Chemical Reaction in the Turbulent Wake of a Cylinder

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A rapid, irreversible, second-order chemical reaction in the turbulent wake of a cylinder was studied. A hypothesis relating the correlation of the fluctuating concentrations directly to the mean concentrations of the reacting species was found applicable, at least, when tested for a pseudo first-order reaction system.

Experimentally, the mean concentration profiles of a colored reactant were obtained with and without reaction by time-exposure photographs. Since the reactant changed from colored to colorless during reaction, the rate of reaction could be determined directly from the rate of color disappearance.

Both the reaction rate and the turbulent intensity divided by the half-width of the wake were found to be inversely proportional to the distance from the source for *x/d* values from 35 to 180. This indicates a direct relationship between them. Consequently, the concentration correlation function also may be related directly to the turbulent intensity divided by the wake half-width.

In addition, it is shown that the rate of reaction is faster in the center of the wake than it is toward the edges.

The relationship of turbulence parameters to the rate of chemical reaction is important not only in the design of chemical reactors, but also in studies of reactions in a hypersonic wake and in supersonic combustion (*1* to *3*). Although many investigations of turbulent transport processes with chemical reaction have been reported, these studies do not relate the rate of reaction to the parameters of the turbulent flow (*4* to *6*). Since experimental data on this relationship are lacking, unjustified simplifying assumptions often have been made to analyze these processes.

Analyses of reaction in turbulent flow can be divided into two broad categories: fundamental (*7, 8*) and phenomenological (*4, 5*). Because of the complexity of the problem, the fundamental work has been restricted to isotropic (macroscopically) homogeneous velocity and concentration fields. Even with this simplification, the mean concentration equation is not exactly soluble for any but a first-order reaction. Since most of the processes of interest involve nonhomogeneous concentrations of reactants in a turbulent shear flow, an analysis of such a process from first principles is difficult if not impossible. Therefore, in the analyses of these problems some phenomenological hypotheses often are made. These hypotheses relate the correlation of the fluctuating quantities to the mean quantities by empirically determined functions.

The general problem of chemical reaction in turbulent flow may be stated as follows. Given the properties of a turbulent flow, the concentrations of reacting species for some time and position, and the kinetics of the reaction, determine the concentrations of the species at any time and position. The solution of this problem from the phenomenological point of view requires a hypothesis relating the correlation of the fluctuating and mean components of the reactant concentrations. The correlation function relating these quantities depends on turbulence parameters such as the turbulent intensity and length scale. An important objective of this research was to examine the dependence of the correlation function on the turbulent flow.

The work presented here is the culmination of that begun by Kiser and Hoelscher (*9, 10*) and continued by Sparks and Hoelscher (*11* to *13*). These investigators constructed a closed-circuit water tunnel and measured some important parameters in the turbulent wake of a cylinder. This well-defined experimental system served as the primary tool for this study of chemical reaction in a turbulent wake.

In this paper an experimental study of rapid, irreversible, second-order reactions in the turbulent wake of a cylinder is described. This system is relatively simple, yet it contains the characteristics of a more general system, that is, inhomogeneity of both concentration and velocity fields. The mathematical description of this two-dimensional system is based on the Townsend model (*14*) of the turbulent wake. First, the turbulence properties such as the turbulent intensity are assumed uniform within the fully developed wake. This assumption is equivalent to restricting the inhomogeneity of the turbulent field to the direction of flow. Second, the mean velocity in the wake is considered constant in the equations for the conservation of species.

The experiments performed during this work were designed to produce a pseudo first-order reaction system when one of the reactants was supplied in great excess. This simplifies the process and its mathematical description, yet retains the essence of the problem. The main objective of the experiments was to obtain the mean concentration of one reactant as a function of position with and without another reactant present. In addition experiments were made to compare the reaction process occurring within the turbulent core (the region of the wake which at any time is completely turbulent) with the reaction process occurring throughout the wake.

