JUL 9 1964

University of California

Ernest O. Lawrence Radiation Laboratory

~~NOT FOR PUBLIC RELEASE - OFFICIAL
DISTRIBUTION MAY BE MADE. OFFICIAL
REQUESTS MAY BE FILLED. REPORT
CONTAINS NOTHING OF PATENT INTEREST.
PROCEDURES ON FILE IN RECEIVING
SECTION.~~

Livermore, California

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

MIXING AND CHEMICAL REACTION IN TURBULENT FLOW REACTORS

R. Norris Keeler,* E. E. Petersen and John M. Prausnitz
Department of Chemical Engineering
University of California, Berkeley

ABSTRACT

Mixing and a rapid, second-order irreversible chemical reaction were studied in a turbulent chemical flow reactor, using a point conductivity probe to detect changes in concentration. Using the mathematical theory of Toor, it is shown experimentally that data on the mixing of a passive scalar additive in a non-reactive system may be used accurately to predict the yield of a rapid, irreversible second-order reaction when the hydrodynamics and initial conditions on mixing for the reactive system are identical to those of the non-reactive system. Increasing the bulk average concentration of one reactant is shown to have a strong effect on the overall reaction yield.

* R. N. Keeler is presently at the Physics Department, Lawrence Radiation Laboratory, University of California, Livermore, California.

Background

One of the important problems of chemical engineering is prediction of fractional conversion at the outlet of a turbulent flow reactor in which the reactants are introduced and are allowed to mix and react. This problem may be rephrased in terms of chemical engineering reactor design: Given the hydrodynamic or mixing properties of a flow reactor, and the kinetics of a chemical reaction, calculate the conversion accomplished in the reactor. This problem has been completely solved for a very limited number of cases, but until recently, little experimental or theoretical work had been done on the problem of simultaneous mixing and rapid chemical reaction.

Chemical reactions carried out in turbulent flow reactors can be divided into two limiting cases and one intermediate case. The two limiting cases occur when either hydrodynamics or kinetics completely dominate a combined mixing-reaction system. In the first limiting case the kinetics of the reaction are so rapid that the reaction is complete when mixing is complete. Reactions of this type are commonly called "diffusion-controlled" reactions, since the molecular diffusivity is the only reaction-limiting factor in the fine structure of the flow field. The second limiting case is the situation in which the kinetics of the chemical reaction are so slow that the reactants are completely and thoroughly mixed before any appreciable reaction has taken place. This corresponds to the most frequently encountered case in reactor design. The intermediate case, where both the hydrodynamics of mixing and the kinetics of reaction play a significant part in determining reaction yield, is interesting and becomes important in certain systems such as flame

reactors. In this work we consider the case of the diffusion-controlled reaction.

Previous Work

There have been at least two attempts to find exact solutions for the cases where the conversion in a chemical reactor is affected by the rate at which the reactants mix. In 1957, Danckwerts [1] defined a quantity he called "intensity of segregation", which is a property of all mixing systems. He related this quantity to the conversion in a reacting system with flow characteristics identical to those of the mixing system. Unfortunately he defined the intensity of segregation in such a way that it was not possible to obtain this quantity from a mixing experiment and then use it to predict conversion rates in a chemical reactor.

In two papers published in 1959 and 1962, Toor [2,3] used transformations first proposed by Burke and Schumann [4] to solve mathematically the limiting case of an instantaneous or diffusion-controlled reaction. The fact that the diffusion-controlled chemical reaction and mixing system are mathematically equivalent when flow hydrodynamics are identical has been well known for some time; however, the application of the required transformations to turbulent flow systems required considerable insight and understanding of turbulent mass transfer processes. The solution was presented in such a way that the degree of completion of reaction could be predicted by using data obtained from a mixing experiment carried out under identical hydrodynamic conditions. In order to investigate the applicability of the theory of Toor, both chemical reactions and mixing experiments were carried out in the same turbulent flow reactor.

Experimental Equipment

The flow reactor used in this investigation was originally constructed for the work of Cairns [5]. A schematic of the flow system is shown in Figure 1. The incoming water passed through the flow distributor and into the calming section, a part of the flow development section. The flow distributor had the function of generating a flat velocity profile. The flow then passed the injector, the turbulence producing grid and entered the reactor. The reactor was simply a 2' or 4' section of 2" or 4" i.d. pyrex pipe which terminated in a stainless steel overflow tank. The overflow tank maintained a constant head on the flow system and permitted establishment of a constant flowrate through the system.

