Residence Time Distribution in Screw Extruders

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A companion article (Chen and Hu, 1993a) discussed a statistical theory that for a system consisting of two closed subsystems, the residence time density (RTD) functions of the two subsystems will be statistically independent, if a two-dimensional perfect mixing exists at the subsystem boundary. In this case, the overall RTD function is related to that for individual subsystems through the convolution integral. This theory has been validated experimentally using two die-screw combinations.

Based on this theory, a predictive RTD model for an intermeshing counterrotating twin-screw extruder has been developed. The screw in the longitudinal direction has been treated as C-chambers in series. The overall RTD of leakage flows has been calculated from the RTDs of these individual chambers and then converted into the RTD of the extruder. The predicted RTD has been tested against experimental results with success.

Introduction

The residence time distribution (RTD) in a screw extruder is not only of fundamental interest, but it is also an important parameter for polymer extrusion processes since it is related to the thermal loading and mixing quality of polymer materials inside the extruder. Its importance is certainly much more relevant for extrusion processes where chemical reactions can occur, such as polymerization, functionalization, crosslinking and curing.

The difficulties in analyzing the RTD function for a single- or twin-screw extruder when compared with those for classical chemical reactors result mostly from two inherent problems: (1) complicated geometrical configurations of the flow path and (2) high viscosity and non-Newtonian behavior of polymeric fluids. Consequently, a number of models for the RTD in a screw extruder have been developed during the last two decades. By introducing an eddy diffusivity, which is similar to the molecular diffusivity in the classical axial dispersion model, Todd (1975) developed a theoretical model for the RTD in an intermeshing counterrotating twin-screw extruder. The same method was used recently by Van Zuilichem et al. (1988). Other authors attempted to develop theoretical models using an analysis based upon an assumed flow mechanism. Most of these models only hold for the RTD in a part of the extruder where the flow pattern is relatively simple. For example, Matsubara (1980, 1983) developed models for a T-die and a coat-hanger die, Pinto and Tadmor (1970) developed a model for the RTD for a single-screw extruder, while Kemblowski and Sek (1981) presented a model for mean residence time and variance of the RTD in a single-screw extruder. In other work, Potente and Lappe (1986) studied the effect of different tracers on the accuracy of RTD measurement and proposed a mathematical model for mean and minimum residence time in a single-screw extruder. Kao and Allison (1984) studied effects of some selected process parameters, and the results were explained by flow mechanism in a partly filled channel.

A notable RTD model was developed by Janssen et al. (1979) for an intermeshing counterrotating twin-screw (CRT) extruder. The extruder was treated as individual turns of screw channel (C-chambers) connected in series, along with two important assumptions: (1) the leakage flows in the solid conveying, melting and partly-filled zones are unimportant and (2) the C-chambers in the last fully filled zone are perfect mixers. The

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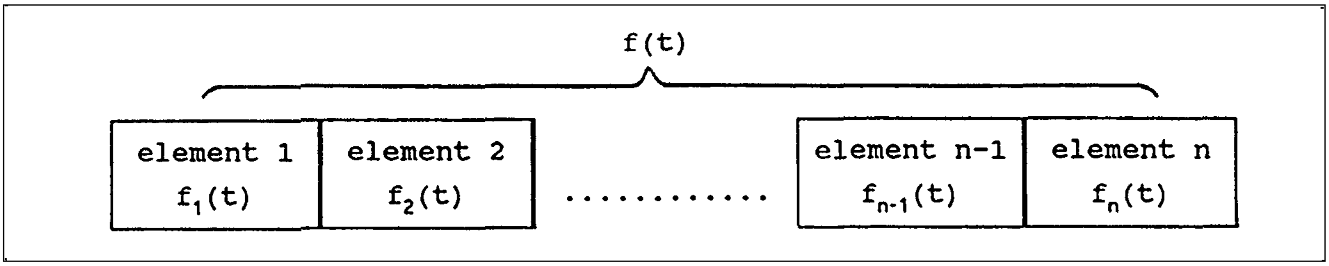

Figure 1. *n* elements in series.

same method was used by Foster et al. (1990) for a nonintermeshing counterrotating twin-screw extruder. However, it is well-known that a single C-chamber is far from a perfect mixer, and the calender leakage flow (leakage flow through the gap between two screws, see Figure 4) is significant even in partly filled screw channels. If these two assumptions are relaxed, a relationship between the overall RTD and those in these connected C-chambers will have to be found. Although much work has been contributed to the RTD in an extruder, very little is known about such a relation (Curry, 1991).

Because the flow mechanism at the boundaries of these elements (for example, the C-chambers) is very complicated, any attempts to describe it by physical models would be unrealistic at the present time. The objectives of this article are threefold: (1) to seek for a theory which relate the overall RTD to those in individual elements without physical models, (2) to develop a predictive model for the RTD in an intermeshing twin-screw extruder based on the theory, and (3) to test the theory and the predictive RTD model against experiments.

Theoretical Background

A fundamental description of polymer flows passing through screw channels, particularly through the boundaries between screw elements in series, is very complicated. Therefore, the development of an RTD model for a twin-screw extruder is also very difficult. This article uses a statistic theory instead of a physical model to relate the overall RTD in an extruder to those in appropriately divided elements. The fundamental basis of the theory is that for a flow passing through a system composed of two elements in series, when fluid particles leaving the first element have the same probability to occupy a given position at the entrance of the second element, the RTDs in these two elements are statistically independent of each other. Then, the overall RTD can be calculated from the individual RTDs using a statistical method.

The RTD analysis for statistically independent network systems was described in sufficient detail by Nauman and Buffham (1983) and is briefly summarized here. For a system consisting of n statistically independent elements connected in series whose residence time density functions are $f_1(t), f_2(t), \ldots,$ and $f_n(t)$, respectively (see Figure 1), the overall $f(t)$ function is related to those of individual elements in the Laplace domain by the following product:

$$\bar{f}(s) = \bar{f}_1(s)\bar{f}_2(s) \ldots \bar{f}_n(s) \quad (1)$$

or in the time domain by n-fold convolution (Chen and Hu, 1993a):

$$f(t) = \int_0^t f_1(\tau_1) \int_0^{t-\tau_1} f_2(\tau_2) \ldots \int_0^{\tau_{n-3}-\tau_{n-2}} f_{n-1}(\tau_{n-1}) \times f_n(\tau_{n-2}-\tau_{n-1}) d\tau_{n-1} \ldots d\tau_2 d\tau_1 \quad (2)$$

For a combination of two elements, Eq. 2 reduces to (Nauman, 1983):

$$f(t) = \int_0^t f_1(\tau) f_2(t-\tau) d\tau \quad (3)$$

Note that the individual $f(t)$ functions in Eqs. 2 and 3 can be interchanged since they are independent of one another.