MATHEMATICAL ANALYSIS

For the particular problem of a second-order, irreversible reaction in a two-dimensional, turbulent wake, the

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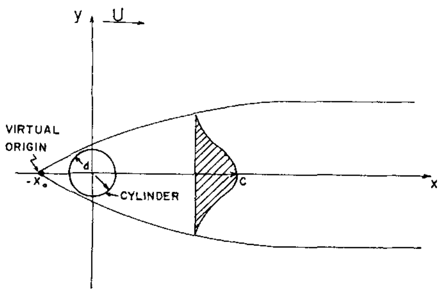

Fig. 1. Schematic of source system.

general equation for the conservation of species reduces to

$$U\frac{\partial C_1}{\partial x}+\frac{\partial(\overline{v'C_1'})}{\partial y}=D\frac{\partial^2 C_1}{\partial y^2}-k\,[C_1C_2+\overline{C_1'C_2'}] \quad (1)$$

where U, C_1, and C_2 are the time-averaged velocity and concentrations of species 1 and 2. The over-barred quantities are the averages of the fluctuating quantities. In this equation the usual assumptions are made based on an order-of-magnitude analysis (*14*). Generally, Equation (1) is coupled with the conservation equation written for species 2.

A further approximation, especially appropriate in liquid systems, is that

$$\frac{\partial(\overline{v'C_1'})}{\partial y}>>D\frac{\partial^2 C_1}{\partial y^2} \quad (2)$$

In addition the velocity-concentration correlation may be related to the mean concentration gradient by a turbulent transport function ϵ defined by

$$\epsilon \equiv -\frac{\overline{v'C_1'}}{\partial C_1/\partial y} \quad (3)$$

This relationship has been shown to be useful for free shear flows. The combination of Equations (1), (2), and (3) yields

$$U\frac{\partial C_1}{\partial x}=\frac{\partial}{\partial y}\left(\epsilon\frac{\partial C_1}{\partial y}\right)-k\,[C_1C_2+\overline{C_1'C_2'}] \quad (4)$$

Now the hypothesis is made that the correlation of the fluctuating components of the concentrations is directly proportional to the mean concentrations times a function of position

$$\overline{C_1'C_2'}=\phi(x)\,C_1C_2 \quad (5)$$

For a pseudo first-order reaction system, the mean concentration of species 2 may be considered constant. Furthermore, the velocity in the wake will be taken as constant; this is a good assumption provided that the velocities at the center and edges of the wake differ by less than 15%. Under these conditions Equation (4) may be written as

$$\frac{\partial C_1}{\partial x}=\frac{\partial}{\partial y}\left(\frac{\epsilon}{U}\frac{\partial C_1}{\partial y}\right)-\frac{kC_2}{U}\,[1+\phi(x)]C_1;$$

$$x+x_o>0, \quad |y|<\infty \quad (6)$$

with the boundary conditions

$$C_1=\frac{\partial C_1}{\partial y}=0; \quad |y|\rightarrow\infty$$

$$\frac{S}{2}=\lim_{\lambda\rightarrow -x_o}\int_o^{\mu}\int_{-x_o}^{\lambda}\left[U\frac{\partial C_1}{\partial x}-\frac{\partial}{\partial y}\left(\epsilon\frac{\partial C_1}{\partial y}\right)+kC_2\,(1+\phi)C_1\right](dx)(dy)$$

As shown in Figure 1, the constant source flux S is considered to emanate from the virtual origin $-x_o$.

