

CONCENTRATION CORRELATION IN A TURBULENT MIXING LAYER WITH CHEMICAL REACTIONS

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Instantaneous concentrations of two reacting species were simultaneously measured using a combined laser and electrode-conductivity technique in a turbulent liquid mixing layer with the influence of second-order irreversible chemical reactions. To investigate the effect of turbulent mixing on the mean reaction rate, the concentration correlation was estimated from the instantaneous concentration measurements for three reactions; a very slow reaction, a moderately fast reaction and a rapid reaction, and the results were compared with several closure models. The results show that the segregation parameter increases from -1 towards zero in the downstream region of the mixing layer and it approaches -1 with increasing Damköhler number. The corrected 3E closure model can best predict the concentration correlation for both moderately fast and rapid reactions. The Lagrangian stochastic model can explicitly predict the concentration correlation for all reactions though it underestimates the correlation.

Introduction

The effect of turbulent mixing on the chemical reaction rate is more significant for higher-order reactions which often occur in industrial reactors or in environmental flows. It is, therefore, of great importance to investigate the effect of turbulent mixing on the reaction rate in estimating both yields of chemical products in chemical reactors and transport of reactive pollutants in environmental flows.

When a second-order, irreversible and isothermal reaction ($A+B \rightarrow C+D$) is considered, the mean reaction rate is expressed as

$$\bar{R} = k (\bar{C}_A \bar{C}_B + \overline{c_A c_B}) \quad (1)$$

where \bar{C} is the mean concentration of one reacting species, c the concentration fluctuation, and k is the reaction rate constant. The concentration fluctuation correlation, $\overline{c_A c_B}$, is very sensitive to the progress of mixing between species A and B and it becomes zero if two species are perfectly mixed. Thus, $\overline{c_A c_B}$ is representative of the effect of turbulent mixing on the mean reaction rate and it is substantial to show explicitly the variation of $\overline{c_A c_B}$ with the evolution of turbulent mixing.

In investigating the effect of turbulent mixing, the most reliable approach is to measure directly $\overline{c_A c_B}$, but this approach requires simultaneous measurement of the instantaneous concentrations of two reacting species with a spatial resolution comparable to the smallest concentration scale, i.e. the Batchelor scale¹⁾. The Batchelor scale for a passive scalar in a liquid turbulent flow is rather smaller than the Kolmogorov scale and therefore the direct

measurement of $\overline{c_A c_B}$ is limited to a few studies^{7, 8)}. Komori *et al.*^{7, 8)} have developed the technique to simultaneously measure the concentrations of two species both in a non-reacting flow and in a reacting flow with a rapid reaction. This technique allows measurement of the instantaneous concentrations with spatial resolution comparable to the smallest concentration scale. In order to investigate the effect of turbulent mixing on the reaction rate, it is worthwhile to utilize the technique of Komori *et al.*^{7, 8)} for the direct measurement of $\overline{c_A c_B}$ in several reacting flows. Especially, direct measurements of $\overline{c_A c_B}$ for a moderately fast reaction are interesting since the time scale of the turbulent diffusion is comparable to that of the chemical reaction. However, there have been no measurements in a reacting flow with a moderately fast reaction.

Numerically speaking, a direct numerical simulation (DNS) is the most useful tool for estimating $\overline{c_A c_B}$ but it also has the same problem of spatial resolution for a liquid flow as in the direct measurement. Therefore, the application of DNS is at present limited to homogeneous reacting gas flows^{4, 5, 11)} where the Batchelor scale is comparable to the Kolmogorov scale. For engineering purposes, the closure modeling for $\overline{c_A c_B}$ is important to design and control chemical reactors. Several closure models have been proposed^{3, 6, 10, 12, 13)} to predict $\overline{c_A c_B}$ in reacting liquid flows and they have been applied to more complicated reactions than simple second-order reactions. However, the reliability of the closure models has not been fully discussed, even for simple second-order chemical reactions. This is just due to the lack of direct measurements of $\overline{c_A c_B}$. Therefore, it is of great importance to present direct measurements of $\overline{c_A c_B}$

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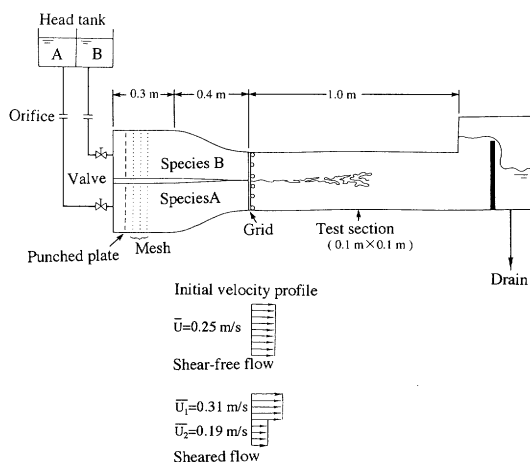


