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**A QUANTITATIVE STUDY OF MIXING IN
ANNULAR AND RASCHIG RING FILLED TANKS**

Mark A. Fredrickson

Chemistry Research and Development
PROCESS CHEMISTRY SUPPORT

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A QUANTITATIVE STUDY OF MIXING IN ANNULAR AND RASCHIG RING FILLED TANKS

Mark A. Fredrickson

ABSTRACT

A quantitative study of mixing, which utilized the exit age distribution theory, has been completed for bench-scale annular and raschig ring filled tanks, in addition to a pilot-scale annular tank. A study of mixing and fluid flow in a full-scale, single-entry, raschig ring filled tank also was initiated. Results showed the bench-scale raschig ring filled tank approached plug-flow (no mixing) behavior. In a bench-scale annular tank, regardless of whether fluid entered tangentially or non-tangentially, the vessel approached continuously stirred tank reactor behavior (perfect mixing). The pilot-scale annular tank approached continuously stirred tank reactor behavior only when tangential entry was used. Mixing in the full-scale raschig ring filled tank did not approach either the perfect or the no mixing situations. Instead, the tank had a flow pattern in which solution channeled through a small volume of the tank, leaving the regions outside the channel effectively inactive.

SUMMARY

A quantitative study of mixing (via recirculation) has been completed for bench-scale annular and raschig ring filled tanks, in addition to a pilot-scale annular tank. Work was also initiated to study mixing and fluid flow in a full-scale raschig ring filled tank. Exit age distribution analysis

was used to obtain the quantitative results. This particular analysis was selected for two reasons. First, it was possible to calculate a theoretical average residence time, which was used to estimate the active volume of a vessel. Second, it was possible to convert raw concentration versus time data from an experiment into a normalized, dimensionless curve. Once the curves were obtained for each of the vessels (corresponding to different flow rates and inlet positions), the tanks were analyzed for the existence of non-ideal flow behavior (short circuiting and dead space). Relative mixing efficiencies also were determined for each vessel by comparing the dimensionless curves with two theoretical ideals (perfect mixing and no mixing).

The following conclusions were reached concerning mixing and fluid flow in each of the vessels. First, the bench-scale raschig ring filled tank approached plug-flow behavior (no mixing). Second, the bench-scale annular tanks approached perfect mixing behavior regardless of the mode of fluid entry. Third, the pilot-scale annular tank was characterized by fluid bypassing and poor mixing when fluid entered the vessel non-tangentially.

When fluid entered the pilot-scale annular tank tangentially, perfect mixing behavior was approached. Fourth, the full-scale raschig ring filled tank, with single entry, had significant problems with dead space and channeling.

INTRODUCTION

For many years, raschig ring filled tanks have been used to store and mix plutonium solutions. The raschig rings in the tanks are made of borosilicate glass and function as neutron absorbers to prevent a criticality accident. The volume of a raschig ring filled tank is generally between 275 and 400 liters; the tank contains rings that are 0.72-inch o.d. X 1.75 inches long with a wall thickness of 0.25 inch.

Raschig ring filled tanks function safely, but there are problems with maintaining them. First, periodic inspections of the vessels are necessary to check for sludge accumulation. Second, the rings require changing because of sludge fouling and breaking during mixing operations.

A frequent problem encountered in operating ring-filled tanks is that mixing in them is inefficient. When process solutions (such as plutonium solutions of different densities) are not mixed well in the vessels, unit operations located downstream from the tanks may be adversely affected. Hence, Chemical Operations is interested in replacing raschig ring filled tanks with a tank that is critically safe in addition to being an efficient mixing vessel. Annular tanks are the vessels currently being considered to replace some or all of the existing raschig ring filled tanks.

An annular tank is a device that, because of its geometry, can be used to safely store and mix solutions containing fissile nuclear materials. Because of its surface area-to-volume ratio, a properly designed annular tank can be used to mix radioactive solutions safely without requiring raschig rings for neutron poisoning. A typical full-scale annular tank is 3-feet o.d. X 7 feet tall with a 2-inch annulus.

This study was initiated to compare the mixing and sludge removal characteristics of annular and raschig ring filled tanks. The first phase of this study consisted of qualitative descriptions of flow and mixing in bench-scale annular and in raschig ring filled tanks. Quantitative experiments, which gathered internal concentration-versus-time data, were also performed.

The second phase of this project consisted of exit age distribution studies of bench-scale annular and raschig ring filled tanks. A pilot-scale annular tank also was studied with this technique. Exit age distribution analysis enabled the comparison of normalized, dimensionless, concentration-versus-time curves with the ideal cases of perfect mixing and plug flow. From this procedure, it was determined whether significant bypassing or stagnant regions existed in the process vessel under consideration.

BACKGROUND AND THEORY

A quantitative analysis of mixing, which details the flow of each molecule as it enters, travels through, and leaves a vessel, is impractical. However, a statistical approach, which yields similar though less detailed information, is available to quantitatively study mixing characteristics in process vessels. This statistical approach enables the investigator to compare the mixing characteristics of one vessel with another. The comparison may be between two completely different vessels of any size. Hence, valuable information for scale-up purposes may be obtained.