If one makes the transformation

$$C_1(x,y)=\Psi(\xi,y)\;e^{-\int\Gamma/U\,dx} \quad (7)$$

where

$$\Gamma=kC_2\,[1+\phi]; \quad \xi=\frac{x+x_o}{d}$$

Equation (6) becomes

$$\frac{\partial\Psi}{\partial\xi}=\frac{\partial}{\partial y}\left(\frac{\epsilon d}{U}\frac{\partial\Psi}{\partial y}\right); \quad \xi>0,\ |y|<\infty \quad (8)$$

with the boundary conditions

$$\Psi=\frac{\partial\Psi}{\partial y}=0; \quad |y|\rightarrow\infty$$

$$\frac{S}{2U}=\lim_{\lambda'\rightarrow 0}\int_o^{\mu}\Psi(\lambda',y)\,dy$$

By seeking groups which are invariant to scale transformations, one finds that the solution of Equation (6) may be written as

$$\Psi(\xi,\eta)=\xi^{-1/2}f(\eta) \quad (9)$$

where

$$\eta=y\,\xi^{-1/2}$$

This analysis restricts ϵ to be a function of η alone.

In the new variables, Equation (8) becomes

$$\frac{d}{d\eta}\left(\frac{2\epsilon d}{U}\frac{df}{d\eta}\right)=-\left(\eta\frac{df}{d\eta}+f\right)$$

From the solution of this equation with the boundary conditions, one can write the solution of Equation (9) in the form

$$\Psi(\xi,\eta)=\frac{S\,\xi^{-1/2}}{2U}\;\frac{e^{-\int\frac{U}{2\epsilon d}\eta\,d\eta}}{\int_o^{\infty}e^{-\int\frac{U}{2\epsilon d}\eta\,d\eta}\,d\eta} \quad (10)$$

When Equation (10) is substituted into Equation (7)

$$\frac{C_1(x,y)}{C_M(\xi)}=\exp-\int\frac{U}{2\epsilon d}\eta\,d\eta \quad (11)$$

where

$$C_M(\xi)=C_1(x,0)=\frac{S\,\xi^{-1/2}}{2U}\;\frac{e^{-\int\frac{\Gamma}{U}dx}}{\int_o^{\infty}e^{-\int\frac{U}{2\epsilon d}\eta\,d\eta}\,d\eta}$$

For $\epsilon=\epsilon_o$

$$C_1/C_M=\exp-\frac{Ud}{4\epsilon_o}\left[\frac{y^2}{d(x+x_o)}\right] \quad (12)$$

for $\epsilon=\epsilon_o(1+\omega\eta^2)^{-1}$

$$C_1/C_M=\exp-\frac{Ud}{4\epsilon_o}\left\{\frac{y^2}{d(x+x_o)}+\frac{\omega y^4}{4[d(x+x_o)]^2}\right\} \quad (13)$$

If Equation (6) is integrated over all y, one gets the normalized reaction rate

$$\frac{U}{C_2\hat{C}_1}\frac{d\hat{C}_1}{dx}=-k(1+\phi) \quad (14)$$

where

$$\hat{C}_1 = \int_{-\infty}^{\infty} C_1\, dy$$

is the quantity of species 1 per unit distance. It is evident that the correlation function ϕ may be computed from the area under the concentration profiles.

EXPERIMENTAL STUDIES

General

The experimental objectives of this research were (1) to test the validity of the correlation hypothesis; (2) to find the dependence of the correlation function ϕ on the distance x; (3) to ascertain if any difference exists between the rate of reaction in the turbulent core and the rate toward the edges of the wake.

The experiments were performed in a closed-circuit water tunnel (*10* to *12*). In this system one reactant was introduced into the wake of a circular cylinder, while the other was circulated in the mainstream of the tunnel. The reactant was introduced into the wake either from the wake-producing cylinder, itself, or from a line source downstream from the wake-producing cylinder. Since one of the reactants was darkly colored, while the other reactant and products were colorless, the rate of reaction was determined from the rate of the disappearance of color. Consequently, the principal data, the mean concentration profiles, could be obtained by time-exposure photographs of the colored reactant.

Equipment

The water tunnel shown in Figure 2 was used in this study. This unit was originally built by Kiser (*9*), modified by Sparks (*11*), and then modified again for this work. Except for the test and riser sections, the tunnel was constructed of brass. The screen section, consisting of eight brass screens varying from 13 to 100 mesh, was designed to produce a uniform velocity profile and to cause the turbulent fluctuations to decay rapidly (*12*). The clear plastic test section, immediately following the screens, held the 1/4-, 3/16-, and 1/8-in. wake-producing cylinders. Both the screen and test sections were 8 in. sq. The Reynolds number for the experiments was varied from 1,300 to 2,600, based on the cylinder diameter.