Fig. 1 Experimental setup

for several reactions with different reaction rate constants in the same turbulent field and to compare the model predictions of $\overline{c_A c_B}$ with direct measurements. In particular, it is worthwhile to show clearly how previously published closure models are reliable for reacting liquid flows. Some closure models have been examined¹⁴⁾ by comparison with measurements of the concentration statistics in a rapidly reacting flow⁷⁾ but there have been no comparisons with measurements in a reacting flow with a very slow or a moderately fast reaction.

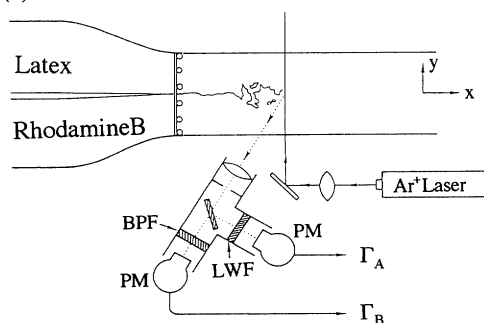
The purpose of this paper is to present reliable direct measurements of $\overline{c_A c_B}$ in reacting flows with different reaction rate constants and to examine the previously published closure models including a stochastic model⁹⁾ by comparison with direct measurements.

1. Experimental

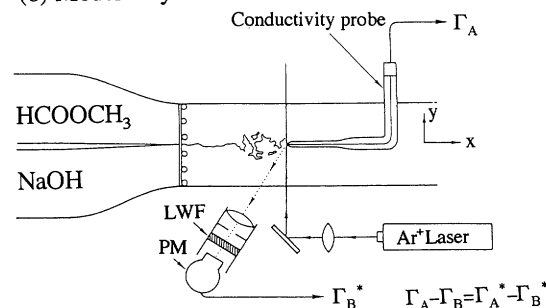
Measurements were carried out in a mixing layer downstream of a turbulence grid as shown in **Fig. 1**. Water solutions consisting of species A and B were pumped up from two large storage tanks to the head tanks, whereupon they passed through a contraction which was separated into two sections by a splitter plate. The test section was 1 m in length and 0.1×0.1 m in cross section. A turbulence grid was installed at the entrance of the test section, and the mesh size and the diameter of the rod were 0.02 and 0.003 m, respectively.

Most measurements were carried out in a shear-free (unsheared) flow where both mean velocities of the nonpremixed A and B streams, \bar{U} , were set to the same value of 0.25 m/s. To investigate the effect of the mean shear on the reaction rate, some measurements for non-reacting and rapidly reacting flows were carried out in a sheared mixing layer with an initial velocity profile illustrated in Fig. 1. For the sheared mixing layer, the velocities of the two streams were 0.31 and 0.19 m/s, respectively, and the Reynolds number based on the mesh size was kept to the same value as in the shear-free mixing layer. Measurements of the concentration were conducted down-

(a) No reaction



(b) Moderately fast reaction



(c) Rapid reaction

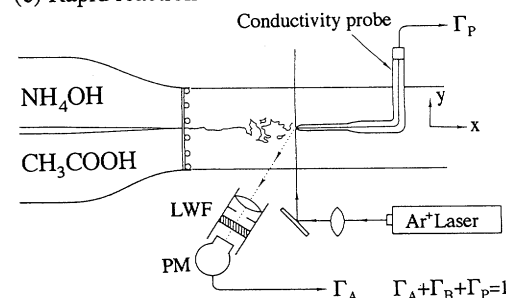


Fig. 2 Measuring system (a) No reaction case (b) Moderately fast reaction case (c) Rapid reaction case

stream of a turbulence grid in the range of $6 \leq x/M \leq 20$.

The reactions used here were a very slow reaction, a moderately fast reaction and a rapid reaction and an individual measuring technique was employed for each reaction as described below.