The statistical analysis just discussed is based on the exit age distribution theory. A detailed description of the theory is given by Himmelblau and

Bischoff.¹ This analysis assumes that an impulse of tracer can be introduced into the system in zero time. The input is referred to as an impulse, or Dirac delta function. Mathematically, the delta function has the following properties:²

$$\delta(t - \tau) = 0 \quad t \neq \tau \quad (1)^*$$

$$\int_{-\infty}^{+\infty} \delta(t - \tau) dt = 1 \quad t = \tau \quad (2)$$

As defined here, the delta function is discontinuous. It is equal to zero everywhere except at one point. In reality, these conditions cannot be met; however, they can be approximated fairly well.

Once the delta function has been delivered to the system, vessel response is determined by measuring the tracer concentration at various points in time near the vessel's exit. A function, $E(t)$, which is defined as the age fraction of tracer particles per unit time that leave the vessel, is calculated from the concentration versus time data. Hence, $E(t)dt$ is the fraction of tracer particles that leaves the vessel in time dt . For convenience, the curve represented by the equation is normalized (Figure 1) and the following equation is obtained:

$$\int_0^{\infty} E(t) dt = 1 \quad (3)$$

It is then possible to compute the theoretical mean residence time of the vessel by calculating the first moment of Equation 3. This is defined as

* Definitions for these and all other symbols and abbreviations used in this report are listed in Appendix A.

$$\bar{t}_{\text{theory}} = \int_0^{\infty} t E(t) dt \quad (4)$$

The theoretical mean residence time may then be compared with the experimental mean residence time, which is defined as

$$\bar{t}_{\text{experiment}} = \frac{V}{Q} \quad (5)$$

Experimentally it was not possible to determine these integrals exactly. Hence, they were approximated by taking finite sums as follows:

$$\sum_{t=0}^{t=\infty} E(t) \Delta t = 1 \quad (6)$$

where

$$E(t) = \frac{C(t)}{\sum_{t=0}^{t=\infty} C(t) \Delta t} \quad (7)$$

and

$$\bar{t}_{\text{theory}} = \sum_{t=0}^{t=\infty} t E(t) \Delta t \quad (8)$$

It can be seen that Equation 7 functions to normalize the raw concentration-versus-time data. Sample calculations are presented in Appendix B.

When it is assumed that all transport of mass into and out of the vessel occurs by bulk flow (no diffusional influx or efflux), the theoretical and experimental mean residence times are the same. When

FIGURE 1. General Exit Age Curve

these boundary conditions are met, $E(t)$ can be made dimensionless by the following equation. A rigorous derivation is given in Himmelblau and Bischoff.³

$$E(\theta) = \bar{t}_{\text{theory}} E(t) \quad (9)$$

Hence, when the operations defined by Equations 7 through 9 are performed on raw data, it is possible to generate dimensionless concentration-versus-time curves.

It is enlightening to compare dimensionless concentration-versus-time curves with two ideal cases. First, consider a continuously stirred tank reactor

(CSTR). In a CSTR, mixing occurs instantly as fluid enters the vessel. Hence, the effluent has the same concentration as the vessel itself. Performing a lumped, macroscopic mass balance on a CSTR (in which no reaction occurs), the following is obtained:

(Accumulation of mass)

$$= \left(\text{input of mass} \right) - \left(\text{output of mass} \right)$$

Written mathematically, the expression becomes

$$V \frac{dc}{dt} = Q C_0 - QC \quad (10)$$

FIGURE 2. Response of a Perfect Mixing Vessel to a Dirac Delta Function Input

FIGURE 3. Response of a Plug Flow Reactor to a Dirac Delta Function Input

Separating the variables and solving results in

$$\frac{C}{C_0} = 1 - e^{-\theta} \quad (11)$$

Himmelblau (see Reference 4) related Equation 11 to $E(\theta)$ and found that

$$E(\theta) = e^{-\theta} \quad (12)$$

Figure 2, a plot of Equation 12, shows the response of a CSTR to a Dirac delta function input at θ equals 0.

Consider now the case of a plug-flow reactor (PFR). In a PFR, fluid moves through the vessel without mixing. The analysis in this case is quite simple. The impulse leaves the vessel exactly as it entered after one mean residence time. Figure 3 shows the PFR response to a Dirac delta function input.