For runs in which mixing experiments were conducted, electrolyte solutions were introduced through the surge relief line into the main stream through the hairbrush injection system, consisting of an array of 156 or 37 hypodermic needles. The main stream was tap water supplied from a constant-head tank on the roof of the laboratory. For the reaction runs, where acetic acid solutions were injected into a main stream of ammonium hydroxide solution, the reservoir for the concentrated acetic acid was a hastelloy piston and cylinder injection system; the ammonium hydroxide solutions were stored in a large lead-lined tank mounted in the roof of the laboratory. For all runs, the main stream flow was gravity flow.

In order to produce turbulence of a known character, square mesh wire grids were mounted at the entrance of the pyrex pipes. These grids, 2" and 4" in diameter, had mesh lengths of $1/4"$, $1/8"$, and $1/16"$, with mesh length to wire diameter ratio of 2.6.

Concentration measurements were made by the conductivity method.

This experimental approach is based on the fact that at low electrolyte concentrations, the conductivity of an aqueous solution is directly proportional to the concentration of electrolyte [6]. The unique experimental feature of this work was the development of a microconductivity probe of pyrex-coated platinum which had higher resolution and greater sensitivity than probes used by previous investigators. A full description of their construction is given elsewhere [7]. The platinum tips of the conductivity probes ranged in size from about one-half micron to 25 microns in diameter, and were capable of detecting r.m.s. concentration fluctuations (relative to average concentration) of 4×10^{-6} , expressed as equivalent noise level.

Block diagrams of the overall electronic circuitry are shown in Figure 2. Measurements were made with two circuits, the load resistor and bridge. Load resistors have been used by numerous investigators, while bridge circuitry was used initially by Kramers et al. [8] and more recently by Gibson and Schwarz [9]. The probe consisted of one electrode, the conducting liquid providing the ground, an arrangement first tested by Lawrence [10]. The fluctuations of concentration were measured as r.m.s. voltage fluctuations on a Ballantine Model 320 r.m.s. voltmeter. For the reaction system, a Hewlett-Packard Type 410B voltmeter was used to measure the average product concentration as an average voltage.

Mixing Studies

In turbulence studies it is customary to measure root-mean-square (or mean-square) quantities and to refer these quantities to their initial root-mean-square, mean-square or average value. The concentration at a particular point in space and time can be divided

into a time-average and a fluctuating term:

$$C(x,y,z,t) = \bar{C}(x,y,z) + c(x,y,z,t) \quad (1)$$

The experimental equipment described in the preceding section was designed to measure $\bar{C}$ and $\sqrt{c^2}$ as time-average and fluctuating voltages, which could be interpreted as their concentration equivalents.

There are several ways of describing how well mixed a continuous single liquid-phase solution may be at a given time. The most popular descriptions are the terms "unmixedness" and "goodness of mixing". Unmixedness has been defined as $1 - \sqrt{c^2} / \bar{C}$ and goodness of mixing as $\sqrt{c^2} / \bar{C}$. Because of the direct relationship of mixing and chemical conversion in flow systems such as the one studied in this research, two new terms will be defined:

$$\eta = 1 - \frac{\sqrt{c^2}}{\sqrt{c_0^2}} \equiv \text{fractional completion of mixing}$$

$$F = \frac{\text{mol fraction average product concentration at a point in reactor}}{\text{mol fraction average product concentration at completion of reaction}}$$

$$\equiv \text{fractional completion of reaction}$$

The mol-fraction-average product concentration at completion of reaction is equal to the initial concentration of the reactant present in smallest stoichiometric quantity; i.e., the initial limiting reactant concentration.

The fractional completion of mixing and the fractional completion of reaction both exhibit the same asymptotic behavior; they are both zero at the entrance of the reactor, and in the case of the diffusion-controlled reaction studied in this research, both quantities approach unity as time (or distance down the reactor) approaches

infinity. That this is indeed the case for diffusion-controlled reactions can be seen if one considers such reactions to be instantaneous. If a fluid containing pockets or eddies of reactants were suddenly mixed on a molecular scale, the reaction would be completed at that instant. Thus, for diffusion-controlled reactions, completed mixing implies completed reaction.