1) Very Slow Reaction (Non-Reacting) Case In a reacting flow with a very slow reaction, the Damköhler number is very close to zero and the diffusive-reactive mechanism can be represented by a non-reacting flow where two passive non-reactive species A (Rhodamine B) and B (0.1μ m diam. latex-particles) are mixed. Thus, measurements of the instantaneous concentrations of two non-reactive scalars were made in the above mixing layer using a combined laser-induced fluorescence and Mie scattering technique developed by Komori *et al.*⁸⁾ (**Fig. 2a**). Both solutions of species A and B with low initial concentrations were fed into the test section as shown in Fig. 1 and fluorescence from the Rhodamine B and the Mie-scattered light from the latex particles were induced by shooting a high power argon-ion (Ar^+) laser through the measuring point. The intensities of fluorescence and scattered light are proportional to the concentrations of species A and B, and therefore the simultaneous measurements of instanta-

neous concentrations of species A and B, were carried out by measuring the instantaneous intensities of the two light sources. The spatial resolution of the laser measurements, which was estimated from the power spectrum⁷⁾ of the concentration fluctuation, was estimated to be $19\text{ }\mu\text{m}$ and it was smaller than the smallest concentration (Batchelor) scale¹⁾ in the present liquid mixing layer.

2) Moderately Fast Reaction Case The saponification reaction ($\text{NaOH} + \text{HCOOCH}_3 \rightarrow \text{HCOONa} + \text{CH}_3\text{OH}$) between sodium hydroxide (NaOH : species A) and methylformate (HCOOCH_3 : species B) was used here. Initial concentrations of the two species were 100 mol/m^3 and the Damköhler number defined by $k(C_{A0}C_{B0})^{1/2}T_L$ was about 0.34. The Damköhler number denotes the relative rate of the chemical reaction. When the ion concentration of Na^+ is constant over the whole region of the flow, the instantaneous concentration of NaOH can be directly determined by measuring the ion concentration of OH^- by means of an electrode-conductivity technique. In order to make the concentration of Na^+ constant, the chemical product (HCOONa), which has the same concentration as the initial concentration of NaOH in stream A, was homogeneously premixed into stream B. Details of both the electrode-conductivity probe with a $10\text{ }\mu\text{m}$ diameter platinum wire and the electric circuit used here are described by Komori *et al.*⁷⁾. The spatial resolution of the measurements by the electrode-conductivity probe was estimated to be $43\text{ }\mu\text{m}$ from the power spectrum of the concentration fluctuation and it was comparable to the Batchelor scale¹⁾.

On the other hand, the instantaneous concentration of another reactant, HCOOCH_3 , cannot be directly measured and therefore the mass conservation equations of reacting species A (NaOH) and B (HCOOCH_3) were used to determine the concentration of species B. When we assume that all the diffusivities of species A and B are equal, the conservation equations for the concentrations nondimensionalized by the initial concentration of $C_0 = C_{A0} = C_{B0}$ give

$$\Gamma_A - \Gamma_B = \Gamma_A^* - \Gamma_B^* \quad (2)$$

where Γ_A and Γ_B are dimensionless concentrations of reacting species A and B. Γ_A^* and Γ_B^* are dimensionless concentrations of species A and B when we assume that the reactants A and B exist with no reaction in the same flow. For non-reacting species, the mass conservation is expressed as

$$\Gamma_A^* + \Gamma_B^* = 1 \quad (3)$$

From Eqs. (2) and (3), we obtain

$$\Gamma_B = \Gamma_A + 2\Gamma_B^* - 1 \quad (4)$$

If we simultaneously measure Γ_A and Γ_B^* , we can determine the instantaneous concentration of reacting species B from Eq. (4). As mentioned above, the concentration Γ_A can be measured using an electrode-conductivity probe. To measure Γ_B^* through the intensity of the laser-induced fluorescence, the fluorescence dye (Rhodamine B) was

homogeneously premixed in stream B. Rhodamine B behaved as a non-reactive species, since it was not affected by the chemical reaction. Thus, the simultaneous measurements of reacting species A and B were attained measuring Γ_A and Γ_B^* by means of a combined electrode-conductivity and laser-induced fluorescence technique, as shown in Fig. 2b.

The above measuring technique is quite original and it enabled us to measure simultaneously the instantaneous concentrations of two reacting species in a reacting flow with a moderately fast reaction. It should be especially emphasized that the present idea of premixing of Na^+ overcomes such a complicated injection method for reactive species as used by Bennani *et al.*²⁾.

3) Rapid Reaction Case The neutralization reaction ($\text{CH}_3\text{COOH} + \text{NH}_4\text{OH} \rightarrow \text{CH}_3\text{COONH}_4 + \text{H}_2\text{O}$) between acetic acid (CH_3COOH : species A) and ammonium hydroxide (NH_4OH : species B) was used. Both initial concentrations of species A and B were 10 mol/m^3 and the Damköhler number was of the order of 10^8 . In the rapid reaction, the instantaneous concentrations of species A (CH_3COOH) and chemical products ($\text{CH}_3\text{COONH}_4 + \text{H}_2\text{O}$: species P) were measured using a combined laser-induced fluorescence and electrode conductivity technique developed by Komori *et al.*⁷⁾, as shown in Fig. 2c. A low-concentration sodium fluorescein dye was homogeneously premixed in both streams. The concentration of species A (CH_3COOH) was measured using the dependency of the fluorescence intensity on pH. The concentration of product P was measured using an electrode-conductivity probe.

