trode diameters, the region of sensitivity is approximately five disturbance volumes.

Sensitivity of the system to changes in concentration in the vicinity of the probe tip was such that concentration variations of the order of one part in 10^6 could be accurately measured. These fluctuations contained measurable frequency components over the range of 0 to 8,000 c/sec. In the absence of mechanical vibration of the probe the signal to noise ratio of the entire system decreased from a value of 20:1 at 20 c/sec. to a value of 3:1 at 8,000 c/sec.

ACKNOWLEDGMENT

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NOTATION

C = capacitance, farads
$C_{1,2}$ = large values of capacitance resulting from orientation of ions and polar molecules in the fluid at electrode surfaces
C_3 = capacitance existing between wires connecting probe to electronic equipment
C_4 = capacitance resulting from presence of dielectric between probe wires
C_5 = capacitance between two exposed electrodes
E = potential, v.
I = current, amp.
j = $(-1)^{1/2}$
K = probe constant, cm.$^{-1}$
k = specific conductance, (ohms)$^{-1}$(cm.)$^{-1}$
R = resistance, ohms
R_P = total resistance of conducting fluid
$R_{1,2}$ = resistances from each electrode to grounded probe sheet; infinite if grounded probe sheet is not used
$R_{3,4}$ = resistances from each electrode to ground via conducting fluid
X_C = capacitive reactance, ohms
Z_T = probe impedance, ohms
ω = angular frequency, radians (sec.)$^{-1}$

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Mixing in a Jet-Stirred Reactor

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Measurements were made of the over-all mixing in a gas phase flow reactor stirred by the entering feed jets. The mixing was studied by following the decrease in exit concentration after sharp cutoff of a radioactive tracer gas krypton —85. The data showed that over the entire range of average residence times investigated, about 0.4 to 16 sec., the reactor behaved as though 85% of its volume was perfectly stirred and the remaining 15% was in piston flow in series with the stirred region. Relocation of the feed jets would probably increase the fraction of volume that is effectively perfectly stirred to about 95%.

A study was made of the over-all quality of mixing in a gas phase flow reactor stirred by the incoming feed jets. The reactor was designed to be completely stirred, that is completely uniform in composition, and the intent of this study was to determine how closely the design objective had been realized. The mixing study was essentially a study of the distribution of residence times of gases flowing through the reactor. Because the ideal completely stirred reactor represents one extreme of distribution of residence times varying exponentially from 0 to ∞, its theory has been analyzed extensively by a number of workers, for example (*1 through 5*). At the other extreme is the ideal piston flow reactor in which all material has the same residence time. Despite the theory no gas-phase reactor has been shown to be perfectly stirred in practice, although Longwell and Weiss (*6, 7*) found it useful to assume perfect stirring for their spherical combustion reactors.

EXPERIMENTAL PROCEDURE

The jet-stirred reactor used in this study was a squat cylindrical vessel with an inside diameter of 3.5 in. and an inner length of 1.125 in. Its net reaction volume, between inlet and outlet holes, was 161 cc. The photograph in Figure 1 presents an exploded view of the reactor. The flow through the reactor is shown schematically in Figure 2.

The gas feed, entering at the left, jets into the reactor tangentially through holes in the sixteen feed quills. Each quill has eight 0.0135-in. holes spaced 0.125 in. apart. The jets cause the reactor contents to swirl around the axis, and the gases finally leave the reactor through sixty holes, 0.0595-in. diameter, in the cylindrical surface on the reactor axis.

The mixing was studied by means of a tracer-decay technique developed in this laboratory (*8*) with krypton-85 gas which has a half life of 10.3 yr. During the experiments nitrogen gas was passed through the reactor at atmospheric temperature and pressure and at constant rates ranging from 10.4 to 407 cc./sec. Added to this main nitrogen stream was a tracer stream amounting to about 2 vol. % of the main stream and consisting of 0.7 vol. % krypton in nitrogen. The concentration of krypton was measured in the exit line by the intensity of γ radiation picked up by two scintillation probes mounted on opposite sides of the exit line. Digital scalers were used to count the emission rates, and the outputs were recorded on three channels simultaneously to cover the full range of radiation intensities used.