Initial Conditions on Mixing

The quantity $\sqrt{c_o^2}$ which appears in the denominator of the second term in the definition of fractional completion of mixing is some initial value of the root-mean-square tracer concentration fluctuations. This quantity can be calculated directly for systems which are one dimensional. In practice, this quantity must be determined experimentally or empirically.

For an ideal, one-dimensional system, such as that shown in Figure 3, a traverse may be drawn in the plane of injection, and the mean-square value of the concentration along this line is given by:

$$\overline{c_o^2} = \frac{\sum C_1^2 \delta_1}{l} \quad (2)$$

where δ_1 = element base length of concentration eddy,

C_1 = concentration of additive in the incoming solution, and

l = length of line traversed.

The term C_1^2 can be taken out of the summation term. It is a constant across all concentration eddies encountered along the line l , since molecular diffusion has not yet had a chance to smear the infinitely steep concentration gradients at the tracer injection point. Now the remaining term, $\sum \delta_1 / l$ is simply equal to the ratio of the injection rate to the total flow rate in volumetric units. Therefore

$$\sqrt{c_o^2} = \sqrt{c_1^2} \sqrt{\frac{I}{G}} = c_1 \sqrt{\frac{I}{G}} \quad (3)$$

where I = injection rate, volumetric units per unit time, and

G = overall flow rate, volumetric units per unit time.

Thus, the initial value of the r.m.s. concentration fluctuations is proportional to the concentration of the tracer in the injected stream, and to the square root of the injection rate. The concentration fluctuations at any point in a one-dimensional flow system show the same dependence on flow rates and tracer concentration as that expressed in Equation (3) for the "initial" concentration fluctuations.

Equation (3) can also be expressed in terms of the average concentration in a one-dimensional turbulent mass transfer system, where $\bar{C}$ is constant, since $\bar{C} = C_1 \frac{I}{G}$,

$$\sqrt{c_o^2} = \bar{C} \sqrt{\frac{G}{I}} \quad (4)$$

Thus it is suitable to normalize the value of r.m.s. concentration fluctuations at a point in the reactor by the average concentration, provided the ratio of G to I remains constant.

Since it is necessary to disturb a flow field to introduce a side stream into it, strong anisotropy of both average local velocity and average concentration exists at the tracer injection point. Therefore, the assumption that concentration fluctuations appear to originate at the tracer injection point with an intensity calculable from Equation (3) or (4) is subject to serious scrutiny. While it is possible to evaluate various theories of mixing using a reduced r.m.s. concentration fluctuation which has been normalized by an "initial" value of r.m.s. concentration fluctuations calculated by Equation (3) or (4), the use of these equations must be modified

when comparison between reacting and mixing systems are made.

Theory of Scalar Decay

Since Toor has established correspondence between mixing and diffusion-controlled reaction, it is of interest to investigate the effects of changing such parameters as Schmidt number, turbulent velocity microscale and viscous dissipation rate upon the rate of scalar decay.

Most theoretical work before 1962 indicated that for the scalar temperature and concentration, which are governed by the diffusion equation

$$\frac{\partial C}{\partial t} + U_1 \frac{\partial C}{\partial x_1} = D \frac{\partial^2 C}{\partial x_1^2}, \quad (5)$$

one could expect a decay law of the form

$$\overline{c^2} = \text{constant} (D\tau)^{-3/2} \quad (6)$$

where D = molecular or thermal diffusivity, and

τ = time.

However, the work of Batchelor [11,12] in 1959 on the spectrum of a scalar in an isotropic turbulent flow field, cast some doubt on the role of molecular or thermal diffusivity in the time decay of scalar quantities in turbulent flow fields. In 1962, Corrsin [13] utilized Batchelor's findings to develop a new theory of scalar decay. For the case of decaying velocity field and a high Schmidt number which was studied in this research, Corrsin derived the expression

$$\frac{dc'}{c'} = \frac{du'}{u'} \frac{2 + \frac{1}{Sc}}{3.9 \left(\frac{L_c}{L}\right)^{2/3} + \frac{2.6}{Re_\lambda} \log Sc} \quad (7)$$

where $u' \equiv \sqrt{\overline{u^2}}$, $c' \equiv \sqrt{\overline{c^2}}$ and $Re_\lambda \equiv \frac{u'\lambda}{\nu}$.

u' = r.m.s. fluctuating velocity component, isotropic turbulent flow field,

c' = r.m.s. fluctuating concentration component,

λ = velocity microscale,

Re_λ = Reynolds number based on λ , and

ν = viscosity.