The line sources, with outer diameters of 0.032 and 0.047 in., were made from stainless steel tubing (*15*). Actually, the "line" was simulated by a series of fifty holes spread over 5 in. In the smallest diameter tube the holes were 0.004 to 0.006 in. in diameter, while in the larger one they were 0.006 in. in diameter.

Photographic Methods

The mean concentration profiles of the darkly colored reactant were obtained from time-exposure photographs with the aid of a microdensitometer. An example of a microdensitometer tracing is shown in Figure 3. The film density as recorded on the tracing was directly proportional to the concentration provided the density measurements were made along the linear portion of the characteristic curve and the light source was effectively monochromatic. The latter was produced by a combination of du Pont "Cronar" Ortho A film and a No. 21 Wratten filter. A photograph of a dark blue reactant with this combination yielded an extremely high contrast negative. From a calibration curve (*15*) the density from the tracing could be converted into units of concentration.

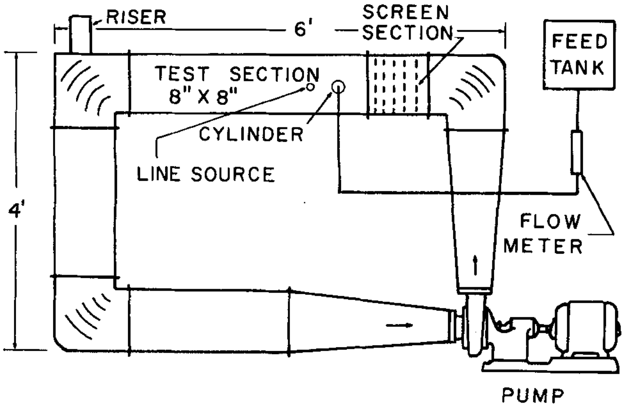

Fig. 2. Water tunnel.

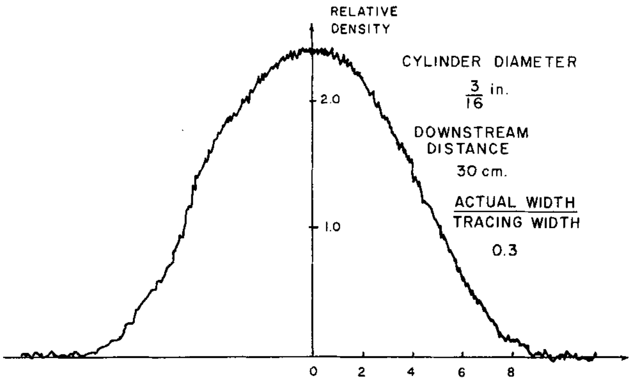

Fig. 3. Microdensitometer tracing.

Reaction Requirements

The reactions used in this work had to fulfill the following requirements: (1) produce a rapid, irreversible, second-order reaction in aqueous solution; (2) involve one darkly colored reactant (without intermediate colors) and one colorless reactant; (3) form colorless and soluble products.

Several reactions of iodine in the presence of starch satisfy all these conditions. Those used in this work were the reactions with sodium hydroxide and sodium thiosulfate:

1. $I_3^- + OH^- = 2I^- + HIO$

$(3HIO + 3OH^- = 2I^- + IO_3^- + H_2O)$

2. $I_3^- + 2S_2O_3^{=} = 2I^- + S_4O_6^{=}$

Procedures and Techniques

In order that the experiments would correspond to the mathematical analysis, the concentration of one reactant had to be made much greater than that of the other. It was found that the iodine-hydroxide reaction produced a conveniently measurable rate of reaction under the conditions imposed. As a comparison, some experiments were made with an iodine-thiosulfate reaction in which the concentrations of both reactants were of the same order of magnitude. In all experiments the temperature was kept at 25°C.