When deviations from plug flow are small, as in flow through long pipes and packed beds, an additional model may be used to analyze the data. Such flow occurs when bench-scale raschig ring filled tanks are mixed via recirculation. The model is called the axial dispersion model. It is based on a microscopic mass balance, which accounts for bulk mass transport and dispersion in the axial direction only. When written in dimensionless form, the axial dispersion model (see Reference 5) is

$$\frac{\partial C}{\partial \theta} = \frac{D}{UL} \frac{\partial^2 C}{\partial Z^2} - \frac{\partial C}{\partial Z} \quad (13)$$

Although Equation 13 appears rather formidable, it has been solved for boundary conditions similar to those found in the raschig ring filled tank.⁶ The dispersion number (D/UL) is the critical

parameter that determines how well fluid is mixed as it passes through the tank. The first step in calculating the dispersion number is to find the variance of the $E(\theta)$ versus θ curve. This is defined as follows:⁷

$$\sigma_{\theta}^2 = \frac{\frac{\sum C_i t_i^2}{\sum C_i} - \left(\frac{\sum C_i t_i}{\sum C_i} \right)^2}{\bar{t}^2} \quad (14)$$

The variance is then placed into the following equation to calculate the dispersion number.

$$\sigma_{\theta}^2 = 2\left(\frac{D}{UL}\right) - 2\left(\frac{D}{UL}\right)\left(1 - e^{-UL/D}\right) \quad (15)$$

When the dispersion number is zero, perfect plug flow occurs in the vessel. When the dispersion number is infinite, perfect mixing occurs in the vessel. In general, if the dispersion number is as large as 0.2, the vessel is considered well mixed.

The axial dispersion model does not, however, apply to the annular tank. This is because significant mass transfer occurs in the rotational direction around the annulus. Hence, one cannot assume that the concentration is uniform at any given cross-section of the vessel. Unless this assumption can be made with confidence, the dispersion number has little significance.

APPARATUS AND PROCEDURE

Table 1 lists the apparatus used in this investigation. Figures 4 through 8 show the tanks.

Figure 9 is a flowsheet for the bench-scale experiments. Water from a process supply was run through the purifying apparatus to the feed storage flask.

Feed storage was necessary since the purifier could supply water at a maximum rate of only 10 ml/sec, whereas some experiments required feed rates over 30 ml/sec.

The same general procedure was used to study both the bench-scale annular and raschig ring filled tanks. Only the flow rate or mode of fluid entry into the vessel was changed between experiments. The first step in setting up an experiment was to connect the desired fluid entry point on the vessel with the feed storage flask. For the annular tank with tangential entry, experiments were performed in which fluid entered directly above the drain and at points located 90°, 180°, and 270° clockwise from the drain. For the annular tank with non-tangential entry, mixing in the tank was studied when fluid entered directly above the drain and at a point 180° opposite the drain. Mixing in the ring-filled tank was studied when fluid entered through a 3/8-inch i.d. X 1-inch long pipe, a Bete Fog nozzle distributor, and an ordinary nole, all of which directed fluid into the center of the rings. Experiments also were performed in which fluid entered through an off-center hole.

Once the entry point was connected to the flask, the vessel was filled with water; the pump was then turned on to a predetermined flow rate (between 12 and 30 ml/sec). A constant volume was maintained in the vessel by opening the drain valve just as the pump started, and adjusting the drain valve until a steady state was achieved. Hence, in this procedure, the vessel did not function as a batch recirculation tank; instead, it was treated as part of a continuous process with the tank receiving fresh feed instead of fluid that had already been in the vessel.

TABLE 1. Equipment and Material Used in Study

Equipment	Description
<u>Bench-Scale Work</u>	
Annular tank (with tangential entry)	Made of pyrex glass, 14.5-cm i.d. X 47-cm tall with 2.5-cm annulus
Annular tank (no tangential entry)	Made of pyrex glass, 15.2-cm i.d. X 46-cm tall with a 2.4-cm annulus
Raschig ring filled tank	Made of pyrex glass, 15.2-cm i.d. X 30.5-cm tall, completely filled with 8- X 8-mm raschig rings
Pipe distributor for ring-filled tank	1.27-cm o.d. X 1.27-cm long standard pipe
Nozzle-type distributor for ring- filled tank	1.27-cm nozzle, Model TF 24N, sup- plied by Bete Fog Nozzles Inc.
Pump	Cole Palmer Peristaltic pump, MOD WZ1 R057, Ser 4MA97310, 33 ml/sec capacity
Pump controller	Cole Palmer Masterflex solid state controller
Tubing	3/8-in. (0.9525 cm) i.d. tygon
Conductivity cell	Devon Products, Model 5000
Dye tracer	Durkee Brand red food coloring, No. 4000
Lithium chloride [(LiCl) for salt tracer in bench-scale work]	Low sodium content, supplied by Van Waters and Rogers Scientific
Potassium chloride [(KCl) for salt tracer in pilot-scale work]	Supplied by Mallinckrodt Inc.
Balance	OHAUS Harvard Trip Balance, 2 kg capacity
Syringe	5-ml capacity, plastic disposable

(continued)

TABLE 1. (concluded)

Equipment	Description
Water purifier	Disposable Cartridge Type, IWT Research Model, S-27786, Sargent Welch Scientific
Feed storage	4000-ml Pyrex vacuum flask
<p style="text-align: center;"><u>Pilot-Scale Work</u> (in addition to some of those previously mentioned)</p>	
Annular tank	80-l volume, stainless steel, all connections to tubing are 3/8-in. i.d.
Pumps	Little Giant, Model 3E-12N
Conductivity apparatus	Beckman Instruments, Model SM3 meter (0-5 volt output) used with Model 414 cell and Model SW6 switch
Chart recorder	Fisher Porter Model 7808A0199A10
Raschig ring filled tank	340-l void volume stainless steel, all connections to tubing are 3/8-in. i.d.