Equation (7) was derived for R_λ large; R_λ for this research was relatively low. However, it was possible to vary the parameters of Equation (7) to give the first qualitative test of Corrsin's theory.

When Equation (6) is integrated and rearranged, a decay law of the form originally proposed by Gibson [14] is obtained:

$$\frac{\overline{c^2}}{\overline{c_0^2}} = \alpha \left(\frac{x}{M} \right)^a \quad (8)$$

with the constant α dependent on the velocity field, and the constant a defined by the term in brackets on the right-hand side of Equation (6).

Experimental Test of the Theory of Corrsin

In order qualitatively to verify the predictions of Corrsin's equation, a study was made of the effect of varying grid solidity, molecular diffusivity, and ratio of the integral scale of turbulence to the integral scale of velocity on scalar decay.

Figure 4 shows a comparison of the data of Gibson and Schwarz with the data of this research. Using a least-squares fit and rejecting four points, Gibson and Schwarz obtained a value of -1.38 instead of -3/2 for the exponent in Equation (6). The data points plotted in Figure 4 were points taken from runs where the sum of terms in the denominator of Equation (6) was roughly the same as the sum of these terms in the Gibson and Schwarz work. It can be seen that the slopes of the two sets of data are approximately equal, as would be predicted by Equation (6). The data are displaced vertically, which

is probably a consequence of the different grid solidities. Figure 4 shows that within the limits of experimental scatter, there appears to be no difference between data points for the decay of sodium nitrite concentration fluctuations and the decay of zinc sulphate concentration fluctuations. The Schmidt number for the sodium nitrite runs was one-half the Schmidt number for the zinc sulphate runs, as the diffusivity of the two materials differs by a factor of two; no dependence of scalar decay on diffusivity such as that predicted by Equation (5) was found, confirming the negligible effect of changing diffusivity predicted by Equation (6).

In the past, the effect of changing velocity scale upon the rate of scalar decay has been largely ignored, although physical arguments had been offered for the existence of this effect. Hinze [15], in discussing the work of Towle and Sherwood [16] and McCarter et al., argued that "...reduction in the coefficient of eddy diffusion by decrease in the scale of turbulence is often greater than the increase caused by the augmented intensity of turbulence." These remarks were made in connection with a system in which the scalar was introduced as a point source, but the same concepts are obviously applicable to systems in which there are no radial gradients of concentration.

Equation (7) takes this effect into account, as the ratio of concentration scale, L_c , to velocity scale, L , appears in the denominator. In order to verify experimentally the existence of this effect, two experiments were carried out in which the initial conditions on mixing were identical, but the grids used to produce turbulence in the reactor were of different mesh size, although geometrically similar. The data are plotted in Figure 5. The ordinate of Figure 5 was chosen so that the data of Figure 4

($Re_M = 800-2400$) which fortuitously obeyed the " $-3/2$ law" would be a straight line. Scalar decay at an exponential rate less than $-3/2$ would be represented by a family of curves bending toward the abscissa; more rapid decay rates generate a family of curves bending toward the ordinate. It can be seen that the curve for grid No. 2 has a slope decreasing continuously toward the abscissa, indicating a value of a , the decay constant of Equation (8), less than $3/2$. This is in qualitative agreement with the prediction of Corrsin's Equation (7); viz., decreasing the scale of turbulence decreases the rate of scalar decay.

Theory of Diffusion-controlled Reactions

The simplest imaginable turbulent flow reactor is a box or tank in which the turbulent velocity field is isotropic and decaying, or isotropic and stationary, and the reactants are homogeneously dispersed throughout the reactor. In such a system, there are no spatial gradients of average velocity or average concentration, and the average and root-mean-square quantities vary only with time. Such a system would be the easiest to treat from the theoretical standpoint.