The iodine solution was introduced from the wake-producing cylinders in some experiments and from the line sources in others. When used, line sources were placed either thirty-two or forty-seven diameters downstream from a ¼-in cylinder. As long as the data were obtained 5 to 15 cm. from the line source, the transport and reaction processes essentially were restricted to the turbulent core. The disturbances from line sources of different diameters were compared.

Because the experimental system was not exactly two-dimentional, experiments were performed to determine the significance of this error. In these experiments different ratios of the field depth (the width over which the iodine was introduced) to the cylinder diameter were used. Both the field depth and the cylinder diameter were changed independently.

The photographs of the reactant wake do not relate density to the concentration, but instead relate the density to the concentration times the field depth of the reactant. If end effects are negligible in the system, the field depth will cancel out of any dimensionless quantities.

An additional source of error occurred as a result of the color loss from dilution even without reaction. From a comparison of results with and without reaction, the error from color loss could be compensated for (*15*).

RESULTS AND DISCUSSION

Concentration Profiles*

The mean concentration profiles developed from the 1/4-, 3/16-, and 1/8-in. cylinders, with and without the iodine-hydroxide reaction, were superposable to each other (Figure 4). These concentration profiles indicate that the hypothesis relating the correlation of the fluctuating concentrations to the mean concentrations [Equation (5)] is reasonable. Further, that these profiles are superposable demonstrates the invariance of the hydroxide concentration in the wake. The ratio of the virtual origin to cylinder diameter x_o/d is approximately 55. The profile in Figure 4, taken for values of x/d from 35 to 180, corresponds very closely to that of Fage and Faulkner (*16*) and to that of Sparks and Hoelscher (*12*); it is slightly different from that of Townsend (*14*) taken at an x/d of 500. At these x/d values the velocity profiles also differ slightly (*14, 16*).

Even in the iodine-thiosulfate studies in which both reactants vary across the wake, the similarity of profiles still exists. This is shown in Figure 5. Hence, the correlation hypothesis still may be valid. Although the profiles of both reaction systems are similar, they differ noticeably from each other.

The concentration profiles developed from the line source in the wake of a ¼-in. cylinder are shown in Figure 6. Again, with or without the iodine-hydroxide reaction, the profiles are superposable. For the line sources at thirty-two and forty-seven diameters downstream from the cylinder, the x_o/d ratios are approximately −33 and −51, respectively. A comparison of profiles from the cylinder and line sources shows that the shapes are different, especially in the tails. While the line source profiles are represented accurately by a Gaussian distribution, Equation (12), the cylinder profiles are fitted better by Equation (13). The difference may result from the intermittent character of the wake flow.

Rate of Reaction

The quantity of species 1 per unit distance $\hat{C}_1$ was plotted as a function of distance x. An example of this is shown in Figure 7. From such graphs the normalized rate of reaction was determined as a function of distance. Since log $\hat{C}_1$ is a linear function of log x, the normalized rate of reaction is inversely proportional to distance

$$\frac{U}{C_2 \hat{C}_1} \frac{d\hat{C}_1}{dx} = -k\,[1+\phi(x)] = -\frac{A}{x} \qquad (15)$$

The reaction parameter A for the iodine-hydroxide reaction is shown as a function of the hydroxide concentration C_2 in Figure 8. The ratio of the percentage change in the reaction parameter to the percentage change in the hydroxide concentration is only 0.07.

