

# Experimental Study on Diffusion Field with Series-Parallel Reaction in Liquid Grid-Turbulence

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## ABSTRACT

A diffusion field with a series-parallel reaction ( $A + B \rightarrow R$  and  $R + B \rightarrow S$ ) in liquid grid-turbulence has been experimentally investigated. The instantaneous concentrations of two reactive species (R and S) and a nonreactive dye (conserved scalar) are measured simultaneously by the light absorption spectrometric method. The concentrations of species A and B are determined by the conservation law.

It is found that the mean concentrations of reactants A and B decrease and those of products R and S increase in the downstream direction by a chemical reaction. Furthermore, the correlation coefficient between species A and B has a negative value in the present measurement region and it increases in the downstream direction, while the correlation coefficient between species B and R has a positive value and it gradually decreases.

## INTRODUCTION

Turbulent diffusion and mixing phenomena with chemical reactions are practically important with regard to many engineering and environmental problems.

In order to experimentally clarify these phenomena, it is necessary to measure the instantaneous concentrations of multiple reactive species simultaneously in a turbulent flow. Several studies have been conducted in the past on the simultaneous measurements of the concentrations in turbulent reactive flows; for example, the mixing layer (Bilger *et al.*, 1991), the point-source plume (Brown and Bilger, 1996), and the line-source plume (Li and Bilger, 1996) in gas phase grid-turbulence. In the liquid phase, the studies were conducted on the mixing layer (Komori *et al.*, 1994) and the nearly homogeneous diffusion field (Kubo *et al.*, 2004) in liquid grid-turbulence. However, these

experiments are regarding single second-order reactions. For the development of a practical computational model useful for turbulent reactive flows, experimental studies using more complex reactions are keenly expected.

The present study presents the new experimental results on the diffusion field with a series-parallel reaction (Bourne *et al.*, 1985) in liquid grid-turbulence. The concentrations of all the species related to the chemical reaction are measured simultaneously by using a combined technique involving the light absorption spectrometric method (Sakai *et al.*, 1997) and conservation law. The chemical reaction considered in this study is the following series-parallel reaction:



where A, B, R, and S are 1-naphthol, diazotised sulphanilic acid, monoazo dye (red), and bisazo dye (purple), respectively. The first reaction (1) is a rapid reaction; the reaction rate coefficient is  $k_1 = 12,000 \text{ m}^3/(\text{mol} \cdot \text{sec})$  and that of the second one is  $k_2 = 0.981 \text{ m}^3/(\text{mol} \cdot \text{sec})$ . Such a large difference between the reaction rate coefficients of (1) and (2) is the distinctive feature of this reaction.

The instantaneous concentrations of two reactive species (R and S) and a nonreactive dye (conserved scalar) were measured simultaneously by the light absorption spectrometric method. From the above measurements, the instantaneous concentrations of species A and B could be determined by the conservation law. The statistical properties of the concentrations of all the species are presented in this paper.

## EXPERIMENTAL APPARATUS AND CONDITIONS

Figure 1 shows the schematic of chemical reactor used in this study. The cross-sectional area of the test section is

60 × 60 mm<sup>2</sup>, and its length is 600 mm. The grid (mesh size  $M = 10$  mm) consists of bars with a rectangular cross section of 2 × 4.5 mm<sup>2</sup>, and the solidity of the grid is 0.36. The nozzles (ID = 1.2 mm, OD = 1.5 mm) are distributed on the grid bars (see Fig. 1). An aqueous mixed solution of species B and the nonreactive dye (conserved scalar) is ejected from them into the main flow which includes species A; hence, an approximately uniform diffusion field is formed in the downstream of the grid. The experiments are carried out under the condition of Reynolds number  $Re_M = UM/\nu = 900$ , where  $U$  is the mean velocity of the main flow, i.e.,  $U = 0.11$  m/s, and  $\nu$  is the kinematic viscosity. The ejecting velocity from the nozzle was set to be  $1.5 \times U$ . The initial concentrations of species A and B are  $\Gamma_{A0} = 0.4$  mol/m<sup>3</sup> and  $\Gamma_{B0} = 24$  mol/m<sup>3</sup>, respectively. The distance from the nozzle exit in the downstream direction is denoted by  $x$ .