The turbulent flow reactor used in this research was a one-dimensional system, generating an isotropic decaying turbulent flow field, decaying down the length of the reactor. In the previously described mixing runs, the hairbrush injector distributed the tracer homogeneously into the mainstream as it entered the reactor, and in the reaction runs, one reactant was dispersed homogeneously into the other. Thus, a one-dimensional system was achieved.

Beek and Miller [17] were apparently the first to point out the importance of the magnitude of the mean-square concentration fluctuations in mixing. Although the theories upon which their work was

based have been replaced by the more recent work of Batchelor [11] and Corrsin [13], the authors made the very important point that the yield of a chemical reaction in a flow reactor is related to the decay of concentration fluctuations of a tracer introduced under identical flow conditions. They suggested that concentration fluctuation data can be used to scale turbulent flow reactors, but offered no details as to how this could be done quantitatively.

In the two previously mentioned papers of Toor [2,3], the problem of conversion in laminar and turbulent flow systems was studied. Toor took the basic transformations of Burke and Schumann [4] and applied them to turbulent flow systems.

For turbulent flow systems, the approach of Toor revealed that in diffusion-controlled reactions, solutions will be found in terms of the parameter $\sqrt{c^2}/\sqrt{c_0^2}$, as suggested by Beek and Miller. Making the assumption that the fluctuating component of concentration is normally distributed around the mean, Toor was able to use a relation of Hawthorne et al. [18] to obtain the solution for the yield of a diffusion-controlled reaction in terms of the equivalent mixing problem even when reactants were present in non-stoichiometric amounts. For reactants present in stoichiometric amounts

$$F = \eta \quad (9)$$

where F = fractional completion of reaction, and

η = fractional completion of mixing.

For the case where the reactants are not present in stoichiometric amounts, a more complicated expression is obtained:

$$F = 1 + (\beta - 1) \{1 + g[\gamma(1 - \eta)]\} \quad (10)$$

where β = the initial ratio of reactant concentrations, and

γ = constant depending on β and the initial value of reactant

concentration fluctuations, and

$$g(x) = \frac{x}{\sqrt{2}} \operatorname{ierfc} \left(\frac{1}{\sqrt{2} x} \right).$$

Reaction Studies

In order to test the theory of Toor, the reaction of ammonium hydroxide and acetic acid was carried out in the turbulent flow reactor. This reaction possesses several advantages:

- (1) It is an ionic reaction, very rapid, and hence can be considered diffusion controlled under conditions where the reactants mix and react.
- (2) The reaction is irreversible for all practical purposes.
- (3) The heat of reaction is low, and may be considered negligible for this work.
- (4) The molecular diffusivities of the reactants are almost equal: ($D_A \approx D_B$, one of the assumptions of Toor).
- (5) The electrical conductivity of the products is much greater than the electrical conductivity of the reactants.

The fractional completion of reaction was measured for reacting mixtures in which the ratios of ammonium hydroxide to acetic acid were 1:1, 2:1, and 10:1, respectively. For every reaction run, there was a mixing run carried out under the same conditions of geometry, hydrodynamic flow, and introduction of side stream.

Figure 6 shows the result of a single mixing run and a single reaction run which were carried out at a mesh Reynolds number, Re_M , of 1200, based on the mesh length of the fine grid used to generate the turbulent velocity field. The reactants were introduced in their stoichiometric ratio, i.e., $\beta = 1$. The r.m.s. concentration

fluctuations reduced by their apparent initial value, and the fractional completion of reaction are plotted against distance down the reactor. As expected, the reaction curve is a mirror image of the mixing curve, confirming the prediction by Toor that

$$F = \eta = 1 - \frac{\sqrt{c^2}}{\sqrt{c_0^2}}, \quad (11)$$

i.e., the fractional completion of reaction is exactly equal to the fractional completion of mixing when $\beta = 1$.

In order to examine fully the effect of changing β , the results of the runs with $\beta = 1$, $\beta = 2$, and $\beta = 10$ are plotted against the predictions of Toor given in Figure 6 of reference [3]. Agreement with theory is good.