Using the measured concentrations of species A and P, the nondimensionalized concentration of species B (NH_4OH) was determined by the mass conservation equation:

$$\Gamma_A + \Gamma_B + \Gamma_P = 1 \quad (5)$$

2. Results and Discussion

2.1 Concentration statistics

Figure 3 shows longitudinal variations in the normalized mean concentrations of species A and chemical product P against x/M on the center line of a shear-free mixing layer flow. The mean concentration of species A is equal to 0.5 for a non-reacting case (a very slow reaction case) whereas it decreases in the region of $x/M \leq 10$ in a rapidly reacting flow (a rapid reaction case). For the rapid reaction case, the concentration of the product increases in the region of $x/M \leq 10$. For a moderately fast reaction, the mean concentrations of the reactants gradually decrease with increasing x/M . These variations reflect the progress of reactions in the region downstream of a turbulence grid.

Figure 4 shows longitudinal profiles of the correlation coefficient between the concentration fluctuations of species A and B on the center line of a shear-free mixing layer, $R_{AB} = \overline{\gamma_A \gamma_B} / \gamma_A' \gamma_B'$. The measured values of R_{AB} are

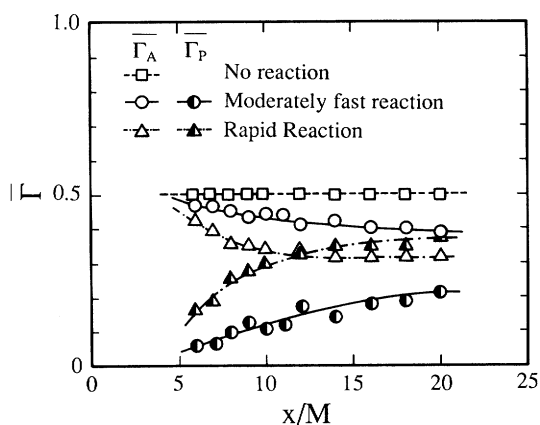


Fig. 3 Longitudinal profiles of mean concentrations of species A and P

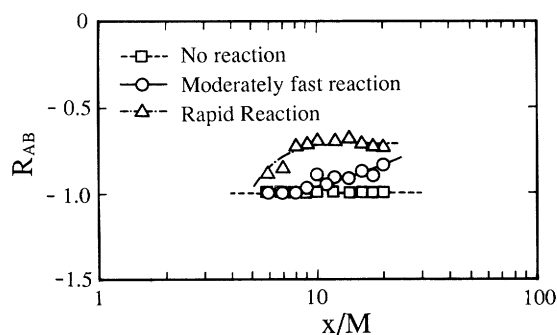


Fig. 4 Longitudinal profiles of the correlation coefficient between concentration fluctuations of species A and B

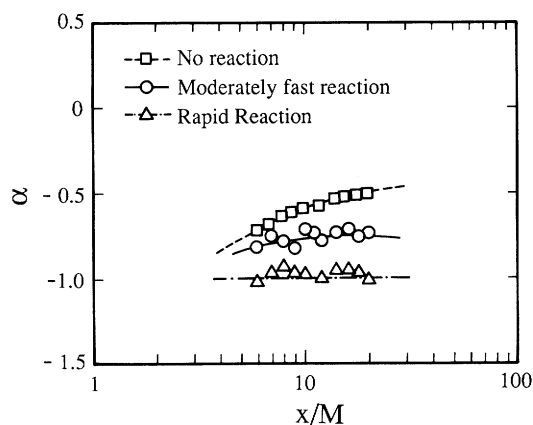


Fig. 5 Longitudinal profiles of the segregation parameter

always equal to -1 in a non-reacting flow since $\gamma_A = -\gamma_B$. For reacting flows, R_{AB} increases with increasing reaction rate (the Damköhler number) and it suggests that the negative correlation between the concentration fluctuations is weakened by progress of the chemical reaction.