The simultaneous measurements of the concentrations of species R and S ( $\Gamma_R$  and  $\Gamma_S$ ) and the conserved scalar ( $F$ ) were performed by the light absorption spectrometric method (Sakai *et al.*, 1997). The concentrations of species A and B ( $\Gamma_A$  and  $\Gamma_B$ ) were determined by the conservation law:

$$\Gamma_A = (1 - F) \Gamma_{A0} - (\Gamma_R + \Gamma_S), \quad (3)$$

$$\Gamma_B = F\Gamma_{B0} - (\Gamma_R + 2\Gamma_S), \quad (4)$$

where the first and second term on the RHS represents the concentrations for the nonreactive case and the consumption by chemical reaction, respectively.

## RESULTS AND DISCUSSIONS

### Flow Field

In order to elucidate the flow field in the present grid-generated turbulence, the velocity measurements were conducted by a laser Doppler velocimeter.

Figure 2 shows the decay of turbulent intensities, where  $u'$  and  $v'$  are the r.m.s. values of velocity fluctuation in the downstream and cross-sectional direction, respectively. The velocities are normalized by the mean velocity,  $U$ , and the downstream coordinate is normalized by the mesh size,  $M$ . Although the present measurement region ( $5 < x/M < 30$ ) is near the grid, the decay of the turbulent intensities can be approximated by the following equations:

$$(u'/U)^2 = 0.0424 (x/M)^{-0.91}, \quad (5)$$

$$(v'/U)^2 = 0.0213 (x/M)^{-0.81}. \quad (6)$$

From the decay of intensities, the turbulent Reynolds number can be estimated to be  $15 < Re_\lambda < 25$  in the measurement region.

### Conserved Scalar

Before the experiments on the reactive scalars, the features of the nonreactive (conserved) scalar field were examined. As the conserved scalar, a direct dye "Rhoduline Red B" (C. I. Direct Red 31; C. I. 29100) was used.

Although the details are not described here, the fundamental characteristics of conserved scalar field are given in the following. The measured mean value of the conserved scalar,  $\bar{F}$ , is approximately constant ( $\bar{F} \approx 0.015$ ) in the measurement region. Here the conserved scalar is

defined as  $F = \Gamma/\Gamma_0$ , where  $\Gamma$  is the concentration of Rhoduline Red B and  $\Gamma_0$  is the initial concentration at the nozzle exit.

The decay of the concentration fluctuation intensity is shown in figure 3. In this figure,  $f'$  denotes the r.m.s. value of concentration fluctuation. Further, it is found that the decay of the concentration fluctuation can be approximated by the following power law:

$$(f'/\bar{F})^2 = 2.91 (x/M)^{-0.956}. \quad (7)$$

### Reactive Scalars

The results concerning the series-parallel reaction are provided in this section.

Figure 4 shows the downstream variations of the mean concentration of each species. The mean concentration,  $\bar{\Gamma}_{i0}$ , is normalized by the initial concentration,  $\Gamma_{i0}$ , and the solid lines denote the mean value of the conserved scalar ( $F$ ). The mean concentrations of reactant species A and B (Fig. 4(a) and (b)) decrease by the chemical reaction in the downstream direction, while those of product species R and S (Fig. 4(c)) increase in this direction.

The downstream variations of the r.m.s. values of concentration fluctuation are shown in Fig. 5. The concentration r.m.s. value,  $\gamma'_i$ , is normalized by the initial concentration,  $\Gamma_{i0}$ , and the solid lines denote the r.m.s. value of conserved scalar,  $f'$ . The r.m.s. value of species A becomes greater than the conserved scalar, whereas that of B becomes smaller. This is because the first reaction is enhanced at a lower concentration of species A (at a higher concentration of species B) in the present measurement condition (i.e.,  $\Gamma_A/\Gamma_{A0} > \Gamma_B/\Gamma_{B0}$ ). Moreover, the r.m.s. values of species R and S gradually increases in the downstream direction by chemical production.