The distribution of residence times within the reactor was measured by allowing the system to come to steady state with constant flow of main and tracer streams. The tracer stream was then cut off sharply by a solenoid valve, and the rate of decay of krypton concentration was observed. Decay rates were corrected for background radiation and for the time lags in the inlet and outlet lines.

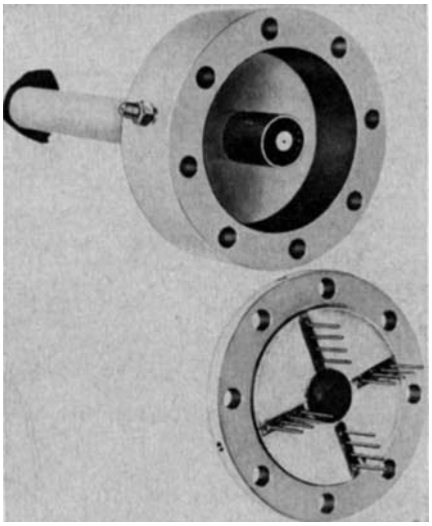

Fig. 1. Exploded view of the jet-stirred reactor.

A few experiments were conducted in which the decay was followed at the reactor walls, instead of at the outlet. However, these measurements did not produce significant results, since the scintillation probes could see only those portions of the reactor space located closest to the walls rather than the total contents of the reactor.

RESULTS AND INTERPRETATION

Experimental conditions for the mixing study are summarized in Table 1. The time lags listed are equal to the inlet or outlet volume (from the tracer injector outlet to the quill outlet or from the exit holes to the scintillation probes) divided by the volumetric flow rate of the main nitrogen stream. That is plug flow was assumed in these regions. Calculations showed that the plug-flow assumption introduced only a small error as judged by the effective time lags calculated by assuming reasonable velocity distributions and back mixing in the inlet and outlet regions.

If the reactor were perfectly stirred, the time lag-corrected rate of fall off of krypton concentration in the outlet stream would be expressed by

$$\frac{C}{C_o} = e^{-\frac{t}{T}} \tag{1}$$

where C is the concentration at time t after cutting off the flow of tracer, C_o is the initial steady state concentration, and T is the average residence time (reactor volume divided by volumetric flow rate). Therefore the experimental data were plotted dimensionlessly as log C/C_o vs. t/T. Over the entire range of flow rates used the experimental data deviated in the same way from the straight line predicted by the perfectly mixed model of Equation (1).

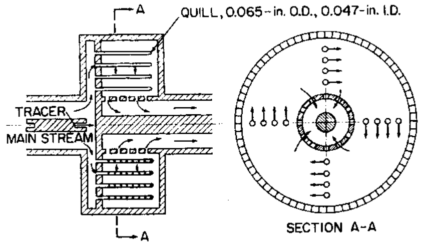

Fig. 2. Schematic diagram of the reactor indicating expected pattern of fluid motion.

Figure 3 illustrates this deviation. Therefore an attempt was made to interpret the results by fitting them to a theoretical mixing model based on some simple combination of ideal reactor units, that is perfectly mixed and plug-flow reactors. The complexity of combinations containing more than one of each unit, or containing other reactor types, was considered to be unwarranted by the data.

Table 2 lists the concentration of tracer, relative to the initial steady state concentration, for two reactor combinations, and compares them to the simple plug flow and perfectly stirred systems. The first combination consists of stirred- and plug-flow reactors in parallel, with the volume of the stirred reactor equal to a fraction x of the total volume. A fraction x of the feed enters the stirred reactor, and the remainder enters the plug flow reactor. Concentrations are given for the total tracer present within both reactors at t and for the tracer in the combined exit stream. Note that the internal and exit concentrations are not the same as they are in the case of a single perfectly stirred reactor. The second combination consists of plug and stirred reactors in series (the results are unaffected by reversing the order of the reactors in the series).

