# Nicholas P. Cheremisinoff

INSTRUVENTATION FOR

# Complex Find Flows

## INSTRUMENTATION FOR

# Complex Fluid Flows

Nicholas P. Cheremisinoff



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"Learning, experimenting, observing, try not to stay on the surface of facts. Do not become the archivists of facts. Try to penetrate to the secret of their occurrence, persistently search for the laws which govern them."

IVAN PAVLOV

### **Foreword**

Today's rapidly changing technologies demand a thorough understanding of the principles of fluid mechanics and a knowledge of how to apply them. Aeronautical, biomedical, chemical, civil, marine, and mechanical engineers, as well as meteorologists and physical oceanographers, encounter a multitude of complex flow phenomena. These complex flows are often comprised of two or more phases in which the interactions between them plays the dominant role in controlling transport processes such as heat and mass exchange and reaction kinetics. Observation through quantitative experimentation is the best approach to unravelling the physics of complex flow behavior and in obtaining invaluable scale-up information for industrial applications.

This volume serves as an overview of laboratory methods aimed at quantitative studies of flow behavior. The book describes the basis for different experimental methods aimed at obtaining information on turbulence structure and intensity, flow regime detection, phase interactions, and holdup, entrainment and deposition, and interfacial phenomena in two-phase flows. Typical experimental set-ups are described along with the advantages/disadvantages and precision of various instruments and methods. Discussions of data interpretation and regression are also covered along with guidelines and recommendations for laboratory automation of advanced experimental methods. Where possible, direction is given for adapting certain techniques to commercial units for trouble-shooting and control. Although the book is primarily written with the graduate research student in mind, the more experienced researcher as well as practitioners will find the contents of value.

Gratitude is extended to Technomic Publishing Co. for the publication of this volume.

NICHOLAS P. CHEREMISINOFF

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### **Biography**

Nicholas P. Cheremisinoff heads the Product Development Group of the Elastomers Technology Division of Exxon Chemical Company, Linden, New Jersey. He is involved in research and engineering on multiphase reactor design, complex flow studies, instrumentation development, products research and development of polymeric materials for the consumers market. He is the author/co-author of over twenty books and numerous papers on the subject of fluid mechanics. Dr. Cheremisinoff received his B.S., M.S. and Ph.D. degrees in Chemical Engineering from Clarkson College of Technology. He is a member of a number of professional and honor societies, including A.I.Ch.E., Tau Beta Pi and Sigma Xi.

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### RESIDENCE TIME DISTRIBUTIONS

INDUSTRIAL APPARATUS OFTEN handle several fluids and/or materials which display fluid-like properties at process conditions. The design, scaleup and operation of chemical reactors and various process equipment and transfer lines requires knowledge of the nature of the flow structure and the interaction between various phases. Transitions in flow regimes or flow structure caused by contacting different materials or due to phase changes and/or phase reactions result in different modes of operation of a piece of equipment. The interaction and overall mixedness between phases is largely responsible for establishing the magnitude of exchange coefficients for mass and heat transport properties. This volume provides an overview of instrumentation and methods for studying and quantifying the structure of complex mixture flows. Many of the methods described were developed for studies under well-controlled test conditions. To the industrial researcher, this is a luxury which is not often encountered; however, with a little ingenuity, many of the systems can be adapted to commercial reactors and in semi-works operations. In addition, some of these methods are well suited as troubleshooting techniques for understanding reactor malfunctions/runaways, and in actually controlling process stability.

This first chapter reviews some basic definitions and concepts on the phenomena of mixing in continuous flow systems. These principles help to outline the general parameters which are of interest in understanding flow dynamics and their relation to reactor and process performance. The concepts of mean residence time and residence time distributions provide a first pass understanding of macro-mixing behavior, from whence later chapters will be aimed at experimental methods for quantifying mixing on the micro-scale. An example of studying the response of a reactor through the use of inert tracer techniques is given in this chapter. This will help to illustrate the usefulness of the residence time distribution concept, but also give the newcomer a flavor for planning and executing experimental programs.

### GENERAL DEFINITIONS AND PROPERTIES OF RTD

To begin, consider any continuous flow system to be comprised of one or more entrances and exits; and assume that no reaction takes place and that the system is at steady state. Further, we define the term particle to denote any conserved entity such as a molecule, Brownian particle or fluid element. Particles enter the system, remain in it for some period of time which may be either deterministic or probabilistic, and eventually exit. We permit the situation where a particle makes a number of temporary exits with subsequent re-entrances, but it is required that each particle has some original, first entrance and an ultimate, final exit. Particles have zero age when they first enter and acquire age at a rate equal to time spent within the system boundaries. Aging stops during any temporary exit but resumes at the previous value when the particle re-enters. The age of a particle at its last exit from the system is referred to as the *residence time*, t.