As discussed previously (Chen and Hu, 1993a), the statistically independent RTDs in elements in series physically implies that perfect mixing exists at the junction or the boundary between any two elements so that fluid particles at this location are totally randomized, and with the limitation that there is no backflow from a downstream subsystem to upstream subsystems.

RTD Model for an Instantaneous CRT Screw Extruder

The theory given by Eq. 1 or 2 was utilized to develop a predictive RTD model for an intermeshing CRT screw extruder.

The screw in an intermeshing CRT screw extruder is treated as consisting of C-chambers connected in series. Every single C-chamber is considered as an element, and the *leakage flows* at the boundaries of each pair of elements are assumed to be sufficiently randomized by squeezing and shearing forces due to the screws rotating in opposite directions. It will be shown below that with these assumptions, the overall RTD for the leakage flows can be determined by the theory presented above using a *moving coordinate*. By an appropriate coordinate transformation, it can then be converted into the RTD of the extruder.

Figure 2 shows the physical model for the C-chambers connected in series. Around a C-chamber are: a stationary screw, the moving barrel and the other rotating screw. If the screw curvature is neglected, the C-chambers can be treated as straight rectangular channels which move forward due to the relative motion between the screw and the barrel (in analyzing the flow in a screw extruder, it is common practice to view the barrel as moving and the screw stationary, although it is the screw that actually moves and the barrel that stays still (Tadmor, 1970)). Figure 2 shows only C-chambers in one thread. If the

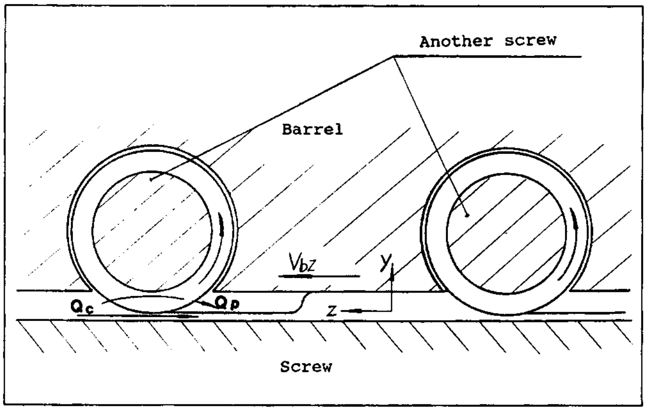

Figure 2. Physical model for the RTD of an intermeshing counterrotating twin screw extruder.

screw contains n threads, there will be n rows of C-chambers which are in parallel. The RTD functions are assumed to be the same for both screws in the CRT screw extruder because of the geometrical symmetry.

Conversion of the RTD of leakage flow into the overall RTD

The overall volumetric flow rate in an intermeshing CRT screw extruder consists of two parts: a plug flow due to the positive displacement character of the intermeshing twin screws, and various leakage flows through different gaps. The overall flow rate is a simple algebraic summation of the volumetric flow rates of these two parts. The overall RTD can also be viewed as some kind of combination of the RTD of the plug flow and that of the leakage flows. However, the latter is not a simple algebraic summation of these two parts since they are related through a coordinate transformation.

Figure 3 illustrates this coordinate transformation using a simple laminar drag flow between two parallel plates as an example. The term "drag flow" is largely used in polymer processing and originates from the fact that the fluid constraint between two parallel plates moves by dragging one of the plates. Figure 3a shows the flow through the plates with a length of L and a residence time density function of $f(t)$. If the lower plate is stationary and the upper plate moves at a

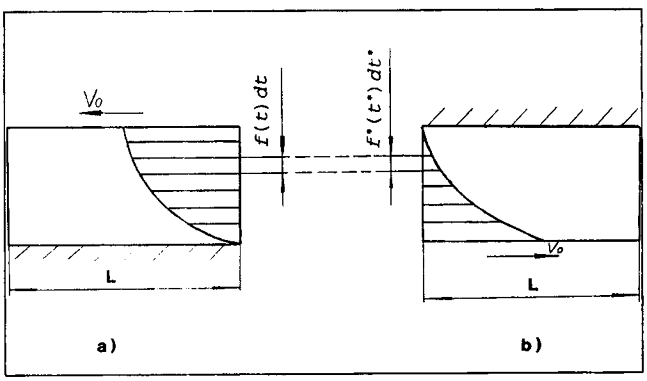

Figure 3. Relationship between the RTD of an actual flow system (a) and that of an imaginary system under a moving coordinate (b).

velocity of v_0, then the minimum residence time t_0 equals to L/v_0 provided that no slip occurs at the plates. If a coordinate is placed on the upper plate moving in the same direction and at the same speed (v_0) as the upper plate, a "backward flow" is observed under this moving coordinate, as shown in Figure 3b. For a small portion of fluid whose residence time is t in the actual flow (Figure 3a) and t^* in the backward flow (Figure 3b), the following expression holds:

$$f(t)dt = -f^*(t^*)dt^* \tag{4}$$

since $f(t)dt$ and $f^*(t^*)dt^*$ actually describe the same portion of fluid. The negative sign in Eq. 4 results from the fact that while t increases, t^* decreases. The velocity of the small portion of fluid observed in the backward flow under the moving coordinate v^* can be related to that in the actual flow velocity v using:

$$v^* = v_0 - v = \frac{L}{t_0} - \frac{L}{t} \tag{5}$$

The residence time observed in the backward flow t^* is:

$$t^* = \frac{L}{v^*} = \frac{t_0 t}{t - t_0} \tag{6}$$

By substituting Eq. 6 into Eq. 4, the relation between the RTD of the actual flow and that of the backward flow is obtained:

$$f(t)dt = -f^*\left(\frac{t_0 t}{t-t_0}\right) d\left(\frac{t_0 t}{t-t_0}\right) \tag{7}$$

Equation 7 allows the conversion of the RTD of the backward flow observed in the moving coordinate into that of the actual flow. Note that the moving coordinate acts like plug flow with a velocity v_0. The actual flow in Figure 3a can be considered as a combination of a plug flow and a backward flow shown in Figure 3b, and so can be the flow in a CRT screw extruder.

