1m}/\alpha_1 w_1$, $\tau_2 = V_{2p}/w_2$ and $\beta_2 = V_{2m}/\alpha_2 w_2$, and (23) reduces to

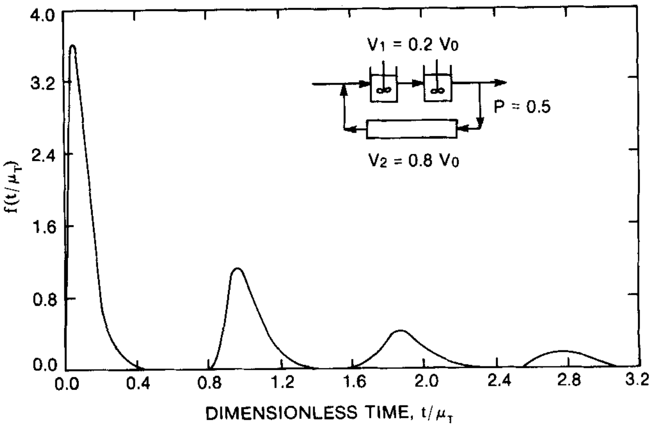

Figure 3. RTD illustration for case B.

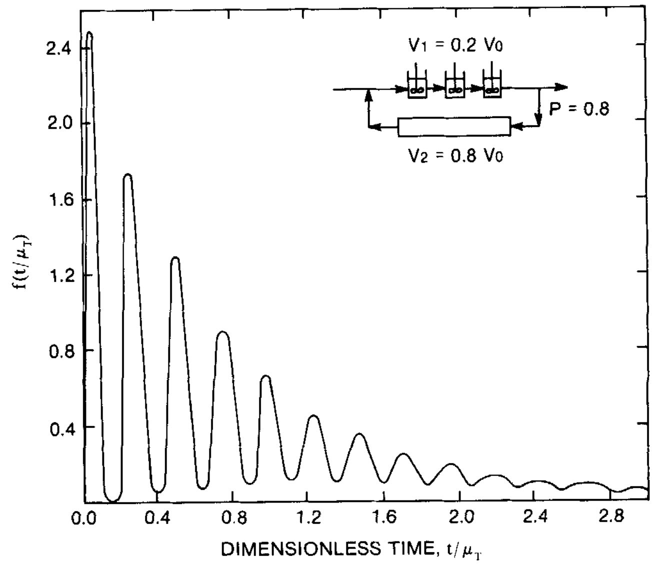

Figure 4. RTD illustration for case B.

$$f(t) = (1-p)\frac{1}{\Gamma(\alpha_1)}\frac{1}{\beta_1}\left(\frac{t-\tau_1}{\beta_1}\right)^{\alpha_1-1} e^{-(t-\tau_1)/\beta_1} + (1-p)\sum_{n=2}^{\infty} p^{n-1}\frac{1}{\Gamma(n\alpha_1)}\frac{1}{\Gamma((n-1)\alpha_2)}\frac{1}{\beta_1}\frac{1}{\beta_2}\cdot \int_{t-n\tau_1-(n-1)\tau_2}^{t}\left(\frac{x-n\tau_1}{\beta_1}\right)^{n\alpha_1} \; {}^{-(x-\tau_1)/\beta_1}$$

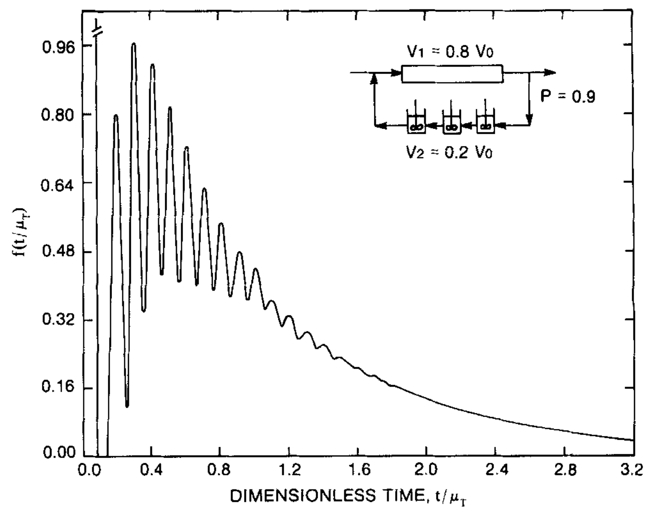

Figure 5. RTD illustration for case C.