At time zero, 5 ml of tracer (either 4500 ppm LiCl or 4500 ppm KCl) was injected into the feed line. The injection required 1-2 sec to perform. Samples were taken near the point through which fluid left the vessel. These samples were taken every 60 sec for flow rates between 12 and 15 ml/sec, and every 30 sec for flow rates greater than 15 ml/sec. Sample sizes ranged between 20 and 40 ml. After the samples were collected, sample analyses were performed with a Devon products conductivity meter.

For pilot- and full-scale work, a modified version of the experimental procedure just discussed was used. The first modification was to not use the water purifier. Since the volume of the pilot-scale annular tank was 80 liters, and that of the ring-filled tank was 340 liters, the only practical approach was to use tap water in the vessels. Hence, when conductometric data were analyzed, background conductivity was subtracted from the overall conductivity to find the tracer concentration. Next, to approximate a Dirac delta function,

23897-6

FIGURE 4. Bench-Scale Annular Tank Without Tangential Entry

50 ml of tracer had to be injected in less than 2 sec. This large quantity of tracer was necessary to obtain a good, measurable response at the vessel outlet.

It was assumed that if the injection required more than 2 sec to perform,

24774-10

FIGURE 5. Bench-Scale Annular Tank With Tangential Entry

the delta function was not approximated satisfactorily. To meet these constraints, a 50-ml syringe was connected to a 0.953-cm-i.d. X 15-cm-long piece of Tygon tubing. All injections were made directly into the pilot-scale annular tank through the flange in which fluid entered the tank. A few experiments were performed in which the tracer was injected directly into the feed line of

FIGURE 6. Bench-Scale Raschig Ring Filled Tank 24774-8

FIGURE 7. 80-Liter Pilot-Scale
Annular Tank 24774-2

25124-3

FIGURE 8. Full-Scale Raschig Ring Filled Tank With Instrumentation and Bench-Scale Vessel in Fore-ground. Author is shown next to tank for size comparison.

FIGURE 9. Bench-Scale Flow Diagram

FIGURE 10. Non-Tangential Entry Arrangement for Pilot-Scale Annular Tank and Entry Device for Ring-Filled Tank ²⁴⁷²⁶⁻⁸

the annular tank; however, the resulting impulse was frequently distorted. For experiments in which tangential entry was not used, injections were made through the short pipe (1/2-inch i.d.) shown in Figure 10. For experiments in which tangential entry was used, injections were made through the long tube (3/8-inch o.d.) shown in Figure 11. The long tube was constructed so the tracer would be input directly into the feed stream as it entered the tank.

Two different tracers were employed to investigate two different mixing situations in the annular tank. The first

FIGURE 11. Tangential Entry Arrangement for Pilot-Scale Annular Tank ²⁴⁷²⁶⁻⁵

situation concerned the case in which a dense solution was added to a light solution already in the tank. For this case, a concentrated potassium chloride (KCl) solution, which was 15% denser than water, was used as the tracer. This salt was used because its specific

conductivity changes linearly with concentration over the entire range from dilute to saturated solution. All concentration data were obtained conductometrically when this salt tracer was used.

The second situation was the case in which the entering fluid had the same density as the solution already in the annular tank. For this case, a dye tracer, which had the same density as water, was used. All concentration data were obtained colorimetrically when dye tracers were used.

The procedure for investigating tangential and non-tangential entry in the annular tank was the same. At time zero, 30 to 50 ml of tracer was injected through the flange as discussed previously. Some experiments required sample collection near the annular tank's exit. These samples were analyzed conductometrically or colorimetrically, depending on the type of tracer used. In these cases, the samples were taken downstream from the pump, at predetermined time intervals, through a special connection to the exit line. Figure 4 shows this arrangement. Before a sample was taken, the sampling valve was opened for 10 to 15 sec to allow trapped, non-representative fluid to escape.

In other experiments, continuous specific conductivity-versus-time data were obtained. The setup used to obtain these data was as follows. First, a Beckman Instruments conductivity cell was lowered through the annular tank to a position close to the vessel's drain. It was positioned so the cell's electrode was not in contact with the steel walls of the tank. The cell was connected to a switch, which was connected to a conductivity meter.

The voltage output from the meter was then recorded by a Fisher Porter chart recorder. With this system, once the tracer injection was performed, concentration-versus-time data were obtained automatically.