Figure 5 shows the longitudinal variations in the segregation parameter on the center line of a mixing layer, $\alpha = \overline{\gamma_A \gamma_B} / \overline{\Gamma_A} \overline{\Gamma_B}$. As mentioned in the introduction, the segregation parameter is defined as the ratio of the second term to the first term in the mean reaction rate \overline{R} in Eq. 1

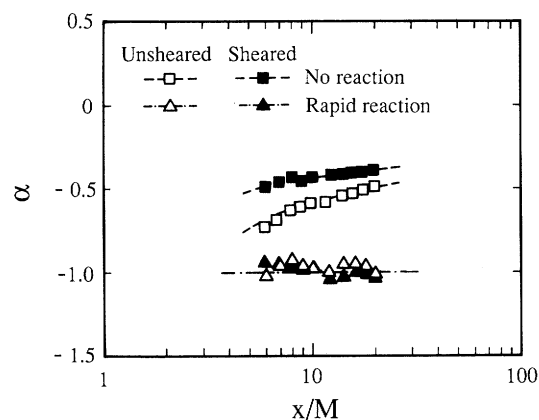


Fig. 6 Comparisons of the segregation parameter between in the sheared and unsheared flows

and it represents the effect of turbulent mixing on the chemical reaction rate. The parameter α also indicates the degree of coexistence between two reactants by molecular diffusion and therefore it shifts from -1 towards zero when species A and B are well mixed in a turbulent flow. In a non-reacting flow, α increases with x/M but the increasing rate gradually decreases in the downstream region. This is attributed to the fact that the mixing is mainly promoted by a turbulent motion behind a turbulence grid but it gradually decays in the downstream region. For a moderately fast reaction, α becomes smaller over the whole range of x/M than that for a non-reacting case. For a rapid reaction with a huge Damköhler number, α drastically decreases and it is always equal to -1. The value of $\alpha = -1$ means that reactive species A and B cannot co-exist because of rapid reaction. Thus, the segregation parameter α decreases with increasing Damköhler number. In this sense, α is an important parameter for indicating the progress of the reaction.

To promote the reaction, increasing the reaction zone by turbulent mixing is required. The mean shear may be a good promoter of turbulent mixing. **Figure 6** shows variations in the segregation parameter against the mean shear in non-reacting and rapidly reacting flows. Here, the mean shear was generated using streams A and B with different mean velocities of 0.19 and 0.31 m/s, as shown in Fig.1. The segregation parameter α considerably increases in the sheared non-reacting mixing layer compared to the shear-free (unsheared) non-reacting layer. Of course, for a rapid reaction, α is always equal to -1 since the time scale for the reaction is much shorter than that for turbulent diffusion even in the sheared flow. The effect of mean shear on the rapid reaction is clearly seen in the distributions of the mean concentration of the product as shown in **Fig. 7**. The effect is quite similar to that in a moderately fast reaction.

2.2 Comparisons between measurements of concentration correlation and model predictions

Several closure models have been developed to predict $\overline{\gamma_A \gamma_B}$ in reacting flows but the models have not been explicitly compared with the local values of $\overline{\gamma_A \gamma_B}$ measured in reacting flows because of lack of measure-

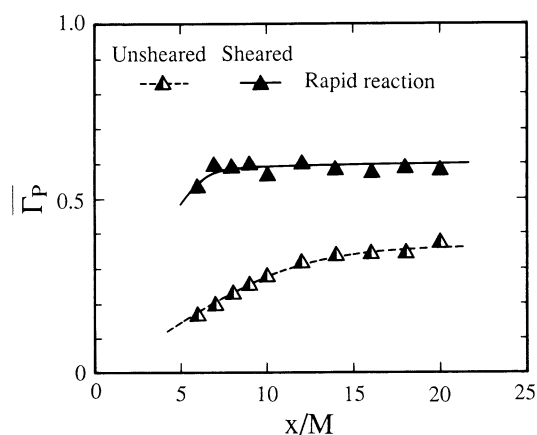


Fig. 7 Comparisons of the mean concentration of species P between in the sheared and unsheared flows

ment. Especially, a series of direct measurements of $\overline{\gamma_A \gamma_B}$ for several reactions in the same turbulent field has been required to carefully examine the previously published closure models. Thus, it is strongly desired that the direct measurements of $\overline{\gamma_A \gamma_B}$ in the present mixing layer flow with no reaction, a moderately fast reaction and a rapid reaction are compared with the predictions by the previously published closure models.