The slopes of the A — ln C_2 relationships are nearly the same for the cylinder and the line source, but the value of A and thus the rate of reaction is 20% greater for the line source. The difference might be attributed to the turbulence properties in the wake, the line source disturbance, or both. Based on the data of Eskinazi (*17*), the turbulent intensity difference created by the line source in this turbulent shear flow should have decayed to approximately 0.01, 5 cm. downstream from the line source. Furthermore, the normalized rates of reaction from line sources of different diameter were found to be nearly identical. Since the line source disturbance does not

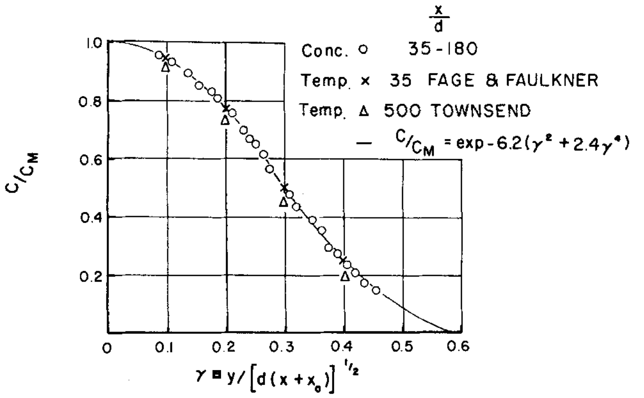

Fig. 4. Mean concentration profiles (cylinder, I_3^- + OH^-).

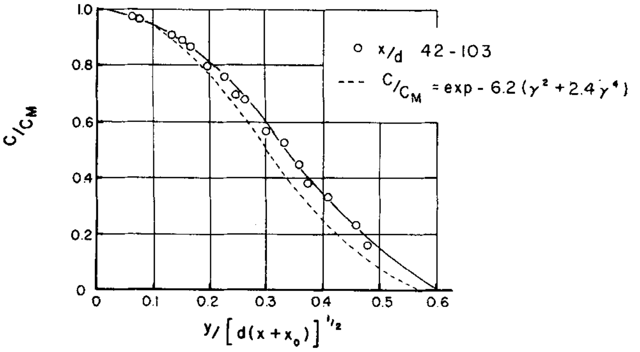

Fig. 5. Mean concentration profiles (cylinder, I_3^- + $S_2O_3^=$).

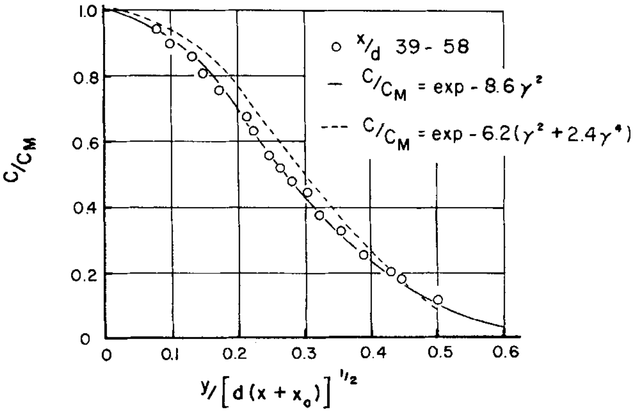

Fig. 6. Mean concentration profiles (line source, I_3^- + OH^-).

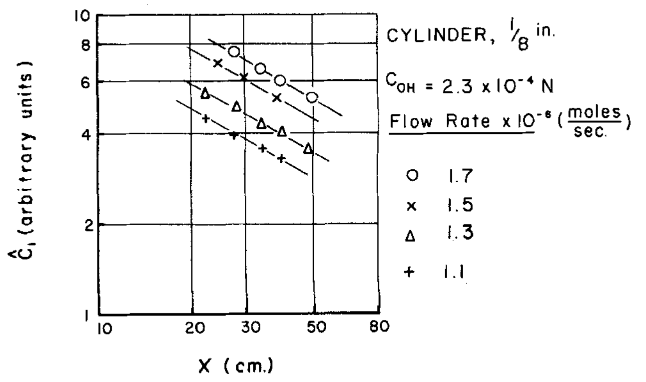

Fig. 7. Quantity of species 1 vs. distance.

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[*] The data upon which the analysis is based are presented in reference *15*.