The procedure for the study of mixing in the full-scale raschig ring filled tank was nearly the same as in the pilot-scale annular tank. The inlet position into the ring-filled tank in this phase of the investigation was always into the center of the rings. The flange shown in Figure 10 was placed directly on top of the rings in the tank so that water would flow into the center of the vessel. The tracer injection technique was the same as that employed when studying mixing in the pilot-scale annular tank with non-tangential entry. At this phase of the research, only KCl tracers were employed. After the injection was made, continuous specific conductivity-versus-time data were obtained at the vessel's exit. Flow rates used to study mixing in the ring-filled tank were between 8 and 15 l/min.

RESULTS AND DISCUSSION

The exit age distribution theory has been proven useful in the quantitative study of mixing in annular and raschig ring filled tanks. It has been valuable for comparing mixing and flow behavior data of bench-scale annular tanks with a pilot-scale annular tank. It has also been used to compare the mixing characteristics of bench-scale annular and raschig ring filled tanks. Finally, exit age distribution analysis has been used to investigate some of the density effects of mixing, and to determine the best inlet flow conditions in the pilot-scale annular tank for optimum mixing.

As discussed in the previous section, the tanks were treated as if they operated continuously rather than batchwise. There may be concern that this does not give information applicable to real-life situations in which tanks do operate batch-wise. However, when continuous-type experiments were performed, it was possible to gain information that could not be gained in other ways. For example, in batch-type experiments in which a tracer was injected and recirculated until mixed, it was difficult to differentiate between convective and diffusive mixing effects. Any system would eventually become mixed by diffusion alone. Such experiments gave little data, however, concerning the effectiveness of existing flow patterns in mixing solutions without the aid of natural diffusion. Also, there were no ideals (such as CSTR or PFR behavior) with which to compare the results of batch-type experiments. By treating the vessel as part of a continuous process, it was possible to detect fluid bypassing, determine if dead space existed, and estimate the degree of mixing by comparing experimental curves with theoretical ideals.

A number of exit age distribution curves have been obtained for the bench-scale raschig ring filled tank. Figure 12 presents the results from three experiments, all carried out at different flow rates. The solid line in Figure 12 shows the results of an experiment in which fluid entered the bench-scale ring-filled tank through an off-center hole in the lid, with no distributor, at 16.2 ml/sec. Visual observations during the experiment determined that channeling occurred down the wall and also through the rings. The curve represented by dashes was obtained from an experiment in which fluid entered the tank through a pipe in the center of the

FIGURE 12. Exit Age Distribution Curves For Raschig Ring Filled Tank

lid at 13.8 ml/sec. Very little channeling down the wall was observed, but channeling down through the rings was seen. The dotted curve shows the results of an experiment in which the fluid entered through a Bete fog nozzle distributor in the center of the lid at 24.6 ml/sec. Again, very little channeling down the tank wall was observed, but channeling down the rings was seen.

After the curves were inspected, the following conclusions were reached. First, since each curve has but one peak, there was no fluid bypassing present. In other words, fluid passed through in

one bulk mass, not two or three. Second, dead space did not exist in the vessel. Generally, if fluid elements require longer than 10 times the mean residence time ($\theta = 10$) to pass through the vessel, that fluid would be effectively stagnant. A curve representing a vessel having dead space would have a long tail extending past $\theta = 10$.

Theoretical average residence times also were in close agreement with experimental average residence times, which indicated that the entire void volume of the vessel was active.

Non-idealities such as bypassing and dead space were not problems with the bench-scale ring filled tank; nevertheless, the situation was far from good. Each curve in Figure 12 resembles a Gaussian distribution and has a high maximum point [$E(\theta) \sim 1.55$]. The axis of symmetry for each curve occurs near $\theta = 1$. A dispersion number of 0.04 was calculated for each curve. The dispersion number indicates that fluid flowed through the rings in a dispersed plug-flow fashion. After the shape of the curves were compared with the plug-flow case, it was evident that the flow of fluid through the raschig ring filled tank approached plug-flow behavior (no mixing) as an ideal. Qualitatively, the situation was imagined like this. After a delta function was injected into the system, each tracer particle passed through the vessel, intermingling (mixing) only slightly with other particles. Bulk flow (convective forces) caused particles to move the axial direction. Dispersion, a macroscopic analogue to diffusion, was the source of particle intermingling. The driving force for dispersion was the concentration gradients that tended to promote mixing. However, since dispersion forces were small and bulk flow was large, the

impulse moved through the vessel relatively unchanged.

After exit age distribution curves for the bench-scale ring filled tank were inspected, it became evident that flow rate had little bearing on mixing performance. In all of the cases investigated, regardless of flow rate and with or without a distribution mechanism, the bench-scale ring filled tank approached plug-flow behavior.

Mixing experiments also were performed in a full-scale raschig ring filled tank to determine if bench-scale findings applied on scale-up. Figure 13 shows the results from an experiment (using a KCl tracer) that was typical of the many experiments performed at full scale. The curve does not resemble those obtained from bench-scale raschig ring filled tank experiments, which indicates that fluid does not flow through the full scale, single entry, raschig ring filled tank in a dispersed plug-flow fashion. The high peak $E(\theta) = 1.35$ indicates channeling; however, the severity of the problem is not obvious until theoretical and experimental average residence times are compared.