Although Eq. 7 is derived from a simple laminar drag flow between two parallel plates, it holds for any laminar flow in any geometry (cf., Appendix 1), provided that the velocity of the moving coordinate is equal to or larger than the maximum velocity of the fluid. Therefore, the RTD of the CRT screw extruder can be determined by converting the RTD of the leakage flow using Eq. 7. The latter can be calculated under a moving coordinate which is placed on the barrel with a velocity equaling to the barrel velocity in the screw channel direction.

Determination of the RTD of the leakage flow

Leakage flows determine how a RTD deviates from plug flow. Generally there are three types of leakage flows in an intermeshing CRT screw extruder (cf., Figure 4): calendar leakage flow Q_c, pressure leakage flow Q_p and flight leakage flow Q_f. The calender leakage flow is the flow through the gap between two screws; the pressure leakage flow is the flow through the tetrahedron gap between the flanks of screw channels, and the flight leakage flow is the flow through the gap

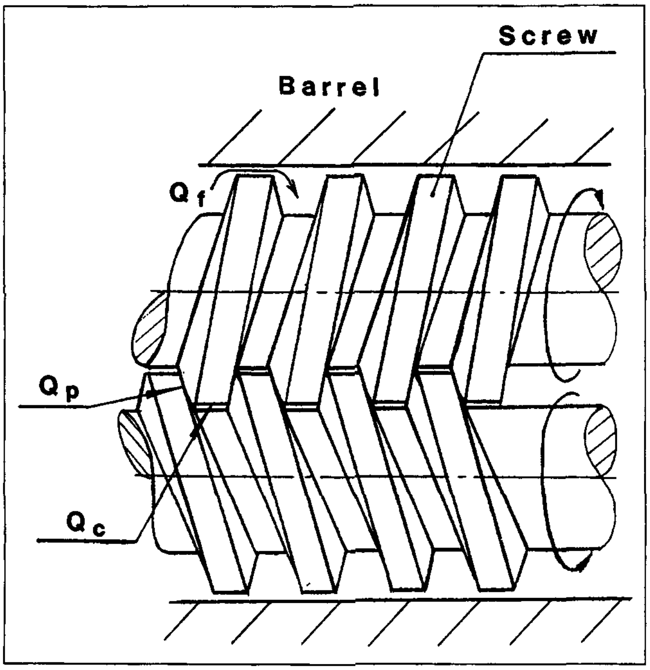

Figure 4. Leakage flows in a CRT screw extruder.

between the screw flight and the barrel. Among the three leakage flows, the calender leakage flow is usually the most important one, and the flight leakage flow is the least important one in a typical extrusion process of polymers (Janssen, 1978). In the experiments discussed in the next section, the flight leakage flow was estimated to be well below 10% of the overall leakage flow, thus neglected in the RTD calculation.

All these leakage flows in an intermeshing CRT screw extruder were quantitatively discussed by Janssen (1978) and Janssen et al. (1976). Assuming that a polymer melt in the extruder is a Newtonian fluid, analytical solutions for these leakage flows were derived. For the calender leakage flow,

$$Q_c = \frac{4(p_z - Bn_z)}{3n_z}\left[\pi N(D-H)h_0 + \frac{\Delta p h_0^3}{6\eta_c\sqrt{(D-H)h_0}}\right] \tag{8}$$

while for the pressure leakage flow,

$$Q_p = \frac{0.0026}{\eta_t}\left(\frac{H}{D}\right)^{1.8}\Delta p D^3 \beta^2 \tag{9}$$

and finally for the flight leakage flow,

$$Q_f = \left(\frac{1}{2}v_n\delta + \frac{\Delta p \delta^3}{12\eta_f(p_z/2 - H\tan\beta)}\right)(\pi - \alpha/2)D \tag{10}$$

The Newtonian fluid assumption is justified by the argument that the shear rate in the intermeshing area of a counterrotating twin screw extruder is relatively low so that the error introduced by this assumption is expected to be small. This is particularly so in the present study, because the screw speed is low (10 rpm).

The overall leakage flow in a single C-chamber observed under the moving coordinate was taken as a flow in a rectangular channel of a single-screw extruder. Clearly, this is a crude approximation, because the channel width-depth ratio in a C-chamber is not as large as in a typical single-screw extruder. A more accurate analysis of the problem is complicated and is beyond the scope of this study. The approximation allows direct calculation of the RTD of the overall leakage flow in a C-chamber using the following analytical expression established by Pinto and Tadmor (1970):

$$f(t)dt = \frac{3\zeta[1-\zeta+(1+2\zeta-3\zeta^2)^{1/2}]}{(1+2\zeta-3\zeta^2)^{1/2}}\,d\zeta \tag{11}$$

where ζ is a dimensionless coordinate; $\zeta = y/H$ ($\zeta < 2/3$) and y is the coordinate in the channel depth direction. The time t in Eq. 11 is given by:

$$t = \frac{L_c}{3v_{bz}\left(1+\dfrac{Q_p'}{Q_d}\right)\sin\theta\cos\theta}\;\frac{3\zeta - 1 + 3(1+2\zeta-3\zeta^2)^{1/2}}{\zeta[1-\zeta+(1+2\zeta-3\zeta^2)^{1/2}]} \tag{12}$$

where L_c is the axial length of fillage in a single C-chamber, V_{bz} is the barrel velocity in z direction (along the channel), θ is the helix angle, and Q_p' and Q_d are the pressure and drag flow rates, respectively. Q_d can be calculated using the expression shown by Tadmor (1970) in a rectangular channel:

$$Q_d = \frac{v_{bz}WH}{2}F_d \tag{13}$$

where W is the width of the screw channel, and F_d is the shape factor given by:

$$F_d = \frac{16W}{\pi^3 H}\sum_{i=1,3,5}^{\infty}\frac{1}{i^3}\tanh\left(\frac{i\pi H}{2W}\right) \tag{14}$$

In fact, the total leakage flow Q_l is the overall flow rate observed under the moving coordinate, that is,

$$Q_l = Q_d + Q_p' \tag{15}$$

If the flight leakage flow is negligible compared with the calender leakage flow, then $Q_l = Q_c + Q_p$. Finally, Q_p' is obtained using:

$$Q_p' = Q_c + Q_p - Q_d \tag{16}$$

where Q_c and Q_p are given by Eqs. 8 and 9, respectively.