The first closure model for the correlation between the concentration fluctuations of species A and B is the first-order closure model presented by Toor¹³⁾. This model shows that the correlation, $\overline{\gamma_A \gamma_B}$, is independent of the chemical reaction and it is always equal to the correlation in a non-reacting flow:

$$\overline{\gamma_A \gamma_B} = \overline{\gamma_A^* \gamma_B^*} \quad (6)$$

The equation is derived by the assumption that the decay of the concentration fluctuations of passive scalars, $d^2(x)$, is merely determined from the flow geometry, that is, the concentration statistics in a non-reacting flow:

$$d^2(x) = \frac{\overline{\gamma_A^* \gamma_B^*}}{\overline{\gamma_{A0}^* \gamma_{B0}^*}} = \frac{\overline{\gamma_A^{*2}}}{\overline{\gamma_{A0}^{*2}}} = \frac{\overline{\gamma_B^{*2}}}{\overline{\gamma_{B0}^{*2}}} \quad (7)$$

where the subscript 0 indicates the value at the initial position of $x = 0$. When Toor's model is applied to a reacting flow, the correlation $\overline{\gamma_A \gamma_B}$ is expressed as

$$\overline{\gamma_A \gamma_B} = \alpha \overline{\gamma_{A0} \gamma_{B0}} d^2(x) \quad (8)$$

where α is defined by $\overline{\gamma_A \gamma_B} / \overline{\gamma_A^* \gamma_B^*}$ in a reacting flow. To predict $\overline{\gamma_A \gamma_B}$ by Eq. (8), the measured value of α is required. This means that Toor's model cannot explicitly predict $\overline{\gamma_A \gamma_B}$ in a reacting flow. Also, it should be noted that Toor's model cannot predict $\overline{\gamma_A \gamma_B}$ even in a non-reacting flow, since mixing ($d^2(x)$ in Eqs. (7) and (8)) is just an input parameter in the model. However, comparison between the measurements and the predictions by Eq. (8) can show whether Toor's assumption is applicable to a reacting flow or not.

The second closure model is the three environmental (3E) model of Ritchie and Tobgy¹²⁾ and it assumes that

the volume in a plug flow reactor consists of three environments; two entering environments for each reactant stream and a single leaving one. The closed form for the concentration fluctuation correlation is given by

$$\overline{\gamma_A \gamma_B} = -I_S (\overline{\gamma_A} \overline{\gamma_{B0}} + \overline{\gamma_{A0}} \overline{\gamma_B} - \overline{\gamma_A} \overline{\gamma_B}) \quad (9)$$

where I_S is the intensity of segregation for a non-reacting flow. The intensity I_S is given by

$$I_S = \exp\left[-\frac{t}{\tau_m}\right] = \frac{\overline{\gamma_A^{*2}}}{\overline{\gamma_{A0}^{*2}}} = \frac{\overline{\gamma_B^{*2}}}{\overline{\gamma_{B0}^{*2}}} \quad (10)$$

where τ_m is a time scale for turbulent micromixing and t is the time. To predict $\overline{\gamma_A \gamma_B}$ using the 3E model, the mean concentrations $\overline{\gamma_A}$ and $\overline{\gamma_B}$ should be given by the measurements and τ_m should be determined from the flow field. In this sense, the 3E model is not an explicit model. If the measured values of the segregation parameter for a non-reacting flow are introduced into Eq. (9) as

$$I_S = -\alpha^* = \overline{\gamma_A^* \gamma_B^*} / \overline{\gamma_A^*} \overline{\gamma_B^*} \quad (11)$$

a corrected 3E model can be defined. Then, α^* in Eq. (11) is given by the measured values of α shown by the squares in Fig. 5. Of course, the corrected 3E model is a kind of a parameter fitting model as is the original 3E model.

The third closure model was developed by Patterson¹⁰⁾. By using the concept of interdiffusion, the model assumes that the correlation $\overline{\gamma_A \gamma_B}$ decays with variation of $(\overline{\gamma_A^2} / \overline{\gamma_A^2})$ or $(\overline{\gamma_B^2} / \overline{\gamma_B^2})$ in a plug flow reactor:

$$\overline{\gamma_A \gamma_B} = -\frac{\overline{\gamma_A^2}}{\beta} \left[\frac{\overline{\gamma_A^2}}{\overline{\gamma_A^2}} \right] = -\frac{\overline{\gamma_B^2}}{\beta} \left[\frac{\overline{\gamma_B^2}}{\overline{\gamma_B^2}} \right] \quad (12)$$

where β is equal to 1.0 when species A and B are stoichiometrically supplied as in the present reacting flows. To estimate $\overline{\gamma_A \gamma_B}$ from Eq. (12), measurements of the mean concentration and squared value of concentration fluctuation of one species are required. Therefore, Patterson's model is not an explicit model either. Patterson's model can also be corrected by introducing a parameter ϕ into Eq. (12):

$$\overline{\gamma_A \gamma_B} = -\frac{\overline{\gamma_A^2}}{\beta} \left[\frac{\overline{\gamma_A^2}}{\overline{\gamma_A^2}} \right] \phi^* = -\frac{\overline{\gamma_B^2}}{\beta} \left[\frac{\overline{\gamma_B^2}}{\overline{\gamma_B^2}} \right] \phi \quad (13)$$

The value of ϕ in the corrected Patterson's model is determined from the comparison between the measured $\overline{\gamma_A^* \gamma_B^*}$ for a non-reacting flow and the prediction by Eq. (13). Of course, this corrected model is a parameter fitting model.