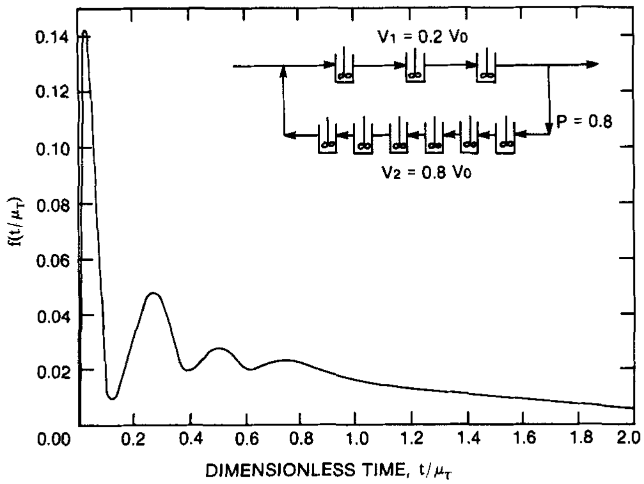

Figure 6. RTD illustration for case D.

$$\left(\frac{t - n\tau_1 - (n-1)\tau_2 - x}{\beta_2}\right)^{(n-1)\alpha_1 - 1} e^{-(t - n\tau_1 - (n-1)\tau_2 - x)\beta_2} dx \quad (43)$$

The mean and the variance of the RTD for this case are

$$\mu_T = \frac{1}{1-p}\left[\tau_1 + \alpha_1\beta_1 + p(\tau_2 + \alpha_2\beta_2)\right] \quad (44)$$

and

$$\sigma_T^2 = \frac{p}{(1-p)^2}(\tau_1 + \alpha_1\beta_1 + \tau_2 + \alpha_2\beta_2)^2 + \frac{1}{1-p}(\alpha_1\beta_1^2 + p\alpha_2\beta_2^2) \quad (45)$$

Figure 6 shows the $f(t)$ of one illustrative system for Case D.

Note that all the RTD densities calculated above exhibit oscillations. In fact, these oscillations are characteristic of recycle systems although in many instances, the oscillations are damped because of internal mixing in the regions. When oscillations are experimentally observed in RTD measurements they indicate that the fluid is recirculating in the system (Levenspiel 1972). Useful information on the system configuration and the parameters of a recycle flow model can be obtained from the frequency (or cycle time) of these oscillations.

Mann et al. (1973) have shown that the mean cycle time in a closed, circulating system is

$$\mu_c = \mu_1 + \mu_2 \quad (46)$$

The same relation also applies for a continuous recycle system, since as long as a fluid element remains within the system, it has the same characteristics as an element in a closed circulating system. The only difference between the two systems is that the element can leave a continuous system at any time, whereas its residence time in closed circulating system is infinite. For a given recycle system with volume V_0 and flow rate w_0, the mean cycle time is thus given by

$$\mu_c = \frac{1-p}{w_0}\left(V_1 + \frac{V_2}{p}\right) = (1-p)\mu_T + \frac{(1-p)^2}{p}\frac{V_2}{w_0} \quad (47)$$

Hence, when μ_c is experimentally measured and V_1 and V_2 are known one can easily determine p directly from (47). Similarly, when p is known, V_2 and V_1 can be calculated. In general, when μ_T and μ_c are known or experimentally measured, one can use (21) and (47) to estimate the parameters of a recycle model without using the RTD expression. By using these relationships, the number of unknown configuration parameters of a recycle model to be determined is reduced from three (p, V_1, V_2 or p, μ_1, μ_2) to one.