The fillage length in a single C-chamber L_c in Eq. 12 is given by:

$$\frac{L_c}{\sin\theta} = \frac{V_m}{WH} \tag{17}$$

where V_m is the volume of the material in the C-chamber, which can be determined through a mass balance:

$$2n_z\left(V_m\frac{N}{60} - Q_l\right) = Q \tag{18}$$

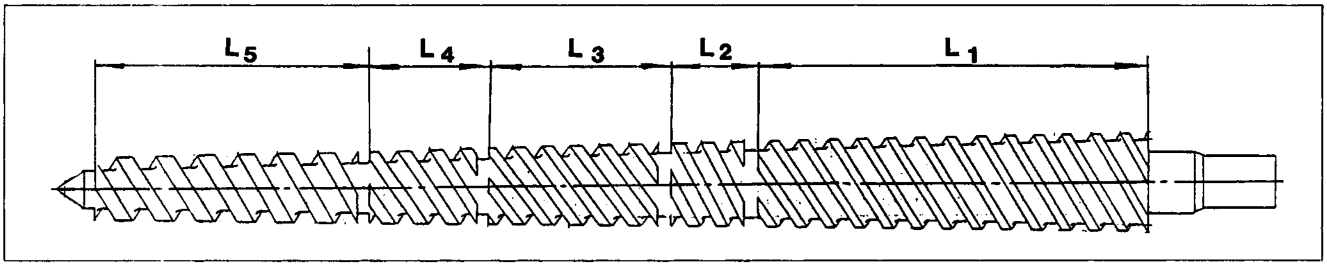

Figure 5. Screw geometry of the twin screw extruder.

where Q is the volumetric throughput rate in M^3/s, and the coefficient "2" takes account of the twin screws.

In the fully filled zone, L_c equals to $L_c = p_z - (\alpha/2\pi)p_z$, which is the screw pitch corrected for the twin-screw overlap. The length of fully filled zone is determined by the die pressure, the throughput Q and Eqs. 8-10.

Finally, the RTD of the leakage flow in a single C-chamber can be calculated by Eqs. 11 and 12. Assuming that the leakage flow is sufficiently randomized when it passes through the calender gaps and the tetrahedron gaps, the overall RTD of the total leakage flow can be determined by those of the leakage flow in the individual C-chambers using Eq. 2. It can then be converted into the RTD in the extruder by Eq. 7.

Experiment

The validity of the statistical theory presented above was experimentally examined using two combinations: (1) a die and a single screw and (2) a die and a twin screw. A tracer technique was adopted to measure the RTDs. A suspension polymerized polystyrene (PS) was chosen as the polymer fluid and 1-aminoanthraquinone was chosen as the tracer. The use of this dye was motivated by the following advantages that it possesses over other possible candidates: (1) it can be finely dispersed in a polystyrene matrix, which is important for accurate measurements of the RTD, (2) it is chemically and thermally stable, and (3) it is highly sensitive to ultraviolet light with a maximum absorption wavelength of 462 nm which is far away from the maximum absorption wavelength of an aromatic substance of a single benzene ring, such as PS and toluene (around 265 nm), thus making the tracer concentration measurement accurate. It was found that the absorption obeys the Beer-Lambert law well at a low concentration ($<0.01\%$). At the working temperature (190°C) of the experiments, the zero shear rate viscosity of the polystyrene used was 4,500 Pa·s. The nominal shear rate was estimated to be around 10 s^{-1}, and the corresponding non-Newtonian index was about 0.6.

A single-screw extruder (Haake rheomex 254; screw diameter D = 19 mm; length-diameter ratio $L_s/D = 25$) and a laboratory conical intermeshing counterrotating twin-screw extruder (Haake rheomex TW-100; tip-screw diameter D = 19.7 mm; screw length $L_s = 331.78$ mm) were chosen as the extruders. The RTD results obtained in the twin-screw extruder were further used to test the RTD model developed in this study (cf., Figure 5 and Table 1 for the screw configurations). The die pressure was recorded by a Dynisco pressure transducer.

A small die (Figure 6a) was used as an adaptor to measure the RTD of the screw (the volume of its channel is very small so that the residence time in it is negligible). A large die (Figure 6b) having a long cylindrical channel was used to measure the RTD in the combinations of the die and the screws. No die plate (breaker plate) was placed between the screw and the die.

Table 1. Screw Geometries of the Twin Screws

Zones	L_1	L_2	L_3	L_4	L_5
Zone Length (mm)	123	27	57	37	87
Initial Channel Depth (mm)	2.0	2.9	3.05	3.25	3.45
Channel Width (mm)	6.0	5.0	5.0	5.0	6.5
Flight Width (mm)	2.5	3.5	3.0	4.0	5.5
Pitch (mm)	36.8	26.7	36.8	29.1	25.5
No. of Thread	4	3	4	3	2

A mixture of the polystyrene and the tracer (75/25 by weight) was blended using a Banbury mixer at 160°C. This concentrate was then pelletized into small particles having similar sizes of the polystyrene. The concentrate pellets (0.5 g) were injected into the screw through the hopper as a pulse after the extrusion process was steady. Samples were collected at the die at chosen time intervals, cut in the direction perpendicular to the flow direction. Tracer concentrations in the samples were determined by UV analysis. The reproducibility was examined by repeated experiments, and it was found to be very good.

The normalized RTD function was calculated on the basis of the following expression:

$$f(t_i) = \frac{C_i}{\int_0^\infty C(t)dt} \tag{19}$$

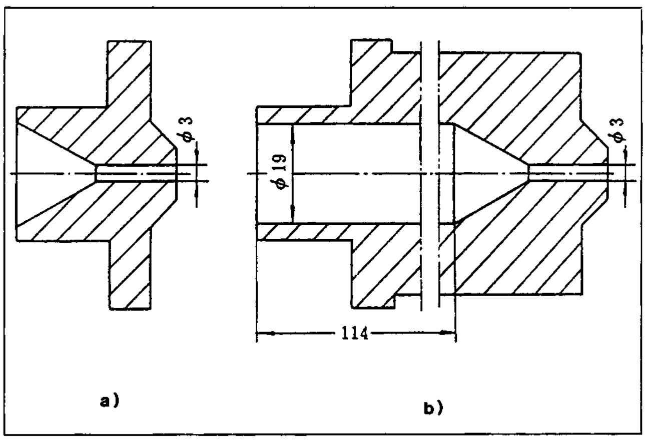

Figure 6. Dies used for RTD measurements.