In systems with several types of conserved entities, attention will be restricted to some agreed upon class of entities and the flow rate through the system will refer to the flow rate for that class.

The mathematical definition of residence time takes the form of the cumulative distribution function, F(t), where F(t) = Probability that a particle had a residence time less than t.

When observations are made for a large number of particles, F(t) defines the fraction of the particles or the fraction of the particle flow rate which had residence times less than t. Since residence times cannot be negative, F(t) is defined over  $(0,\infty)$ ; and, due to the probability interpretation, F(t) must be nondecreasing over that interval. The range of F(t) is 0 to 1 with F(0-)=0 and  $F(\infty)=1$ . From a physical standpoint it would seem that F(t) is continuous, however, by discretizing F(t) we allow for isolated points of discontinuity where F(t) undergoes a step change. One such point of discontinuity may be at the origin with F(0+)=b. In this case a fraction of the fluid stream bypasses the system and experiences zero residence time. The situation  $F(\infty) < 1$  corresponds to stagnancy or dead volume, and it is usually preferred to redefine the system volume so that  $F(\infty) \equiv 1$ .

The decay or washout function, W(t), is defined as:

$$W(t) = 1 - F(t) \tag{1}$$

W(t) refers to the fraction of particles which experienced residence times greater than t so that W(0-) = 1 and  $W(\infty) = 0$ .

The differential distribution or frequency function is obtained by differentiating F(t):

$$f(t) = \frac{dF}{dt} \tag{2}$$

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This gives a non-negative function defined over  $(0,\infty)$ . It is common practice to use the Dirac delta function,  $\delta(t-t_o)$ , to represent f(t) at points where F(t) undergoes a step change.

Moments of the residence time distribution are defined in the usual way for non-negative random variables. The moments about the origin are:

$$\mu_n = \int_0^\infty t^n f(t) dt \tag{3}$$

where n = 0, 1, 2, ...

The zeroth moment is equal to unity as a consequence of requiring that  $F(\infty) = 1$ . The first moment is the mean of the residence time distribution and is usually denoted at  $\bar{t}$ . One would expect  $\bar{t}$  to be finite although this is not a direct consequence of the mathematical formulation. In fact, F(t) can be devised such that it satisfies the restrictions on cumulative distribution functions yet maintain infinite means. Higher moments from a mathematical sense may also be infinite; however, molecular diffusion ensures that all moments are finite in real systems.

A useful alternative to Equation (1) is:

$$\mu_n = n \int_0^\infty t^{n-1} W(t) dt \tag{4}$$

which can be verified through integration by parts. This expression is true whenever  $t^n W(t)$  approaches zero in the limit of large t which in turn will be satisfied if the next higher moment,  $\mu_{n+1}$ , exists.

Since t > 0, the  $\mu_n$  forms an ordered set such that  $\ln(\mu_n)/n$  is a non-decreasing function of n. If  $\mu_n$  is finite then so are all lower moments, and if  $\mu_n$  is infinite then so are all higher moments. If  $\mu_1 = 1$ , the distribution is normalized and the ordering of the set takes the form:

$$1 < \mu_2 < \mu_3 < \mu_4 \dots$$
 (5)

The limiting case with  $\mu_n = 1$  for all *n* represents a delta function distribution where all molecules have an identical residence time.

The moments of F(t) about the mean are defined as:

$$\mu_n' = \int_0^\infty (t - \bar{t})^n f(t) dt \tag{6}$$

where  $\mu_1' = 0$  and  $\mu_n > 0$  for all higher n. The various  $\mu_n$  and  $\mu_n'$  may be

related in the conventional manner for probability distributions; for example:

$$\mu_2' = \mu_2 - (\bar{t})^2 = \sigma_t^2 \tag{7}$$

and

$$\mu_3' = \mu_3 - 3\bar{t}\mu_2 + 2(\bar{t})^3 \tag{8}$$

where  $\mu_2' = \sigma_t^2$  is the variance of the distribution and  $\mu_3'$  is the skewness. The special case where  $\mu_n = 1$  for all n gives  $\mu_n' = 0$  for all n > 0 since there is no variation in residence times about the mean.