TOTAL REGIONAL RESIDENCE TIME DISTRIBUTIONS

As indicated above, in many cases a recycle process depends on the total time a fluid element stays in a certain section of the system—not on its residence time in the system as a whole or its number of cycles. These cases are conveniently characterized by the total regional residence time distributions (TRRTDs). How such distributions are obtained and a discussion of their properties in region "1" and "2" follows.

The total time a fluid element resides in region "1" during its passage through the system, T_1, is a random variable. It may vary from particle to particle and passage to passage. Total regional residence time distribution (TRRTD) in region "1", $F_1(t)$, is thus defined by

$$F_1(t) = P\{T_1 \leq t\} \quad (48)$$

An expression for $F_1(t)$, can be derived using an argument similar to the one in the derivation of the joint distribution. However, this can also be obtained directly from (19), by noting that total residence time in region "1" is equal to the residence time in the system, if the residence time in region "2" were zero. By substituting a unit step function at the origin for $H_2(y)$ into (19) one obtains

$$F_1(t) = (1-p)H_1(t) + (1-p)\sum_{n=2}^{\infty} p^{n-1}[H_1^{*n}](t) \quad (49)$$

Similarly, the Laplace transform of $F_1(t)$ is obtained by substituting $\hat{H}_2(s) = 1$ into (20), viz.

$$\hat{F}_1(s) = \frac{(1-p)\hat{H}_1(s)}{1 - p\hat{H}_1(s)} \quad (50)$$

Note that $F_1(t)$ and its parameters depend only on p and $H_1(x)$ and not on $H_2(y)$. It follows that the mean and variance of $F_1(t)$ are

$$E[T_1] = \frac{1}{1-p}\mu_1 \quad (51)$$

and

$$\mathrm{Var}[T_1] = \frac{p}{(1-p)^2}\mu_1^2 + \frac{1}{1-p}\sigma_1^2 \quad (52)$$

Since $\mu_1 = V_1/w_1 = (1-p)\,V_1/w_0$ and $\mu_T = (V_1 + V_2)/w_0$, (51) becomes

$$E[T_1] = \frac{V_1}{w_0} = \frac{V_1}{V_1 + V_2}\mu_T \quad (53)$$

Hence, the mean of the total residence time in region "1" is proportional to the volume fraction of that region and the mean residence time in the system.

The TRRTD in region "2" and its parameters may be obtained in a similar way, i.e.,

$$F_2(t) = P\{T_2 \leq t\} = (1-p) +$$

$$(1-p) \sum_{n=2}^{\infty} p^{n-1}[H_2^{*(n-1)}](t) \quad (54)$$

The Laplace transform of $F_2(t)$ is

$$\hat{F}_2(s) = \frac{1-p}{1-p\hat{H}_2(s)} \quad (55)$$

and its mean and variance are

$$E[T_2] = \frac{p}{1-p}\mu_2 \quad (56)$$

$$\text{Var}[T_2] = \frac{p}{(1-p)^2}\mu_2^2 + \frac{p}{(1-p)}\sigma_2^2 \quad (57)$$

It is easy to verify that

$$E[T_2] = \frac{V_2}{w_0} = \frac{V_2}{V_1+V_2}\mu_T \quad (58)$$

Figures 7 and 8 show the densities of $F_1(t)$, $F_2(t)$ and $F(t)$ for two illustrative recycle systems.