The experimental average residence time was calculated on the assumption that the entire void volume, up to the level of the rings, was active (340 liters). Since the theoretical average residence time was only about 1/6 of the experimental number, much less of the tank's void volume was actually involved in the experiment. Using the theoretical mean residence time, the theoretical active volume of the tank was found to be only 59 liters. Hence, about 83% of the full-scale raschig ring filled tank's volume was dead space. This number serves only as a rough estimate of the actual dead space in the tank because of approximations involved in the computation. The

FIGURE 13. Exit Age Distribution Curve for Full-Scale Raschig Ring Filled Tank

number does indicate, however, that channeling and dead space are severe problems in the ring-filled tank. With so much dead space, mixing is extremely inefficient in single entry, raschig ring filled tanks.

The next step in this investigation was to examine exit age distribution curves for the bench-scale annular tanks. First, the case of non-tangential fluid entry was considered. The flow rates used in each of the experiments was 12 ml/sec. As indicated in Figure 14, only the placement of the inlet relative to the drain was changed between experiments.

An analysis of the curves yielded the following conclusions. First, bypassing and dead space were not problems. This was indicated since the curves had one peak, the tails did not extend past $\theta = 10$, and theoretical average residence times agreed well with experimental average residence times. Second, the curves approach CSTR behavior (perfect mixing) as an ideal. For most experiments with the bench-scale annular tanks, the peaks were much lower (and occurred earlier) than for the ring-filled tank. Generally, for the annular tank, the peak was at $E(\theta) = 0.9 \pm 0.1$. Hence, for the bench-scale annular tank with non-tangential entry, it was concluded that the position of the inlet did not affect

FIGURE 14. Exit Age Distribution Curves for Bench Scale Annular Tank With Non-Tangential Entry

the vessel's performance. The same type of experiments performed at higher flow rates yielded similar results.

Figure 15 shows exit age distributions for the bench-scale annular tank with tangential entry. The flow rate used in the experiments that generated this family of curves was 30 ml/sec, which resulted in a mean residence time of 2.09 min. The major purpose of this phase of the research was to determine how exit age distributions for tangential inlets were affected by flow rate and inlet position. As shown in Figures 15 and 16, the inlet position had no significant effect on the shape of the curve. When the flow rate was slowed

from 30 ml/sec to 12 ml/sec, the family of curves shown in Figure 16 was obtained. The major difference between curves in Figures 15 and 16 is that the curves for high flow rates rise faster and peak slightly earlier. This means that at high flow rates, fluid elements required a smaller fraction of the vessel's mean residence time to pass through the tank than at low flow rates.

On comparing tangential versus non-tangential entry at the bench scale, the following conclusions were reached. First, for both cases the annular tank approximated CSTR mixing behavior. Second, because tracer particles had a shorter distance to travel when they entered non-tangentially, the curve

FIGURE 15. Exit Age Distribution Curves for Annular Tank With Tangential Entry

shifted farther to the left than at the same flow rate when tangential entry was used. Hence, it was concluded that the mode of fluid entry had little bearing on the performance of the bench-scale annular tank.

To determine whether bench-scale findings apply on a larger scale, pilot-scale annular tank experiments were performed. As in the bench scale, tangential versus non-tangential entries were compared to determine if the mode of fluid entry affected mixing performance. For each mode of entry, different fluid flow rates, inlet positions, and tracer densities were used to find the conditions for optimum mixing.

Figure 17 presents the results from two experiments in which non-tangential entry was used. In the first experiment, the tracer was a concentrated KCl solution that had a density 15% greater than water. The solid curve in Figure 17 represents this experiment and shows a rapid, high peak. The peak is then followed by a long tail. This curve, as a whole, does not approach either of the ideals discussed earlier; hence, its interpretation is more difficult, but it may be understood as follows.

First, a significant fraction of the total area is contained under the first peak. This means that a large fraction of the tracer particles passed through

FIGURE 16. Exit Age Distribution Curves for Annular Tank With Tangential Entry (Slow Flow Rate)

the tank in a short time. When non-tangential entry was used in the pilot-scale annular tank, channeling developed, which enabled some of the fluid to pass through the tank without being well mixed.

As the bulk of the impulse traveled through the vessel, the outermost layer of molecules tended to be washed away by convective forces. In addition, a large concentration gradient existed between the pulse and water in the tank. Hence, diffusional forces tended to draw molecules from the pulse. The tail on the exit age curve corresponds to tracer particles that left the bulk pulse but remained in the tank for a long time.

The second experiment, indicated by the dashed line, made use of a dye tracer that had the density of water. That experiment was performed to determine what effect tracer density changes had on altering the shape of the curve. It was thought that a low-density tracer would have a lower propensity for channeling. This was not the case, however. It may be concluded from this information that channeling, when non-tangential entry was used, was a function of convective forces and not gravitational, density forces.