(a) die for the RTD of the screw; (b) die for the RTD of the combination of the screw and the die.

where C_i is the tracer concentration in the sample collected at time t_i ($i = 1, 2, \ldots, n$), and $C(t)$ is the instantaneous tracer concentration. The truncation errors in the integral for our measurements were found to be 0.4-1.1% using an empirical expression suggested by Todd (1975):

$$R' \doteq \int_{t_n}^{\infty} Ae^{-kt}dt = \frac{C_n}{k} \tag{20}$$

where R' is the amount of the tracer remained in the extruder after the last sample is taken, and k is a coefficient determined by regression.

Results and Discussion

Experimental verification of the theory

The validity of the theory was experimentally examined using two combinations: a die and a single screw, and a die and a twin screw. In both cases, the junction between the screw and the die was first chosen as the boundary. This is assuming that the flow is sufficiently randomized when it passes across the boundary so that Eq. 3 can be used to determine the overall RTD from those of the screw and the die. Denote $\phi(t)$ as the RTD of the screw which was determined experimentally with the small die, and $\psi(t)$ as that in the large die which can be calculated by the RTD model for a fully developed tubular laminar flow of a Newtonian fluid:

$$\psi(t) = \begin{cases} 0 & t < t_0 \\ \dfrac{2t_0^2}{t^3} & t \geq t_0 \end{cases} \tag{21}$$

where t_0 is the minimum residence time in the die. It can be calculated by the following expression:

$$t_0 = \frac{\pi R^2 L_d}{2Q} \tag{22}$$

where L_d and R are the length and diameter of the cylindrical die. The Newtonian melt assumption for the polystyrene melt is supported by very low shear rates on the die wall ($\dot{\gamma}_w < 0.1$ s^{-1}), and the fully developed assumption is based on the fact that the developing length is less than 10% of the total channel length (Middleman, 1977).

The experimental RTD results and those predicted by Eq. 3 for both combinations are shown in Figures 7a and 7b, respectively. The predicted values are in good agreement with the experimental ones for the combination of the large die and the single screw (cf., Figure 7a), but there is a little disagreement between them for the combination of the large die and the twin screw (Figure 7b). This disagreement is expected since there is a long transition zone (about 45 mm long, that is, 2-3 times of the tip-screw diameter) between the twin screw and the die, as shown in Figure 8a. The boundary which was chosen to separate the RTD in the twin screws from that in the die is very far from the screw tips, and the flow at this boundary is not sufficiently randomized to apply Eq. 3. In fact, the measured $\phi(t)$ includes both the RTDs in the twin screws and the transition zone. The location at which the flow is most randomized should be where the twin-screw tips are located. If the latter is chosen as the boundary, the RTD in the transition zone, $\psi^*(t)$, should be subtracted from $\phi(t)$ and added to the RTD in the die $\psi(t)$ (Figure 8b). Upon considering the transition zone and the real twin screws as a combination of two elements and the twin screw tips as the boundary, the overall RTD in this combination $\phi(t)$ can be related to the individual RTDs in the transition zone, $\psi^*(t)$, and in the real screws, $\phi^*(t)$, by applying Eq. 3:

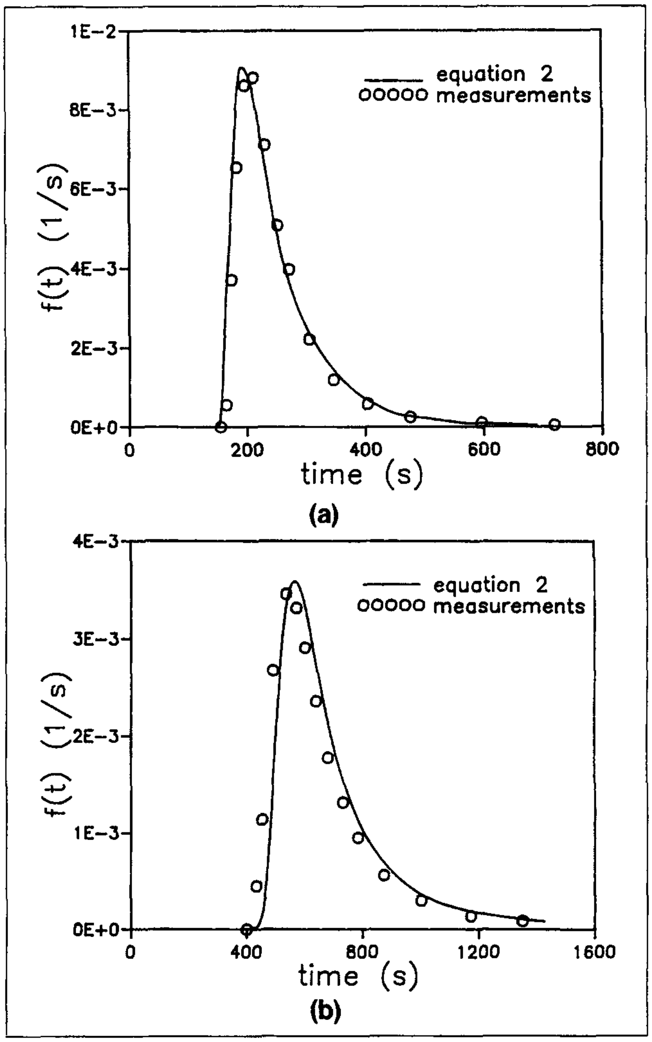

Figure 7. Comparison between the RTD predicted by Eq. 3 and experimental results.

(a) single screw and die combination; (b) twin screw and die combination.

$$\phi(t) = \int_0^t \phi^*(\tau)\psi^*(t-\tau)d\tau \tag{23}$$

In this equation, $\phi(t)$ was experimentally measured with the small die, and $\psi^*(t)$ can be calculated approximately by the tubular laminar flow model of a Newtonian fluid (Eqs. 21 and 22). Thus $\phi^*(t)$ can be obtained by solving Eq. 23 through deconvolution. Equation 23 is the so-called first kind Volterra equation. An accurate solution to this kind of equation is