JOINT PARAMETERS

In the previous three sections it has been shown how the NCD, RTD, and TRRTD and their parameters can be evaluated. While these distributions are useful in many cases, they do not provide complete information on the history of a fluid element in the system. In processes where more than one of these variables is important, the joint behavior of N and T, N and T_1, T_1 and T_2, etc. should be considered. Such behavior is expressed by the joint distribution of the interesting pair of variables. In principle, one can derive and compute such a joint distribution. However, in many cases the explicit expressions are complicated and cannot be evaluated numerically. In such cases useful information on the joint behavior of each pair of variables can be obtained from the covariance and correlation coefficient of that pair.

The covariance of N and T is defined as

$$\text{COV}[N,T] = E[(N-E[N])(T-E[T])] \quad (59)$$

and it can be conveniently evaluated from the relation

$$\text{COV}[N,T] = E[N\cdot T] - E[N]E[T] \quad (60)$$

The term $E[N\cdot T]$ is defined by

$$E[N\cdot T] = \sum_{n=1}^{\infty} \int_0^{\infty} n t d_t P\{N=n, T\leq t\} \quad (61)$$

which can be calculated from $\hat{G}(z,s)$ by

$$E[N\cdot T] = -\frac{d}{ds}\frac{d}{dz}\hat{G}(z,s)\Big|_{\substack{z=1\\s=0}} \quad (62)$$

Thus, an expression for the covariance of N and T is obtained by substituting (13) into (62), the latter into (60) and using (16) and (21). The result is

$$\text{COV}[N,T] = \frac{p}{(1-p)^2}(\mu_1+\mu_2) \quad (63)$$

The correlation coefficient of N and T is defined by

$$\rho(N,T) = \frac{\text{COV}[N,T]}{(\text{Var}[N])^{1/2}(\text{Var}[T])^{1/2}} \quad (64)$$

Substituting (63), (17), and (22) into (64) one obtains

$$\rho(N,T) = \left[1 + \frac{1-p}{p}\frac{(\sigma_1^2+p\sigma_2^2)}{(\mu_1+\mu_2)^2}\right]^{-1/2} \quad (65)$$

For recycle systems, the correlation coefficient of N and T varies between 0 and 1; when $p = 0$, (no recycle), $\rho(N,T) = 0$, and when $p = 1$ (batch circulating system), $\rho(N,T) = 1$. For small values of p, the number of cycles is very small, and the value of N provides little information on T. When p is large, N varies over a wide range and, thus, for each value of N one can obtain estimates for T. Statistical procedures for such estimation problems will be discussed in a separate article.

The covariance and correlation coefficient of N and T_1 can be obtained by first deriving the joint distribution of N and T_1 and then following the procedure used in obtaining the covariance of N and T. However, they are obtained directly from (63) and (65) by substituting $\mu_2 = 0$ and $\sigma_2 = 0$, viz.

$$\text{COV}(N,T_1) = \frac{p}{(1-p)^2}\mu_1^2 \quad (66)$$

$$\rho(N,T_1) = \frac{\mu_1}{\left(\mu_1^2 + \frac{1-p}{p}\sigma_1^2\right)^{1/2}} \quad (67)$$

Similarly, the covariance and correlation coefficient of N and T_2 are obtained from (63) and (65) by substituting $\mu_1 = 0$ and $\sigma_1 = 0$, viz.

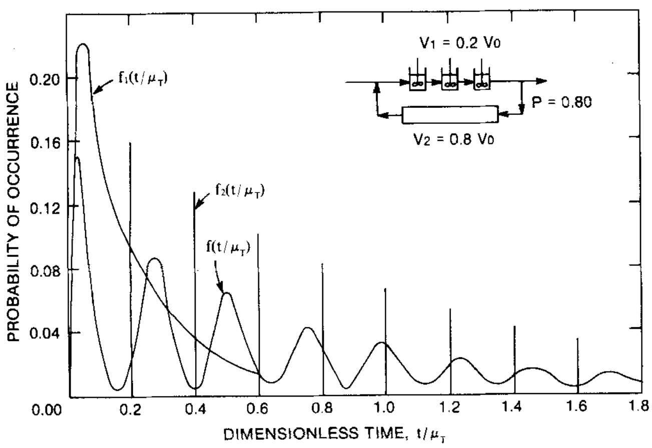

Figure 7. RTD and TRRTDs illustration for case B.