Every point plotted on Figures 7, 8, and 9 is a point for which mixing and reaction data were taken under identical hydrodynamic conditions. The addition of one reactant in excess exerts a strong effect upon the fractional completion of reaction, forcing the reaction to completion long before mixing is complete. This suggests that when one of the reactants is inexpensive and easily recovered, it would be feasible to add this reactant in great excess, decreasing the reactor length required for a desired fractional completion of reaction. This technique would be particularly useful for reactions where almost complete conversion is desired.

Experimental Initial Mixing Conditions

The initial conditions on mixing for the injection system described in this investigation were such that $\sqrt{c_0^2} \approx \bar{c}$. This means that almost 90% of mixing was completed by the time the distribution of tracer became homogeneous. It was observed that considerable mixing took place just before and around the grid, as would be

characteristic of turbulent jets. Extrapolation of mixing runs revealed an initial value of effective concentration fluctuations of about one-tenth of the value calculated from Equation (3). Measurements taken with the conductivity probe close to the grid revealed that when the distribution of tracer became homogeneous, at a distance of about one centimeter downstream from the grid, the value of $\sqrt{c^2}$ was approximately equal to the average concentration. The value of $\overline{c_o^2} \approx \bar{c}$ was used to normalize r.m.s. concentration fluctuations in calculating the value of fractional completion of mixing for Figures 7, 8, and 9.

In an anisotropic system, it is possible for considerable mixing to take place before homogeneity of tracer distribution is achieved, by the criterion $\eta = 1 - \sqrt{c^2} / \sqrt{c_o^2}$, if $\sqrt{c_o^2}$ is calculated from Equation (3). In a system where the reactants are present in near stoichiometric amounts, and one reactant is introduced in a stream which constitutes only a very small volumetric fraction of the overall flow, the rate of approach to completion of reaction will be very slow until the two reactant streams are homogeneously mixed. Toor realized this, and specified that $\sqrt{c_o^2}$ must be referred to a point where the distribution of tracer is homogeneous, and further, the fractional completion of reaction must be set to zero at this point. Unfortunately, this constraint makes it impossible to predict the overall yield of a chemical reaction from mixing data alone. In this work, this difficulty was circumvented by assuming that very little reaction took place between the injection needles and the region just downstream of the grid -- a justified assumption under the conditions discussed previously. Nevertheless, it is felt that this assumption and the error in assigning an effective initial value

of r.m.s. concentration fluctuations are responsible for the scatter of the data plotted on Figures 7, 8, and 9:

Conclusions

The decay of concentration fluctuations in an isotropic turbulent flow field was studied with a high-resolution electrical conductivity probe. The decay rate was found to agree, at least qualitatively, with the most recent theories of Corrsin. The model of Toor was used to predict the conversion of a diffusion-controlled chemical reaction in a turbulent plug flow reactor. Further advances in the field now depend upon resolution of the problem of mixing and chemical reactions in anisotropic systems such as turbulent jets.

Acknowledgments

The authors would like to express their appreciation to Mr. Maarten van Gent for assistance in constructing the micro-conductivity probes used in this work and to Mr. Frank Upham for helpful advice regarding electronic circuitry. One of us (R.N.K.) would like to acknowledge the financial support of the Standard Oil Company of California, the National Science Foundation, and the Woodrow Wilson Foundation in the form of fellowships and research grants.