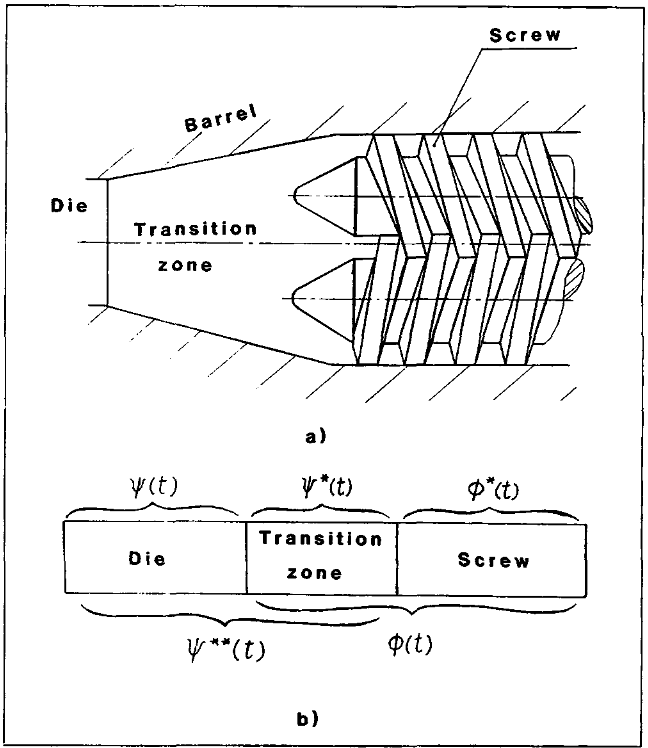

Figure 8. Division of elements between the twin screws and the die.

(a) transition zone between the twin screws and the die; (b) redivision of the elements for the RTD calculation.

the scope of this work. The deconvolution was done in this study using both a linear algebra approach and one based on optimization, both yielding similar results. The RTD function in the entire extruder $f(t)$ can then be related to the RTD of the real screw, $\phi^*(t)$, and that of the combination of the transition zone and the die $\psi^{**}(t)$ by:

$$f(t) = \int_0^t \phi^*(\tau)\psi^{**}(t-\tau)d\tau \tag{24}$$

where the RTD function $\psi^{**}(t)$ can also be calculated by the tubular laminar flow model of Newtonian fluid in the whole channel of the transition zone and the die. As expected, the predicted results by Eq. 24 are in better agreement with the experimental ones (Figure 9), further supporting the validity of the theory (Eq. 2 or 3).

One may argue that the errors seen in Figure 7b between the experiment and Eq. 3 may be introduced by the various assumptions or approximations involved, such as treating the polystyrene melt as a Newtonian fluid and assuming a fully developed flow in the die. Thus, the slight improvement in Figure 9 compared with Figure 7b may not be well-based. It is difficult to assess, based upon current knowledge, and the effect of these assumptions on the error. A key point here is that mixing at the junction between the twin screws and the large die is not as intense as at the twin-screw tips, so that choosing the latter as the boundary is preferred.

In separate work (Chen et al., 1993b), the validity of Eqs. 2 and 3 were further confirmed using nonintermeshing counterrotating twin-screw zone combinations. A polystyrene solution in ethylbenzene (30 wt.% polystyrene) was used as the fluid. Compared with the polystyrene melt, the polystyrene solution behaves more similarly to a Newtonian fluid. Also, the experimental conditions needed for accurate RTD measurements can be more easily satisfied.

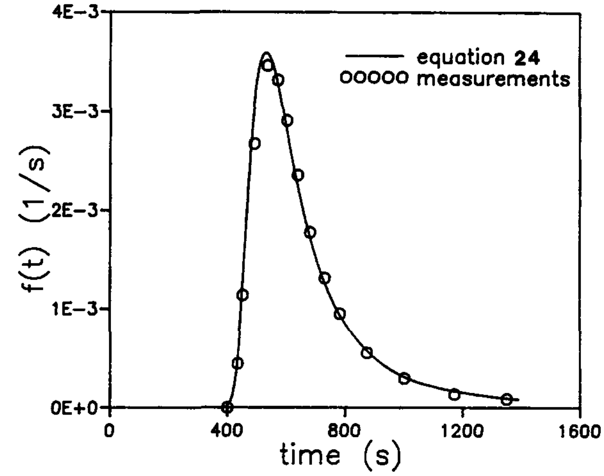

Figure 9. Comparison between the RTD predicted by Eq. 24 and that experimentally obtained for the combination of the twin screw and the die.

Selected characteristic values of the RTD between the theory and the experimental results are compared in Table 2. In this table, the mean residence time $\bar{t}$, the variance σ^2, and the third moment M_3 are defined as:

$$\bar{t} = \int_0^\infty t f(t)dt \tag{25}$$

$$\sigma^2 = \int_0^\infty (t-\bar{t})^2 f(t)dt \tag{26}$$

$$M_3 = \int_0^\infty (t-\bar{t})^3 f(t)dt \tag{27}$$

Table 2. Comparison of Some Characteristic Values of RTD between the RTD Predictive Model and the Experiment

Type of Extruder	Characteristic Values of RTD	Theory	Experiment
Single Screw $Q=0.281$ g/s $N=30$ r/min	Mean Residence Time, s	255.6	258.6
	Variance s^2	6,512	6,376
	Third Moment s^3	1.35×10^6	1.13×10^6
Twin-Screw $Q=0.104$ g/s $N=10$ r/min	Mean Residence Time, s	656.4	659.8
	Variance s^2	2.9×10^4	3.4×10^4
(Calcul. from Eq. 24)	Third Moment s^3	7.3×10^5	1.18×10^6

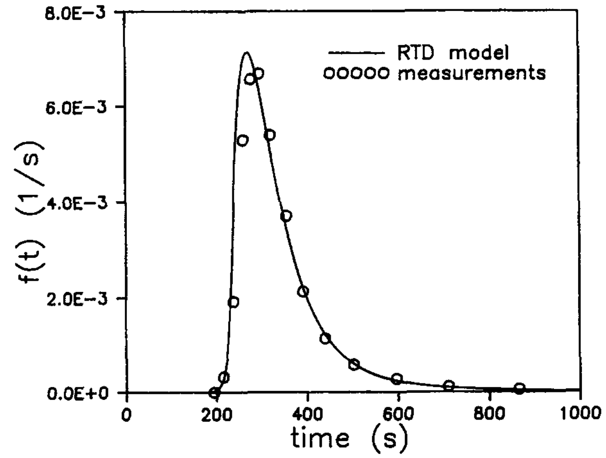

Figure 10. Comparison between the RTD predicted by the model and the experimental results.

Mean residence time and moments: predicted: $\bar{t}=337.9$ s, $\sigma^2=1.33\times10^4$, $M^3=4.30\times10^6$; experimental: $\bar{t}=336.6$ s, $\sigma^2=1.14\times10^4$, $M^3=3.38\times10^6$.