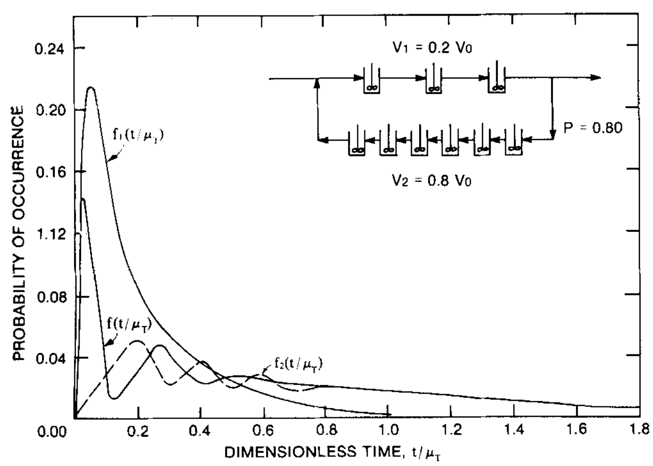

Figure 8. RTD and TRRTDs illustration for case D.

$$\mathrm{COV}[N, T_2] = \frac{p}{(1-p)^2}\mu_2 \tag{68}$$

and

$$\rho(N, T_2) = \frac{\mu_2}{[\mu_2^2 + (1-p)\sigma_2^2]^{1/2}} \tag{69}$$

The covariance of T_1 and T_2 is obtained by noting that $T = T_1 + T_2$, and using the relation

$$\mathrm{Var}[T] = \mathrm{Var}[T_1] + \mathrm{Var}[T_2] + 2\,\mathrm{COV}[T_1, T_2] \tag{70}$$

Since all parameters except the covariance are known, one can obtain it by substituting (22), (52), and (57) into (70), viz.

$$\mathrm{COV}[T_1, T_2] = \frac{p}{(1-p)^2}\mu_1\mu_2 \tag{71}$$

The correlation coefficient of T_1 and T_2 defined as

$$\rho(T_1, T_2) = \frac{\mathrm{COV}[T_1, T_2]}{(\mathrm{Var}[T_1])^{1/2}(\mathrm{Var}[T_2])^{1/2}} \tag{72}$$

is given by

$$\rho(T_1, T_2) = \frac{p\mu_1\mu_2}{(p\mu_1^2 + (1-p)\sigma_1^2)^{1/2}(p\mu_2^2 + (1-p)p\sigma_2^2)^{1/2}} \tag{73}$$

The relationships derived above completely describe the history of a fluid element in the system. For a fluid element residing in the system a given time t, one can estimate with a specified confidence level what number of cycles it completed, its total residence time in region "1" and its total regional time in region "2."

CONCLUDING REMARKS

The foregoing results provide mathematical means to describe completely the history of a fluid in continuous recycle systems. This article has been directed primarily at introducing the concepts of joint distribution of N and T, NCD, and TRRTD, deriving useful expressions for the NCD, RTD, and the TRRTD, discussing their properties and obtaining relations between N and T, N and T_1, or T_2, and T_1 and T_2. Potential applications of these results are numerous, especially for particulate processes such as coal gasification and liquefaction where particles undergo a change via several simultaneous mechanisms (reaction, agglomeration, attrition, etc.). Knowledge of the particle history in the process should be useful in interpretating data and developing understanding of complex processes.

Part II of this article extends the analysis to a cascade of recycle units.