The fourth model is not a traditional closure model but the Lagrangian stochastic model developed by Komori *et al.*⁹⁾. The closure model was derived from the statistical theory of the trajectories for two marked particles being mixed in turbulence and it could determine the ensemble-averaged values of the concentrations of species A and B in a homogeneous zero-shear mixing layer with a second order chemical reaction. The most superior aspect of the stochastic model in comparison with the above three closure models is that this model can explicitly predict

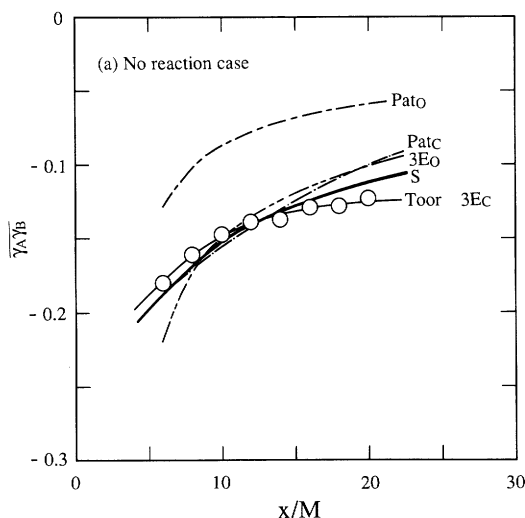


Fig. 8 Comparisons of the concentration fluctuation correlation between the model predictions (curves) and the measurements (open circles)
(a) No reaction case

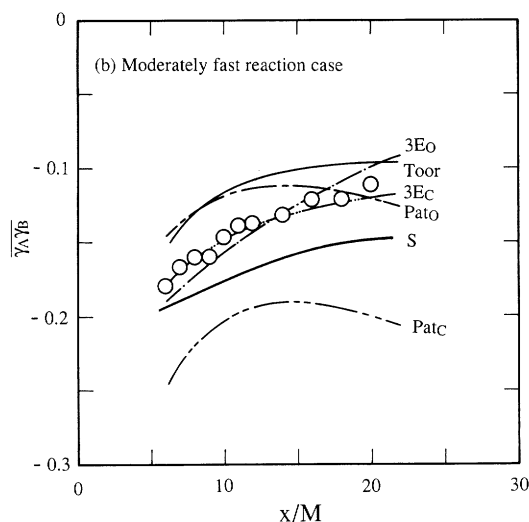


Fig. 8 (b) Moderately fast reaction case

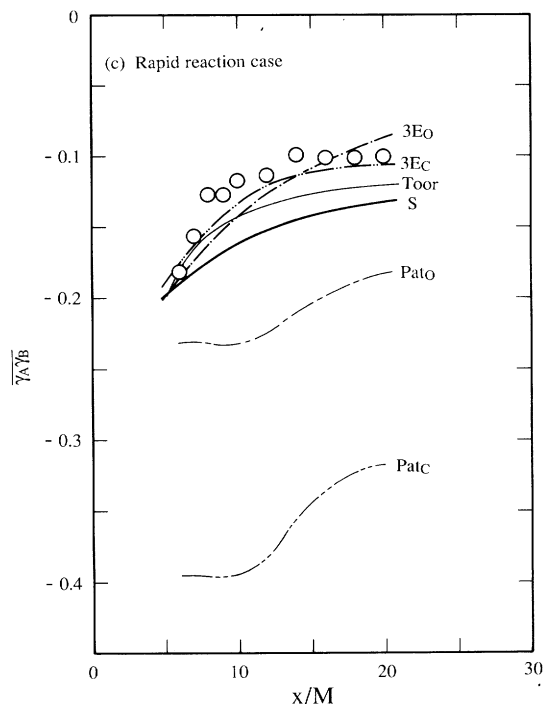


Fig. 8 (c) Rapid reaction case

different profiles from the measurements and the errors for the original and corrected Patterson's models are about +25% and -40%, respectively. Toor's (Toor) model overestimates the measurements by about 20% whereas the stochastic (S) model underestimates by about 20%. The corrected 3E (3E_C) model can best predict the measurements for a moderately fast reaction. For a rapid reaction (**Fig. 8c**), the predictions by the original and corrected Patterson's (Pat_O and Pat_C) models grossly underestimate the measurements by 60 - 190% and the stochastic (S) model and Toor's (Toor) model also underestimate the measurements by about 30% and 20%, respectively. The corrected 3E (3E_C) model best agrees with the measurements and the original 3E (3E_O) model predicts rather well the measurements.