LITERATURE CITED

- [1] Danckwerts, P. V., "The Effect of Incomplete Mixing on Homogeneous Reactions," Chemical Reaction Engineering, 12th Meeting, European Federation of Chemical Engineering, Amsterdam (1957).
- [2] Toor, H. L., and S. H. Chiang, A.I.Ch.E. Journal, 5, 339 (1959).
- [3] Toor, H. L., ibid., 8, 70 (1962).
- [4] Burke, S. P., and T. E. W. Schumann, Ind. Eng. Chem., 20, 998 (1928).
- [5] Cairns, E. J., Ph.D. Thesis, University of California (1959).
- [6] International Critical Tables, Vol. 6, p. 238, McGraw-Hill Book Company, Inc., New York, N.Y. (1929).
- [7] Keeler, R. N., Ph.D. Thesis, University of California, Berkeley (1964).
- [8] Kramers, H., G. M. Baars and W. H. Knoll, Chem. Eng. Sci., 2, 35 (1953).
- [9] Gibson, C. H., and W. H. Schwarz, J. Fluid Mech., 16, 365 (1963).
- [10] Lawrence, W. J., unpublished work, University of California, Berkeley (1958).
- [11] Batchelor, G. K., J. Fluid Mech., 5, 113 (1959).
- [12] Batchelor, G. K., I. D. Howells and A. A. Townsend, ibid., p. 134.
- [13] Corrsin, S., "The Isotropic Turbulent Mixer, II. Arbitrary Schmidt Number." Part of a lecture presented at the 55th Annual Meeting, A.I.Ch.E., Chicago, Ill., 2-6 Dec., 1962.
- [14] Gibson, C. H., Ph.D. Thesis, Stanford University (1962).
- [15] Hinze, J. O., "Turbulence," p. 342, McGraw-Hill Book Company, Inc., New York, N.Y. (1959).
- [16] Towle, W. L., and T. K. Sherwood, Ind. Eng. Chem., 31, 457 (1939).
- [17] Beek, J., Jr., and R. S. Miller, Chem. Eng. Progr. Symp. Ser., No. 25, 55, 23 (1959).
- [18] Hawthorne, W. R., D. S. Weddell and H. C. Hottel, Third Symposium on Combustion and Flame and Explosion Phenomena, p. 266, Williams and Wilkins, Baltimore (1949). Paper presented at A.I.Ch.E. Meeting, Chicago, Ill., Dec. 1962.

NOTATION

- a - constant in decay law
- a,b - stoichiometric coefficients for the diffusion-controlled reaction $aA + bB \rightarrow \text{products}$
- c - fluctuating component of concentration
- c' - root-mean-square concentration fluctuations, $\sqrt{c'^2}$
- $\sqrt{c_o^2}$ - initial root-mean-square concentration fluctuations, calculated or effective
- C - concentration
- C_1 - concentration of tracer in injected stream
- $\bar{C}$ - time average concentration
- $\bar{C}_1$ - space (and time) average concentration of tracer
- d - mesh diameter
- D - molecular or thermal diffusivity
- F - fractional completion of reaction
- G - overall flowrate through reactor
- I - tracer injection rate
- ℓ - length of traverse line
- L - integral scale of velocity
- L_c - integral scale of concentration
- M - mesh length
- R - resistance, ohms
- Re_M - mesh Reynolds number, $\bar{U}M/\nu$
- Re_λ - turbulence Reynolds number, $u'\lambda/\nu$
- Sc - Schmidt number, ν/D
- t - time
- u - fluctuating velocity component
- u' - root-mean-square concentration fluctuation, $\sqrt{u'^2}$
- U - velocity

- $\bar{U}$ - average velocity
- $\overline{v^2}$ - r.m.s. voltage
- $\bar{V}$ - average voltage
- x - distance
- x, y, z - space coordinates

Greek Letters

- α - pre-exponential factor in decay equation
- β - stoichiometric parameter, $a\bar{C}/b\bar{C}$
- γ - initial value of r.m.s. reactant concentration
- δ - increment of distance through concentration eddy
- η - fractional completion of mixing, $1 - \frac{\sqrt{c^2}}{\sqrt{c_o^2}}$
- λ - microscale of velocity fluctuations
- ν - kinematic viscosity
- τ - time

Subscripts

- o - initial value
- l - tracer
- c - concentration
- M - mesh
- λ - microscale

FIGURE CAPTIONSFigure

1. Flow system.
2. Electronic circuitry. (a) Probe and auxiliary circuitry.
(b) Overall Q-circuitry.
3. Initial conditions on injection of tracer. (a) Physical configuration at injection system. (b) Concentration distribution along a line in the plane $t = t_0$ (after Corrsin).
4. Decay of concentration fluctuations behind a square-mesh grid.
5. Effect of changing turbulence scale on rate of decay of concentration fluctuations.
6. Rate of approach to completion of reaction with decay of concentration fluctuations.
7. Theoretical predictions and experimental results, $\beta = 1$.
8. Theoretical predictions and experimental results, $\beta = 2$.
9. Theoretical predictions and experimental results, $\beta = 10$.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

GLL-641-210

Figure 7

GLL-641-209

Figure 8

GLL-641-208

Figure 9