ACKNOWLEDGMENT

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NOTATION

$F(t), F_1(t), F_2(t)$ = total residence time distribution for the system, region "1" and region "2" respectively (dimensionless)

$\hat{F}(s), \hat{F}_1(s), \hat{F}_2(s)$ = Laplace transform of $F(t)$, $F_1(t)$, and $F_2(t)$ respectively (dimensionless)

$f(t), f_1(t), f_2(t)$ = densities of $F(t)$, $F_1(t)$, and $F_2(t)$, respectively (time^{-1})

$G(n, t)$ = joint distribution of the residence time and the number of cycles defined by Equation (4) (dimensionless)

$H_1(x), H_2(y)$ = residence time distribution in flow regions "1" and "2," respectively (dimensionless)

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

$h_1(x), h_2(y)$ = densities of $H_1(x)$ and $H_2(x)$, respectively (time^{-1})

j, n = integer

N = number of cycles or number of passages in region "1" (dimensionless)

p = recycle fraction, w_2/w_1 (dimensionless)

$Q(n)$ = distribution of N, NCD, (dimensionless)

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

R = recycle ratio, w_2/w_0 (dimensionless)

s = variable of Laplace transformation

T, T_1, T_2 = total residence time in the system, region "1" and region "2," respectively (time)

t = time (time)

V_0, V_1, V_2 = volume of the system, region "1" and region "2," respectively

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

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

w_0, w_1, w_2 = net volumetric flow rate through the system, region "1" and region "2", respectively (volume/time)

X, Y = residence time in regions "1" and "2" (time)

x, y = time

z = transform variable (dimensionless)

Greek Letters

$\alpha, \alpha_1, \alpha_2$ = parameters of gamma distribution

β, β_1, β_2 = parameter of gamma distribution

$\Gamma(\,)$ = gamma function

μ_1, μ_2 = mean of $H_1(x)$ and $H_2(y)$, respectively (time)

μ_c = mean cycle time (time)

μ_N = mean of $Q(n)$ (dimensionless)

μ_T = mean of $F(t)$ (time)

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

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

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

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

τ, τ_1, τ_2 = parameters of shifted gamma distribution

APPENDIX A: THE CONVOLUTION OPERATOR

Let $H_1(x)$ be the distribution function of a non-negative random variable X and let $H_2(y)$ be the distribution function of another non-negative random variable Y. The convolution of $H_1(x)$ and $H_2(y)$ is defined (see for example Feller 1971) by

$$[H_1 * H_2](t) = \int_0^t H_1(t-y)\, dH_2(y) = \int_0^t H_2(t-x)\, dH_1(x) \tag{A-1}$$

It has been shown that the convolution of distribution functions is both commutative and associative. Furthermore, if X and Y are independent, the distribution function of $X + Y$ is $[H_1 * H_2](t)$, viz.

$$P\{X + Y \leq t\} = [H_1 * H_2](t) \tag{A-2}$$

If $h_1(x)$ and $h_2(y)$ are the densities of $H_1(x)$ and $H_2(y)$, the density of the convolution $[H_1 * H_2](t)$ is given by

$$[h_1 * h_2](t) = \int_0^t h_1(t-y)\, h_2(y)\, dy$$
$$= \int_0^t h_2(t-x)\, h_1(x)\, dx \quad \text{(A-3)}$$

The distribution function of the sum of n mutually independent random variables, $X_1 + X_2 + \cdots + X_n$, with a common distribution $H(x)$ is the n-fold convolution of $H(x)$ with itself which is denoted by $[H^{*n}](t)$. Hence,

$$[H^{*1}](t) = H(t),$$

$$[H^{*2}](t) = [H * H](t) = \int_0^t H(t-x)\, dH(x),$$

and

$$[H^{*(n-1)}](t) = [H^{*n} * H](t) = \int_0^t [H^{*n}]\,(t-x)\, dH(x) \quad \text{(A-4)}$$

Similarly, if $h(x)$ is the density function of $H(x)$,

$$[h^{*1}] = h(t),$$

$$[h^{*2}](t) = [h * h](t) = \int_0^t h(t-x)\, h(x)\, dx,$$

and

$$[h^{*(n-1)}](t) = [h^{*n} * h](t) = \int_0^t [h^{*n}](t-x)\, h(x)\, dx \quad \text{(A-5)}$$

where $[h^{*n}](t)$ is the density of the convolution $[H^{*n}](t)$.