Thus, the corrected 3E model is best for both moderately fast and rapid reactions and other models give errors of more than 20%. However, none of the closure models can explicitly predict the experimental results for three reactions. As mentioned above, the 3E, Toor's and Patterson's closure models are not explicit models and they always require measurements of the concentration of at least one species in reacting and non-reacting flows as the input data (see Eqs.7 to 13). In this sense, the stochastic model that does not need any concentration measurements may be considered superior, though it underestimates $\overline{\gamma_A \gamma_B}$. The reason why the stochastic model underestimates $\overline{\gamma_A \gamma_B}$ is that the micro mixing at scales less than the Kolmogorov scale is assumed to be under well-mixed conditions. To improve the stochastic model, micro mixing at scales less than the Kolmogorov scale should be well described by developing a new structure function⁹⁾

turbulent mixing and $\overline{\gamma_A \gamma_B}$ without using any other assumptions and measurements of concentration statistics except for the integral time scale and intensity of turbulence. Details of the stochastic model are shown in the work of Komori *et al.*⁹⁾

Figure 8 shows comparisons of the predictions with the measurements. For a non-reacting case (**Fig. 8a**), it should be noted that only the predictions by the original Patterson's (Pat_O) model and the stochastic (S) model can be compared fairly with the measurements since other models are adjusted to agree with the measurements. Predictions by the original Patterson (Pat_O) model are about 50% in excess of the measurements whereas the stochastic (S) model agrees rather well with the measured values. For a moderately fast reaction (**Fig. 8b**), the original and corrected Patterson's models (Pat_O and Pat_C) show rather

which determines the separation rate of pairs of fluid particles. Furthermore, the effect of turbulence decay in the streamwise direction should be considered. Both improvement of the stochastic model and development of more useful closure models which can be explicitly applied to all reacting flows will be interesting future works.

Conclusions

To estimate the concentration correlation in reacting flows, simultaneous measurements of the instantaneous concentrations of two reacting species were carried out in the mixing layer with no reaction, a moderately fast reaction and a rapid reaction. The first direct measurements of the concentration correlation were proposed for three reactions in the same turbulent flow field. The results can be summarized as follows.

- 1) The segregation parameter tends to increase from -1 to zero with progress of turbulent mixing in a non-reacting flow. The parameter approaches -1 with increasing Damköhler number in reacting flows and it becomes -1 for a rapid reaction. The mean shear effectively accelerates progress of the chemical reaction.
- 2) The corrected 3E model can best predict the concentration correlation for both moderately fast and rapid reactions. However, there is no explicit closure model that can predict well the concentration correlation in all reacting flows. The Lagrangian stochastic model is an explicit model for the concentration correlation but a more advanced stochastic model which can quantitatively predict all the measurements is desired.

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Nomenclature

C_i	= instantaneous concentration of species i	[mol·m ⁻³]
c_i	= concentration fluctuation of species i	[mol·m ⁻³]
$d^2(x)$	= decay function in Eq. (7)	[-]
I_s	= intensity of segregation in Eq. (10)	[-]

k	= reaction rate constant	[m ³ ·mol ⁻¹ ·s ⁻¹]
M	= mesh size of a turbulence grid	[m]
\bar{R}	= mean reaction rate in Eq. (1)	[mol·m ⁻³ ·s ⁻¹]
R_{AB}	= correlation coefficient between concentration fluctuations of species A and B	[-]
T_L	= integral time scale of turbulence	[s]
U	= mean velocity in the longitudinal x-direction	[m·s ⁻¹]
U_1	= mean velocity in stream 1	[m·s ⁻¹]
U_2	= mean velocity in stream 2	[m·s ⁻¹]
x	= longitudinal distance from turbulence grid	[m]
y	= transverse distance from the center line of the test section	[m]
α	= segregation parameter	[-]
β	= initial concentration ratio in Eqs. (12) and (13)	[-]
Γ_i	= normalized instantaneous concentration of species i	[-]
	= C_i/C_{i0}	[-]
γ_i	= normalized concentration fluctuation of species i	[-]
	= c_i/C_{i0}	[-]
κ_i	= molecular diffusivity of species i	[m ² ·s ⁻¹]
τ_m	= time scale of mixing characteristic	[s]

<Subscripts>

A	= species A
B	= species B
P	= species P
0	= initial value

<Superscripts>

'	= rms value
—	= time-averaged value
*	= value in a non-reacting flow

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