In an effort to improve the vessel's performance, fluid flow patterns were

FIGURE 17. Exit Age Curves for Pilot-Scale Annular Tank, Entry Opposite Exit

modified. In some experiments, the non-tangential inlet was repositioned with respect to the drain. Different inlet flow rates (2.79 l/min to 12 l/min) also were used in the investigation. For all experiments in which non-tangential entry was used, a high peak followed by a long tail was observed. Unfortunately, the height of the peak and the shape of the tail were not highly reproducible. The lack of reproducibility was due to the many random events that occurred when non-tangential entry was used. Random events were induced by the tur-

bulent channeling effects in the vessel; hence, how distorted an impulse was when it exited the vessel and the time at which it exited were slightly different from one experiment to the next.

When tangential entry was used in the pilot-scale annular tank, major problems such as fluid bypassing and channeling were eliminated. Figure 18 presents the results from two typical experiments. The solid curve is for a case in which the inlet was opposite the drain. There is one minor peak, which indicates a

FIGURE 18. Exit Age Curves for Pilot-Scale Annular Tank With Tangential Entry

small degree of bypassing. However, the area under the small peak, which corresponds to the fraction of particles involved in bypassing, is insignificant compared to the total area. Thus, bypassing was not a major problem. When the entrance was placed above the drain, it was more difficult for any bypassing to develop since the particles were required to traverse a longer distance to reach the drain. In short, the optimum position for the tangential inlet was directly above the drain. However, even when a less than optimum position was used, the mixing was still quite good.

For experiments in which tangential entry was used, data were more reproducible. This indicates that good, predictable results will be possible when actual production operations start using annular

tanks with tangential entry. Results from pilot-scale experiments involving tangential entry have shown that at higher flow rates, the exit age curves shift close to the $E(\theta)$ axis. This indicates that, as one would expect, mixing in the system at higher flow rates more closely approximates perfect mixing. As in the non-tangential entry case, it was determined that the density of the tracer did not affect the resulting exit age distribution curve.

After comparing bench-scale annular tanks with the pilot-scale annular tank, similarities and differences both were found. Exit age distribution studies have shown that when fluid entered tangentially, an annular tank on either scale responded the same. When non-tangential entry was used, the relative

FIGURE 19. Exit Age Curve for Pilot-Scale Annular Tank With Non-Tangential Entry (Low Flow Rate)

responses were different. In an effort to determine why channeling occurred under these conditions, an experiment was performed in which the fluid entered the pilot-scale annular tank at 0.96 1/min. This flow rate was in the range used in the bench-scale experiments (0.70 to 1.50 1/min). If the absolute flow rate alone was responsible for channeling, the vessel would have approached CSTR behavior. Figure 19 presents results from the experiment just discussed, in which a dye tracer was used. As seen in the graph, a high peak

is followed by a long tail. The peak of the curve actually occurred at $E(\theta) = 7.5$. It was determined that approximately 15 to 20% of the tracer channeled through the tank. Similar results were obtained when a salt tracer was used instead of the dye. For the pilot-scale annular tank with non-tangential entry, channeling occurred regardless of both the inlet flow rate and tracer density.

Tangential entry tended to eliminate channeling and bypassing problems. Since

the fluid was forced to rotate around through the annulus, sufficient opportunity was available for particle intermingling. The probability was small that a significant fraction of fluid particles would find a short circuit to the vessel exit, against the mainstream flow. As the flow rate increased, it became more difficult to short circuit; homogenization therefore was accomplished faster.

CONCLUSIONS

Upon analyzing exit age curves for each of the vessels studied, the following conclusions were reached:

1. Bench-scale raschig ring filled tank approached plug-flow behavior (no mixing).
2. Bench-scale annular tanks approached continuously stirred tank reactor (perfect mixing) behavior.
3. Pilot-scale annular tank behavior depended on the mode of fluid entry into the tank.
 - a. Non-tangential entry was characterized by fluid bypassing and less-than-ideal mixing.
 - b. Tangential entry caused mixing to be highly efficient.
4. The full-scale raschig ring filled tank was characterized by severe amounts of dead space and channeling.

RECOMMENDATIONS

Tracer studies should be used to consider different inlet positions relative to the drain in the full-scale raschig ring filled tank. Dye tracers also

should be used to consider density effects on mixing in the raschig ring filled tank. In other words, the importance of convective versus diffusive effects in the mixing of fluids in the ring filled tank should be determined. The theoretical average residence times and exit age curves from dye tracers may be different from those obtained using salt tracers. This possibility should be explored. Actual recirculation studies also should be performed to determine the time required to mix stratified layers in the full-scale raschig ring filled tank.

For the annular tank, exit age studies employing a full-scale vessel should be performed to determine if pilot-scale results apply on further scale-up. Sludge removal experiments should be performed, too.