The  $\mu_n$  definitions are particularly useful as normalized distributions with  $\mu_1 = 1$ . They then provide a measure of the shape of f(t) which is independent of the magnitude of  $\bar{t}$ . Hence, to define a dimensionless normalized distribution for a given  $\bar{t}$ :

$$f_N(\theta) = \bar{t} f(\theta \bar{t}) \tag{9}$$

The moments of  $f_N(\theta)$  are related to those of f(t) by:

$$(\mu_n)_{\theta} = (\mu_n)_t / (\bar{t})^n \tag{10}$$

with a similar relationship holding for the  $\mu_n$ . The moments of  $f_N(\theta)$  are seen to provide dimensionless measures of the shape of the residence time distribution. The dimensionless variance

$$\sigma^2 = \int_0^\infty (\theta - 1)^2 f_N(\theta) d\theta = \frac{\sigma_t^2}{(\bar{t})^2}$$
 (11)

has proven particularly useful in fitting models to residence time distributions. It has a theoretical range of zero to infinity but the actual range in many practical situations is  $0 < \sigma^2 < 1$ .

It is useful to employ the Laplace transform of f(t), defined as:

$$\bar{f}(s) = \int_{0}^{\infty} e^{-st} f(t) dt$$
 (12)

where s is the transform parameter. If  $\bar{f}(s)$  is known, the moments of f(t) can be obtained by differentiation:

$$\mu_n = (-1)^n \frac{\text{Lim}}{s \to 0} \frac{d^n \overline{f}}{ds^n}$$
 (13)

Laplace transforms of residence time distributions are used for summing flow networks in a manner analogous to their use in summing electrical networks in linear circuit theory. Let  $f_1(t)$  and  $f_2(t)$  represent the differential distribution functions for two flow systems in series and let g(t) be the overall distribution function for the composite system. Suppose that the two vessels are statistically independent so that the time a particle spends in one vessel has no effect on the time it will spend in the other. Then the various distribution functions are related in the time domain by convolution.

$$g(t) = \int_{O}^{t} f_{1}(t - \tau) f_{2}(\tau) d\tau = \int_{O}^{t} f_{1}(\tau) f_{2}(t - \tau) dt$$
 (14)

and in the transform domain by multiplication

$$\bar{g}(s) = \bar{f}_1(s)\,\bar{f}_2(s). \tag{15}$$

If the flow systems are in parallel,

$$g(t) = wf_1(t) + (1 - w) f_2(t)$$
(16)

where w = fraction of the total particle flow which goes to flow system #1. This same additive rule applies in the transform domain:

$$\bar{g}(s) = w\bar{f}_1(s) + (1 - w)\bar{f}_2(s)$$
 (17)

Recycle flows can also be analyzed using the Laplace transform approach. There is no simple, time domain solution for the composite residence time distribution of the recycle system shown in Figure 1-1. The transform domain solution is:

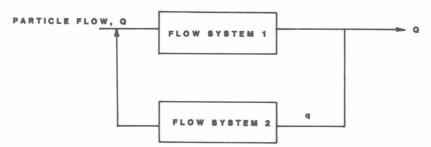


Figure 1-1. Illustrates a recycle system.

$$\bar{g}(s) = \frac{\bar{f}_1(s)}{1 + \frac{q}{O} \left[ 1 - \bar{f}_1(s) \bar{f}_2(s) \right]}$$
(18)

The above expressions enable the determination of residence time distributions (RTD) in flow networks of any complex. A key assumption in the calculations is that the subsystems are independent so that time spent in one vessel has no influence on time spent in another vessel or on the time spent in the same vessel should a particle return to that vessel. It should be noted that this assumption is not always satisfied in practice. Laminar flow systems routinely violate it as do fluid-particle systems where particle velocities depend on some parameter such as size.

Equation (15) is readily extended to i vessels in series, and the mean residence time for this series combination can be found using Equation (19)

$$(\bar{t})_{series} = \sum_{i} \bar{t}_{i} \tag{19}$$

Taking the second derivative and applying Equation (7) gives

$$(\sigma_t^2)_{series} = \sum_i (\sigma_t^2)_i$$
 (20)

so that means and variances are additive for series combinations of statistically independent flow systems. This is also true for the skewness,  $\mu_3$  but not for higher moments.