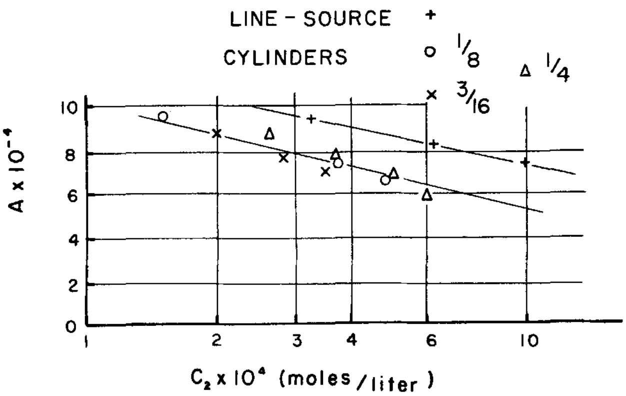

Fig. 8. Reaction parameter vs. hydroxide concentration.

account for the difference in the values of A, the difference is caused probably by the variation of the turbulence properties across the wake. As a consequence of this variation, the rate of reaction is faster in the center of the wake than at the edges.

The errors in the data due to end effects were less than 5%. The corrections for color loss from dilution were at most 3, 6, and 10% for the 1/8-, 3/16-, and 1/4-in. cylinders, respectively.

Correlation Function

An examination of the properties of the concentration correlation function ϕ and the reaction system indicates that the function varies from -1 to 0 (*4*). Although a numerical value relating ϕ and x is desirable, the rate coefficient k has not been measured. Provided the experiments described in this work are two-dimensional, the rate coefficient is the same as that for the microscopically homogeneous reaction system.

When the variation of the normalized rate of reaction with distance is compared to the variation of turbulence parameters with distance, a relationship seems indicated. For x/d values of 35 to 180, the turbulent intensity $(\overline{u'^2})^{1/2}/U$ divided by the half-width of the wake is inversely proportional to the distance downstream (*15, 17, 18*). Since the normalized rate of reaction is also inversely proportional to distance downstream [Equation (15)], the following equation is suggested:

$$\frac{U}{C_2 \hat{C}_1}\,\frac{d\hat{C}_1}{dx} = -k\,[1 + \phi(x)] = K\,\frac{(\overline{u'^2})^{1/2}/U}{b_{1/2}}$$

Thus, the normalized rate of reaction and the concentration correlation function ϕ may be related linearily to the turbulent intensity divided by the half-width.

The relationship between the normalized rate of reaction and these turbulence parameters may be rationalized physically: An increase in the turbulent intensity increases the rate of reaction, but an increase in the length scale decreases the rate of reaction.

CONCLUSIONS

Experiments were performed with a rapid, second-order chemical reaction in the turbulent wake of a cylinder. A hypothesis relating the correlation of the fluctuating concentrations directly to the mean concentrations of the reactants was found applicable, at least when tested by a pseudo first-order reaction system.

A direct relationship is indicated between the rate of reaction and the turbulent intensity divided by the half-width of the wake, since both vary inversely with distance for x/d values of 35 to 180. Therefore, the concentration correlation function ϕ also may be linearly related to the turbulent intensity divided by the half-width.

The rate of reaction is faster in the center of the wake than it is toward the edges.

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NOTATION

A = reaction parameter
$b_{1/2}$ = half-width of the velocity profile of the turbulent wake
C_i = mean concentration of species, = 1, 2
C_i' = fluctuating component of concentration, = 1, 2
$\hat{C}_1$ = quantity of species 1 per unit distance x
C_M = scaling function
D = molecular diffusivity
d = cylinder diameter
f = dimensionless dependent variable
k = reaction rate coefficient
K = proportionality constant
u' = fluctuating component of velocity in x direction
U = free stream velocity
v' = fluctuating component of velocity in y direction
x = direction of flow
x_o = virtual origin
y = direction perpendicular to flow

Greek Letters

Γ = coefficient of the reaction term
γ = dimensionless independent variable
ϵ = turbulent transport coefficient
η = dimensionless independent variable
ξ = dimensionless independent variable
ϕ = concentration correlation function
Ψ = transformed mean concentration variable
ω = constant

Subscript

i = species 1 or 2

Superscripts

$\overline{}$ = mean, time-averaged
$'$ = fluctuation, unless otherwise noted

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Laminar Dispersion in Capillaries:

Part I. Mathematical Analysis

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Since 1953, when G. I. Taylor first considered the problem, numerous studies of the miscible displacement of fluids in capillaries have produced several approximate mathematical solutions which are purported to be valid under different conditions. Their form and ranges of applicability have been in conflict to some extent, since no exact solution is available to check these expressions.