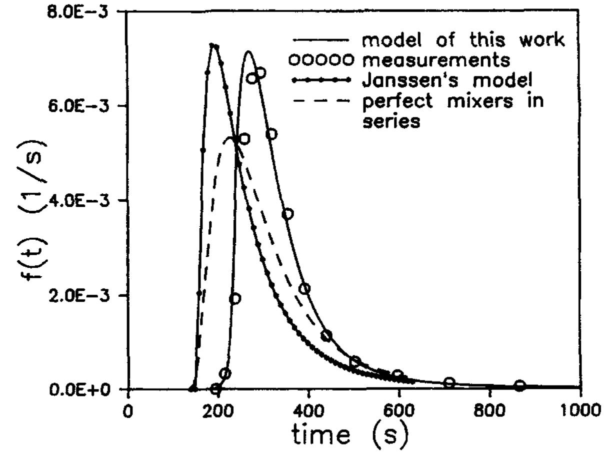

Figure 11. Comparison between Janssen's model and the experimental results.

Examination of the Predictive RTD Model

The polystyrene pellets, together with the tracer concentrate, undergo three transport zones in which phase transformation occurs progressively, as in a typical plasticating extrusion operation. These zones include a solids conveying zone, a melting zone, and a partly filled and fully filled melt conveying zones. In our RTD model construction, two simplifications were made: (1) the flow in the first zone (solids conveying zone) was plug flow, and (2) the mixture consisting of polymer solids and melt in the melting zone was treated as a single liquid. The second assumption is justified by the fact that the solids in an intermeshing counterrotating twin-screw extruder are squeezed into small particles when passing through the intermeshing areas and are commingled with the melt. This results in pseudo-homogeneous liquid behavior. This situation is in contrast to the melting mechanism in a single-screw extruder where solids bed is formed separating from the melt liquid.

The RTD model developed for an intermeshing CRT screw extruder was tested against the experimental results generated from the twin-screw extruder used in this study. As shown in Figure 10, the predicted results are in satisfactory agreement with the experiments. Such an agreement is particularly encouraging, considering the complexity of the flow mechanisms in an intermeshing CRT screw extruder and the assumptions involved.

Janssen's model (Janssen, 1979) was used to test against the experimental results to show how a flow passing through the channels of a twin-screw extruder deviates from the plug-flow or perfect-mixing-flow model or a combination of both. In comparison to the experimental results, the peak of the prediction using Janssen's model appears noticeably sooner (Figure 11). The disagreement between the experimental results and Janssen's model is probably caused by two model assumptions: the flow in the first three zones is plug flow (no leakage flow), and every single C-chamber in the last zone is a perfect mixer. Obviously, the plug-flow assumption underestimates the residence time, since the leakage flow which is ignored by this assumption tends to reduce the average flow velocity (or to increase the holdup) and therefore increases the residence time. The perfect mixer assumption for the last zone also underestimates the residence time. This is illustrated by the dashed curve in Figure 11, which is obtained by assuming that all the four zones of the extruder are perfect mixers so that the length of the plug-flow zone is zero. Thus, it is clear that the plug- and perfect-mixing-flow assumptions implicitly underestimate the RTD of the flow in an intermeshing CRT screw extruder.

It should be pointed out that the success of this model is mainly attributable to the incorporation of the statistical theory developed in the present study. By introducing Eq. 2, the perfect mixer and the plug-flow assumptions can be relaxed. Also note that the validity of this theory only requires a perfect two-dimensional mixing at chosen boundaries for division of elements. This is particularly applicable for screw extruders in which the flow channels are usually very long and narrow. By virtue of such flow channels, a two-dimensional perfect mixing can be readily achieved at the locations of high shear, elongational or squeezing mixing elements (kneading discs, calender gaps, and so on) so that fluid particles at these locations can be distributed randomly across the entire sections to which Eq. 2 can be applied. By contrast, a three-dimensional perfect mixing is much more difficult to achieve in screw extruders just because of the long and narrow nature of the screw channels.

Conclusions

In this article, a statistical theory which relates the overall residence time distribution (RTD) in a screw extruder to those in appropriately divided elements has been presented. The core of the theory is that if the flow at the boundary between two elements is sufficiently randomized so that any fluid particles leaving the first element have the same probability to occupy a given position at the entrance of the second element, the RTD functions of these two adjacent elements are independent of each other. Hence, the overall RTD can be related to the individual ones by a statistical method described by Eqs. 1-3. This theory has been validated experimentally using two types of combinations: (1) a die and a single screw and (2) a die and an intermeshing twin screw.

This theory is particularly useful for screw extruders of which the flow channels are usually very long and narrow. In such channels, a flow can be sufficiently randomized at the locations of mixing elements (kneading discs, calender gaps) to the extent that the theory can be applied.

This theory has been used for the development of a RTD model for an intermeshing counterrotating twin-screw extruder. The screw in the longitudinal direction has been treated as C-chambers in series. The overall RTD of leakage flows has been calculated from the RTDs of these individual chambers, and it has been finally converted into the RTD of the extruder. By introducing this theory, the perfect-mixer- and plug-flow assumptions which are far from reality can be relaxed. The model predictions have been tested against the experimental results successfully.

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Notation

A = coefficient in Eq. 20
B = thread distance, m
$C(t)$ = tracer concentration distribution, kg/m^3
C_i = tracer concentration of sample i, kg/m^3
D = diameter of the screw, m
$\bar{f}(s)$ = Laplace transform of $f(t)$
$f(t)$ = residence time density function, s^{-1}
$f^*(t)$ = $f(t)$ function of an imaginary system, s^{-1}
$f_i(t)$ = residence time density function of element i, s^{-1}
F_d = shape factor defined by Eq. 14
$F(t)$ = cumulative RTD function
$F^*(t)$ = cumulative RTD function of an imaginary system
h_0 = clearance of the calender gap, m
H = depth of the screw channel, m
k = coefficient in Eq. 20
L = length of the plates, m
L_c = length of fillage of a single C-chamber, m
L_d = length of the channel of the die, m
L_i = length of screw in zone i, m
L_s = length of the screw, m
M_3 = third moment of RTD function, s^3
n_c = number of C-chamber
n_z = number of threads
N = screw speed, r/min
p_z = pitch, m
Q = volumetric throughput, m^3/min
Q_c = volumetric calender leakage flow rate, m^3/s
Q_d = volumetric drag flow rate, m^3/s
Q_f = volumetric flight leakage flow rate, m^3/s
Q_k = volumetric leakage flow from one screw to the other, m^3/s
Q_l = total volumetric leakage flow rate, m^3/s
Q_p = volumetric pressure leakage flow rate, m^3/s
Q_p' = volumetric pressure flow rate, m^3/s
Q_s = volumetric leakage flow between two adjacent C-chambers of the same screw, m^3/s
R = radius of the die, m
R' = amount of tracer remained in the extruder after time t_n, kg
t = time, s
$\bar{t}$ = mean residence time, s
t_0 = minimum residence time, s
v = velocity of the fluid in the plates, m/s
v^* = velocity of the fluid observed in the moving coordinate, m/s
v_{bz} = velocity of barrel in z direction, m/s
v_n = velocity of the barrel in the axial direction, m/s
v_0 = velocity of the moving plate, m/s
V_m = volume of material in a single C-chamber, m^3
W = width of the screw channel, m
y = coordinate in the screw channel depth direction, m