Closely related to the Laplace transform of f(t) is the generating function for the cumulants of the distribution

$$K_n = (-1)^n \frac{\text{Lim}}{s \to 0} \frac{d^n}{ds^n} (1n\bar{f}(s))$$
 (21)

Note that this relation gives the mean, variance and skewness directly without resorting to Equations (7) and (8). The fourth and higher cumulants are related to moments in a more complex manner. For example, the kurtosis of the distribution,  $\mu_4$ , depends on the second and fourth cumulants:

$$\mu_4' = K_4 + 3 K_2^2. \tag{22}$$

The cumulants have the useful property that they are strictly additive for independent systems in series.

### MEASUREMENT OF RTD

The ease of measuring residence time distributions depends on the nature of flow in the inlet and outlet streams rather than on the nature of the flow within the system. The ideal situation is to have uniform velocity profiles and no diffusion in the inlet and outlet transfer lines. Such a system is said to be closed, and closed systems allow easy determination of residence time distributions both mathematically and experimentally through transient response techniques. To illustrate this, consider a continuous flow system with a single inlet and single outlet. If we replace some fraction of the incoming particles with tracer particles which are identical in flow characteristics but which have some non-flow attribute which allows easy detection, then the tracer particles will follow the same paths through the system as did the original particles they replaced. Furthermore, they will have the same distribution of residence times as the original particles. The easiest way to measure this distribution is to inject the tracer particles in a short duration pulse which can be mathematically represented as a delta function,  $\delta(t)$ . The closed system under consideration allows such an injection and ensures that particles which once enter the system will stay until they finally leave. All the tracer particles will have entered the system at exactly the same time but will leave at varying times. By recording the times when particles leave, a histogram can be constructed which, with a large sample size, will converge to the differential distribution function, f(t). In experimental practice, a quantity of tracer is rapidly injected at the inlet to the system and the outlet concentration is monitored as a function of time, c(t). Then c(t) is normalized to give a differential distribution function, f(t), with  $\mu_o = 1$ :

$$f(t) = \frac{c(t)}{\int_{0}^{\infty} c(t) dt}$$
 (23)

Note that f(t) is the impulse response function for a closed system. The response of the system to more complex input signals may be found by convolution

$$c(t) = \int_{-\infty}^{t} c_{in}(\tau) f(t - \tau) d\tau$$
 (24)

Note that the lower limit on this expression differs from that on Equation (14) since  $c_{in}(\tau)$  is defined for all  $-\infty < \tau < t$  while  $f(\tau)$  is defined only for  $0 < \tau < t$ . Equation (24) allows determination of f(t) by using input signals such as sine waves and even random noise. Step change inputs are useful. Suppose  $c_{in}(t) = 0$  for t < 0 and  $c_{in}(t) = c_o = 1$  for  $t \ge 0$ . Then the system response gives the cumulative distribution function, F(t), directly. Similarly, a step-down change of the form  $c_{in}(t) = c_o = 1$  for t < 0 and  $c_{in}(t) = 0$  for t > 0 gives the residence time washout function, W(t).

Perfect delta function and step change inputs are possible only in the mathematical sense. There, they are commonly used as initial conditions to establish the residence time distribution corresponding to a mathematical model of the system. For example, the dynamic model for a perfectly mixed, stirred tank reactor is

$$\frac{d(Vc)}{dt} = Q_{in} c_{in} - Q_{out} c.$$
 (25)

Assuming V and Q are constant we apply a step change, c = 0 for t < 0 and c = 1 for  $t \ge 0$ , as the initial condition for Equation (25). The solution is

$$F(t) = c(t) = 1 - e^{-t/t}$$
 (26)

where  $\bar{t} = V/Q$ .

This represents the residence time distribution function since the modelled system is closed and since a perfect step change was applied to the inlet.

In experimental determinations it may not be possible to apply a good impulse or step change to the inlet of the system. If, however, the system is adequately closed and if the actual imperfect input signal can be monitored, then the true residence time distribution can still be determined. Equation (24) gives the theoretical justification for this statement:  $c_{in}$  and c are measured and Equation (24) inverted to give f(t). One approach is numerical Laplace transformation followed by numerical inversion, however this is cumbersome. A simpler, time doman approach is available when only the mean residence time, t, and variance,  $\sigma_t^2$  are needed rather than the entire distribution function. This approach was first popularized by Bischoff<sup>(1)</sup> and depends on the additive nature of means and variances in statistically independent series systems. Using the measured concentration response, c(t), one calculates the mean and variance for the composite system which includes the inlet line. Then  $c_{in}(t)$  is used to calculate t and  $\sigma^2$ for the inlet line alone, and these values are subtracted from the composite ones to obtain corrected results for the system alone. This approach is referred to as the imperfect pulse method, but it applies equally well when the input signal is an imperfect step change.