The plotting of some typical curves gives the results shown in Figure 4. Inspection of Figure 4 shows that the general shape of the decay curves for the series combination is the same as the shape of the experimental data illustrated in Figure 3. A best fit of the experimental data from the reactor studied results by assuming that 85% of its volume is prefectly stirred and 15% of its volume is in piston flow, regardless of the actual residence time used. Figure 5 shows how well the data can be matched by assuming this reactor behavior. Since at low tracer concentrations the experimental values of C/C_o are strongly influenced by any error in the background rate, it is more important to match the data with the calculated model for $t/T < 1$. The background rates were estimated from

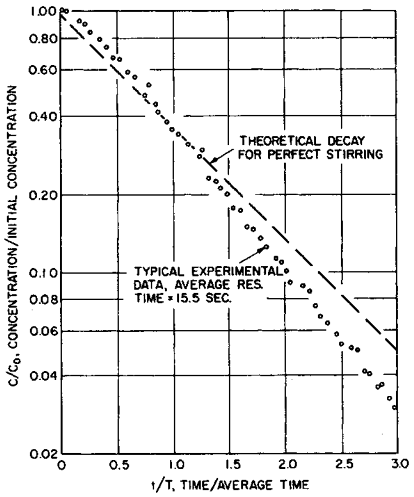

Fig. 3. Typical krypton tracer decay results, flow rate = 10.4 cc./sec.

the final values of the recorded decay and therefore were likely to be somewhat overestimated. For example at a flow rate of 203/sec. an error of 20% in the background rate would lower by 18% the value of C/C_o corresponding to t/T = 3.0. The same error however would decrease C/C_o only by 1.7% at t/T = 1.0.

A plausible interpretation of the results of the decay experiments is suggested by the physical arrangement of the reactor. This indicates that the 85% of the reactor volume which is well mixed precedes the plug-flow zone. It is likely that the gases entering the reactor through the high-velocity jets are rapidly dispersed in the well-mixed zone. The extent of this zone is probably limited to the cylindrical volume which contains the feed jets.

However the gases must eventually leave this well-mixed zone and flow radially towards the center and out of the reactor. The volume of this radial flow zone contained within the well-stirred outer concentric volume represents about 15% of the total reactor volume and may well be the region in effectively plug flow. A scaled diagram of the reactor in Figure 6 shows that the plug-flow section starts just inside the diameter which corresponds to the innermost feed quills if the preceding physical picture of the flow is correct. Therefore improved mixing should be obtained by rearranging the feed quills, bringing some of them closer to the center, or by installing additional quills in order to minimize the fraction of reactor space unoccupied by feed jets.

One of the most interesting results of this work was the independence of mixing pattern from average gas velocity, that is residence time, despite a forty-fold variation in velocity. This result emphasizes that geometry is the critical variable in turbulent mixing and that flow conditions are of little importance as long as the flow regime does not approach laminar conditions at one extreme or large compressibility effects at the other.

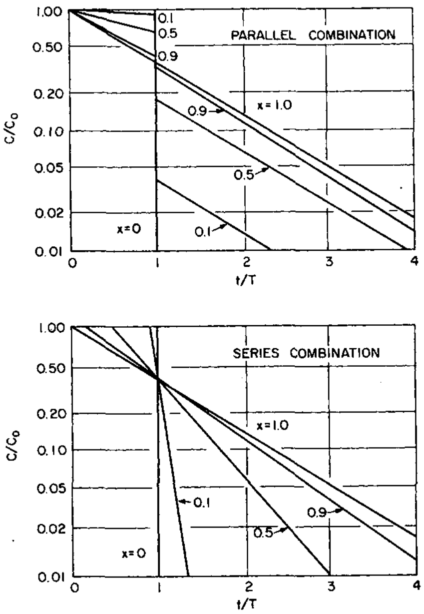

Fig. 4. Exit tracer concentrations for combinations of one stirred and one piston flow reactor.