This study has resulted in exact numerical solutions to this problem with both axial and radial molecular diffusion accounted for. The range of parameters investigated is wide enough for comparison with all known analytical and empirical results and covers τ from 0.01 to 30 and N_{Pe} from 1 to 23,000. It is shown that for sufficiently large values of τ the Taylor-Aris theory is valid and thus results for all τ and N_{Pe} of any practical interest are now known.

Axial molecular diffusion is significant at lower values of the Peclet number but the magnitude of N_{Pe} at which this occurs depends on the value of τ. In general, axial molecular diffusion is important for Peclet numbers less than about 100.

Present results show that there is no justification for Bailey and Gogarty's empirical modification which yields an exponent of 0.541 rather than 0.50 for τ in Equations (35) and (36). ***Also, for the system studied here, no justification was found for the conjecture of Bournia et al. that Aris' low N_{Pe} modification may not account for axial diffusion properly.***

Simple expressions given by Equations (48) and (50) were developed empirically and they ***give with good accuracy the average concentration distribution over wider ranges of N_{Pe} and τ than previously reported expressions.***

In 1953, G. I. Taylor (*11* to *13*) discussed the rather complex problem of aperiodic dispersion in tubes that occurs when a solute is transported by a stream of nonuniform velocity; since that time this problem has received a considerable amount of attention from other investigators. Aris (*1*, *2*) extended Taylor's work by using the method of moments, which is an interesting integral type of approach to the problem. Van Deemter et al. (*7*) also employed an integral method and discussed their results in terms of Danckwerts' holdback concept.

More recently, Bailey and Gogarty (*3*) employed an unorthodox numerical method to solve the convection equation with axial molecular diffusion ignored, which, in effect, limits the solution to large Peclet numbers. They used the pure convection solution for short values of time and then continued the solution by a technique such that the convection term was accounted for by moving the solutions down the flow. This approach essentially accounts for radial diffusion and axial convection alternately rather than simultaneously. Also, Phillip (*10*) studied the periodic dispersal problem by using eigenfunction expansions and discussed the applicability of the diffusion model to dispersion systems of this type.

Experimental results for dispersion in tubes have been reported by a number of investigators (*3* to *5*, *11*). For large Peclet numbers these results are in rather good agreement with each other, and for reasonably large values of time all of them agree with Taylor's approximate solutions. However, for slow flows the effect of Peclet number has not been determined yet satisfactorily and the limits of applicability of the Taylor-Aris solution are in doubt. Also, at low velocities, experimental results of Bournia et al. (*5*) yield values of the dispersion coefficient k which are far greater than those predicted by Bailey and Gogarty (*3*) and Taylor (*11*). Furthermore, there is a dearth of information regarding point concentration distribution, which is needed to gain a better understanding of the dispersion process.

Dispersion problems have considerable practical importance in a variety of fields. Taylor originally attacked the problem by stating that it described the dispersion of soluble materials in blood vessels. Phillip said that the dispersal of solutes in water conducting organs of plants follows similar laws. Van Deemter et al. (*7*) and Golay (*8*) pointed out the use of such theories in describing chromatographic columns. Westhaver (*15*) and Aris (*2*) discussed applications to distillation processes. We are interested because of the connection of this problem with the batch washing of ice particles to free them of brine before melting.

In the present work we solve the diffusion-convection equation including the axial diffusion term numerically and our purposes are to:

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