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APPENDIX A ABBREVIATIONS AND SYMBOLS

Abbreviations

C	- Concentration Leaving a vessel
C_i	- Raw concentration data at a point in time
C_o	- Concentration entering a vessel
$C(t)$	- Concentration as a function of time
D	- Axial dispersion coefficient, cm^2/sec
e	- Base of a Napierian logarithm
$E(t)$	- Exit age as a function of time
l	- Length of a vessel to a point
L	- Total length of a vessel
Q	- Flow rate into a vessel
t	- Time
t_i	- Point in time

t

- Mean residence time

U - Flow rate into vessel, same as Q

V - Volume of a vessel

Z - Fractional length of a vessel (1/L)

Greek Symbols

Δ - Finite difference

$\delta(t - \tau)$ - Dirac delta function

θ - Dimensionless time ($t/\bar{t}_{\text{theory}}$)

$E(\theta)$ - Exit as a function of dimensionless time

τ - An arbitrary point along time axis

σ^2 - Variance

σ_{θ}^2 - Dimensionless variance

APPENDIX B SAMPLE CALCULATIONS

Consider the bench-scale raschig ring filled tank with the following raw data. The analysis for all experiments was similar to what is demonstrated here.

Concentration (ppm)	0.00	0.00	0.00	0.00	0.40
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Time (min)	0.0	0.5	1.0	1.5	2.0
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Concentration (ppm)	1.10	1.30	1.10	0.75	0.45
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Time (min)	2.5	3.0	3.5	4.0	4.5
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Concentration (ppm)	0.29	0.15	0.08	0.0
---------------------	------	------	------	-----

Time (min)	5.0	5.5	6.0	6.5
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$$\Sigma C \Delta t = (0.40 + 1.1 + 1.3 + 1.1 + 0.75 + 0.45 + 0.29 + 0.15 + 0.08) (0.5)$$

$$= 5.62 (0.5)$$

$$= 2.81 \text{ ppm/min}$$

To find $E(t)$ at a point, the concentration at that point is divided by $\Sigma C \Delta t$. At $t = 2.5$, we have $1.10/2.81 = 0.39 \text{ min}^{-1}$

<u>t(min)</u>	<u>E(t)(min⁻¹)</u>	<u>tE(t)</u>
0.0	0.00	0.00
0.5	0.00	0.00
1.0	0.00	0.00
1.5	0.00	0.00
2.0	0.14	0.28
2.5	0.39	0.98
3.0	0.46	1.39
3.5	0.39	1.37
4.0	0.27	1.07
4.5	0.16	0.72
5.0	0.10	0.52
5.5	0.05	0.29
6.0	0.03	0.17
6.5	0.00	0.00

Adding up the entries in the $tE(t)$ column, we find

$$\Sigma t E(t) = (0.28 + 0.98 + 1.39 + 1.37 + 1.07 + 0.72 + 0.52 + 0.29 + 0.17) = 6.79$$

To find $\bar{t}_{\text{theory}}$, the $tE(t)$ total is multiplied by Δt :

$$\begin{aligned}\bar{t}_{\text{theory}} &= \sum t E(t) \Delta t \\ &= 6.79 \text{ (0.5 min)} \\ &= 3.39 \text{ min}\end{aligned}$$

Since $\theta = t/\bar{t}$ and $E(\theta) = \bar{t} E(t)$, the following table can be constructed:

θ	$E(\theta)$
0.00	0.00
0.15	0.00
0.29	0.00
0.44	0.00
0.59	0.47
0.74	1.32
0.88	1.56
1.03	1.32
1.18	0.92
1.33	0.54
1.47	0.34
1.62	0.17
1.77	0.10
1.92	0.00

To calculate a dispersion number for this case, the following steps are taken.

$$\begin{aligned}\sum C_i &= (0.40 + 1.1 + 1.3 + \dots + 0.08) \\ &= 5.62\end{aligned}$$

$$\sum t_i C_i = [0.4(2.0) + 1.10(2.5) + \dots + 0.08(6.0)]$$

$$= 19.08$$

$$\begin{aligned}\sum t_i^2 C_i &= [(2.0)^2(0.4) + (2.5)^2(1.10) \\ &\quad + \dots + (6.0)^2(0.08)] \\ &= 69.43\end{aligned}$$

$$\begin{aligned}\sigma_t^2 &= \frac{\sum t_i^2 C_i}{\sum C_i} - \left[\frac{\sum t_i C_i}{\sum C_i} \right]^2 \\ &= \frac{69.43}{5.62} - \left[\frac{19.08}{5.62} \right]^2 \\ &= 0.83\end{aligned}$$

$$\sigma_\theta^2 = \frac{\sigma_t^2}{\bar{t}^2} = \frac{0.83}{3.39^2} = 0.072$$

Using the following equation, D/UL is found by trial and error.

$$0.072 = 2 \frac{D}{UL} - 2 \left(\frac{D}{UL} \right)^2 (1 - e^{-UL/D})$$

Ignoring the second term on the right, the following is obtained as a first approximation.

$$D/UL = 0.036$$

By plugging this number into the entire equation, the equation is satisfied. Thus, D/UL equals 0.036 is our final answer. This indicates that an intermediate amount of dispersion existed in this case.