Greek letters

α = screw overlap angle
β = flight wall angle
γ_w = shear rate at the wall of a die, s^{-1}
δ = clearance between the barrel and the screw flight, m
Δp = pressure difference between two adjacent C-chambers, Pa
ζ = y/H, dimensionless coordinate
η_c = viscosity of the melt in the gap between the screws, Pa/s
η_f = viscosity of the melt in the flight gap, Pa/s
η_t = viscosity of the melt in the tetrahedron gap, Pa/s
θ = helix angle of thread
σ^2 = variance of RTD function, s^2
τ = dummy integral variable of time, s
$\phi(t)$ = RTD function of the screw, s^{-1}
$\phi^*(t)$ = corrected RTD function of the screw, s^{-1}
$\psi(t)$ = RTD function of the die, s^{-1}
$\psi^*(t)$ = RTD function of the transition zone located between the twin screws and the die, s^{-1}
$\psi^{**}(t)$ = RTD function of the die including the transition zone, s^{-1}

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Appendix I: Derivation of Eq. 7 for a Laminar Flow in a Generalized System

Consider a closed laminar flow system of any geometrical configuration with a given cross section (Figure A1) and a contour of L. We set two coordinates: one is stationary with regard to the earth (Figure A1), and the other moves in the flow direction at a velocity $V = L/t_0$ (Figure A2; t_0 is so chosen that V is equal to or larger than L/t_{min} (t_{min} is the minimum residence time observed in the stationary coordinate). Obviously, the inlet of the system in the stationary coordinate becomes the outlet in the moving coordinate, and vice versa. Suppose at time $t = 0$, a given amount of tracer is introduced as a pulse at the inlet of the system observed in the stationary coordinate. The portion of the tracer that has left the system observed in this coordinate is $F(t)$. ($F(t)$ is the cumulative residence time distribution observed in the stationary coordinate.) Similarly, if a given amount of tracer is introduced in the inlet of the system observed in the moving coordinate, the portion of the tracer that has left the system is $F^*(t)$. ($F^*(t)$ is the cumulative residence time distribution in the system observed in the moving coordinate.)

The question now is how to relate $F(t)$ to $F^*(t)$. Since the flow mechanism is the same in both coordinates, a fluid particle, having a residence time t in the system observed in the stationary coordinate (its average velocity, v, is then equal to L/t in flow direction), has a residence time in the system observed in the moving coordinate t^*. Its average velocity observed in the moving coordinate $v^* = L/t_0 - L/t = L(t - t_0)/t_0 t$. Time t^*, by definition, is equal to $L/v^* = t_0 t/(t - t_0)$. Notice that the portion of tracer particles that have left the system at time t observed in the stationary coordinate corresponds to the portion of tracer particles that still remain at time t^* in the system observed in the moving coordinate, namely,

$$F(t) = 1 - F^*(t^*) = 1 - F^*\left(\frac{t_0 t}{t - t_0}\right) \tag{A1}$$

Equation A1 can be expressed as:

$$\int_{t_0}^{t} f(t)dt = \int_{\frac{t_0 t}{t - t_0}}^{\infty} f^*\left(\frac{t_0 t}{t - t_0}\right) d\left(\frac{t_0 t}{t - t_0}\right) \tag{A2}$$

Since the left side of Eq. A2 is equal to the right side for any t, the following relation holds:

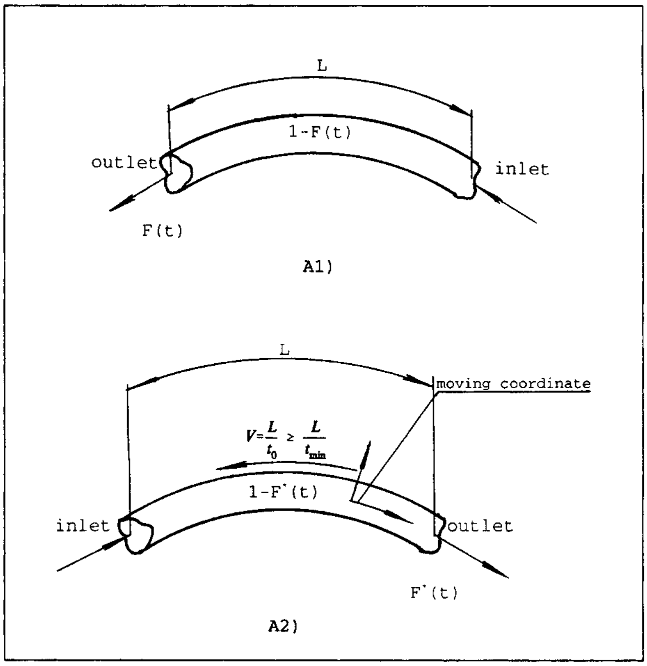

Figure A1. Relationship of the RTD of a laminar flow in a closed system observed in the stationary and moving coordinates.

(A1) system in the stationary coordinate; (A2) system in the moving coordinate.

$$f(t)dt = -f^*\left(\frac{t_0 t}{t - t_0}\right) d\left(\frac{t_0 t}{t - t_0}\right) \tag{A3}$$

Note that this equation is the same as Eq. 7.

The only limitation for Eq. 7 or Eq. A3 is that the velocity of the moving coordinate must be larger than or equal to that of the maximum velocity of the fluid. Otherwise a negative infinite residence time is observed under the moving coordinate and the coordinate transformation loses its sense. Hence Eq. 7 cannot be used for a perfect stirred tank reactor in which the maximum velocity is infinite. Also note that the derivation above is not vigorous.

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