In general, $[H_1{}^{*n} * H_2{}^{*k}](t)$ is the probability that the sum of n random variables, $X_1, X_2, \ldots, X_n$ having common distribution $H_1(x)$ and k random variables $Y_1, Y_2, \ldots, Y_k$ having distribution $H_2(y)$ is not larger than t, i.e.,

$$[H_1{}^{*n} * H_2{}^{*k}](t) = P\{X_1 + X_2 + \cdots + X_n + Y_1 + Y_2 + \cdots + Y_k \leq t\} \cdot \quad \text{(A-6)}$$

In accordance with the operational procedure used in (A-5), $[H_1{}^{*n} * H_2{}^{*k}](t)$ is calculated by

$$[H_1{}^{*n} * H_2{}^{*k}](t) = \int_0^t [H_1{}^{*n}]\,(t-y)\, d[H_2{}^{*k}]\,(y)$$
$$= \int_0^t [H_2{}^{*k}]\,(t-x)\, d[H_1{}^{*n}](x) \quad \text{(A-7)}$$

Finally, since $H_1(\infty) = H_2(\infty) = 1$, it follows from (A-1) that $[H_1 * H_2](\infty) = 1$ and, in general, $[H^{*n}](\infty) = 1$ and $[H_1{}^{*n} * H_2{}^{*k}](\infty) = 1$.

APPENDIX B: GENERATING FUNCTIONS AND LAPLACE TRANSFORM

Let $H(x)$ be a distribution function of a non-negative random variable X and let $h(x)$ be its density. The Laplace transform of $H(x)$ is defined (see for example Feller 1971) by

$$\hat{H}(s) = \int_0^\infty e^{-sx} \frac{d}{dx} P\{X \leq x\} = \int_0^\infty e^{-sx}\, dH(x)$$
$$= \int_0^\infty e^{-sx} h(x)\, dx \quad \text{(B-1)}$$

where s is the transform variable and the symbol $\wedge$ denotes the transformed form. It follows from (B-1) that $\hat{H}(0) = 1$. The Laplace transform of the convolution $[H_1 * H_2]\,(t)$ is derived directly from the definitions of the Laplace transform and the convolutions to give

$$[H_1 \hat{*} H_2]\,(s) = \hat{H}_1\,(s) \cdot \hat{H}_2\,(s) \quad \text{(B-2)}$$

and in general

$$[H \hat{*} n]\,(s) = [\hat{H}(s)]^n \quad \text{(B-3)}$$

The moments of a distribution function, $H(x)$, are evaluated from its Laplace transform by

$$M_k = (-1)^k \frac{d^k}{ds^k} \hat{H}(s)\,|_{s=0} \quad \text{(B-4)}$$

where M_k is the k-th moment.

Let $g(n)$ be a discrete probability density function. The generating function of $g(n)$ is defined (see for example Feller 1968) by

$$\hat{g}(z) = \sum_{n=0}^{\infty} z^n g(n), \quad (|z| < 1) \quad \text{(B-5)}$$

where z is the transform variable. As

$$\frac{d^k}{dz^k} \hat{g}(z)\,|_{z=1} = \sum_{n=0}^{\infty} n(n-1)\cdots(n-k+1)\, g(n) \quad \text{(B-6)}$$

all the moments of $g(n)$ can be evaluated from (B-6). In particular, the first moment is given by

$$M_1 = \frac{d}{dz} \hat{g}(z)\,|_{z=1} \quad \text{(B-7)}$$

and the second moment is expressed by

$$M_2 - M_1 = \frac{d^2}{dz^2} \hat{g}(z)\,|_{z=1}$$

The variance is evaluated from

$$\sigma^2 = M_2 - M_1{}^2 \quad \text{(B-8)}$$

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