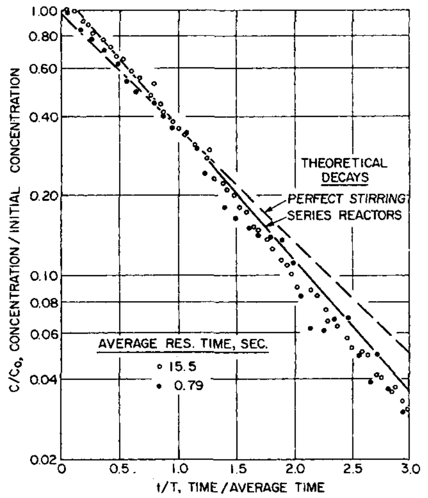

Fig. 5. Correlation of experimental decay results with series model for $x = 0.85$.

CONCLUSIONS

The use of krypton-85 as a γ-emitting tracer permits the study of over-all mixing quality in gas-phase reactors with residence times as short as 0.4 sec. Although shorter residence times could be studied, down to 0.1 sec. or so, this tracer technique does not appear practicable for residence times as short as the 0.3 msec. reached in the reactors of Longwell and Weiss (*6, 7*). The results here show that it is possible to construct a gas phase flow reactor, with residence times of seconds or tenths of seconds, which is 85% perfectly stirred on a macromixing scale at least. By some modifications close to 95% perfect stirring should be attainable.

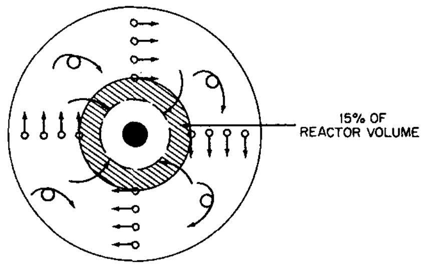

Fig. 6. Physical representation of the mixing in the jet-stirred reactor; the cross-hatched space at the center represents that 15% of the reactor volume in plug flow which follows in series the well stirred 85% of the reactor volume.

TABLE 1. SUMMARIZED EXPERIMENTAL CONDITIONS

Av. residence time, sec.	N_2 flow rate, cc./sec.	Time lag, sec. Inlet	Time lag, sec. Outlet	γ-counting rates, counts/min. Initial	γ-counting rates, counts/min. Background
0.40	407.0	0.02	0.13	3.6×10^5	1.4×10^4
0.63	255.0	0.04	0.20	7.5×10^5	3.0×10^4
0.79	203.0	0.05	0.26	5.6×10^5	1.7×10^4
2.36	68.0	0.15	0.77	1.2×10^5	1.2×10^4
3.95	40.7	0.25	1.28	1.6×10^5	1.0×10^4
15.50	10.4	0.96	5.03	3.0×10^5	1.6×10^3

TABLE 2. DECAY RATES FOR PISTON FLOW AND PERFECTLY STIRRED REACTORS

Model	Relative tracer concentrations: Inside reactor	Relative tracer concentrations: At exit	Time interval
Perfectly stirred	$e^{-t/T}$	$e^{-t/T}$	$t \geqq 0$
Piston flow	$1 - t/T$ 0	1 0	$0 \leqq t \leqq T$ $t \geqq T$
Parallel piston and stirred	$(1-x)(1 - t/T) + xe^{-t/T}$ $xe^{-t/T}$	$(1 - x) + xe^{-t/T}$ $xe^{-t/T}$	$0 \leqq t \leqq T$ $t \geqq T$
Series piston and stirred	$1 - t/T$ $x \exp\left[\dfrac{-t + (1-x)T}{xT}\right]$	1 $\exp\left[\dfrac{-t + (1-x)T}{xT}\right]$	$0 \leqq t \leqq (1-x)T$ $t \geqq (1-x)T$

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