Figure 6 show the downstream changes of the concentration probability density function (PDF) of all species. As expected from figures 4 and 5, it is found that the first moments (mean values) of PDF of reactant A and B decrease and those of products R and S increase in the downstream direction by the chemical reaction. At the same time, the PDF of species A becomes broad in the downstream direction, whereas that of species B becomes narrow in this direction. Furthermore, the PDFs of the species are intricately skewed by the influence of the chemical reactions.

Since the first and second reactions of the series-parallel reaction in this study are second-order reactions, the chemical source terms are represented by  $k_1 \Gamma_A \Gamma_B$  and  $k_2 \Gamma_B \Gamma_R$ . Therefore, the concentration correlation coefficients are very important in the modeling of moment closures (Fox, 2003). The concentration correlation coefficient between species A and B,  $C_{AB} = \overline{\gamma_A \gamma_B} / (\gamma'_A \gamma'_B)$  (where  $\gamma_A = \Gamma_A - \bar{\Gamma}_A$ ,  $\gamma_B = \Gamma_B - \bar{\Gamma}_B$ ), and that between species B and R,  $C_{BR} = \overline{\gamma_B \gamma_R} / (\gamma'_B \gamma'_R)$ , are shown in Fig. 7. The correlation coefficient  $C_{AB}$  has a negative value in the present measurement region and it increases in the downstream direction, while  $C_{BR}$  has a positive value and it gradually decreases in this direction.

Figure 8 also shows the segregation factors,  $\alpha_{AB} = \overline{\gamma_A \gamma_B} / (\overline{\Gamma_A} \overline{\Gamma_B})$  and  $\alpha_{BR} = \overline{\gamma_B \gamma_R} / (\overline{\Gamma_B} \overline{\Gamma_R})$ ; these factors are defined as the concentration correlation normalized by the product of the mean concentrations. The segregation factors represent the difference between the mean value of the concentration product ( $\overline{\Gamma_A \Gamma_B}$  and  $\overline{\Gamma_B \Gamma_R}$ ) and the product of the mean concentrations ( $\overline{\Gamma_A} \overline{\Gamma_B}$  and  $\overline{\Gamma_B} \overline{\Gamma_R}$ ). From this figure, the segregation factor between species A and B,  $\alpha_{AB}$ , is approximately -0.1 and that between B and R,  $\alpha_{BR}$ , decreases from 0.4 to 0.1.

These data are very important for the modeling of concentration correlation (or the chemical source term) in the turbulent reactive flows.

## CONCLUSIONS

The diffusion field with a series-parallel reaction in liquid grid-turbulence has been experimentally investigated. The instantaneous concentrations of two reactive species (R and S) and a nonreactive dye (conserved scalar) are measured simultaneously by the light absorption spectrometric method. The concentrations of species A and B are determined by the conservation law. The main conclusions are summarized as follows.

- It is found that the mean concentrations of reactants A and B decrease and those of products R and S increase in the downstream direction by the chemical reaction.
- The r.m.s. value of species A becomes greater than that of the conserved scalar, whereas that of B becomes smaller under the present measurement condition (i.e.,  $\Gamma_A / \Gamma_{A0} > \Gamma_B / \Gamma_{B0}$ ).
- The correlation coefficient between species A and B has a negative value in the present measurement region and it increases in the downstream direction, while that between species B and R has a positive value and it gradually decreases in this direction.

## ACKNOWLEDGEMENT

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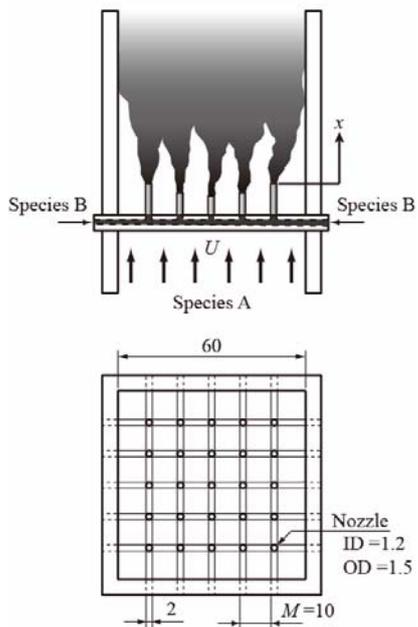


Figure 1: Schematic of the chemical reactor.

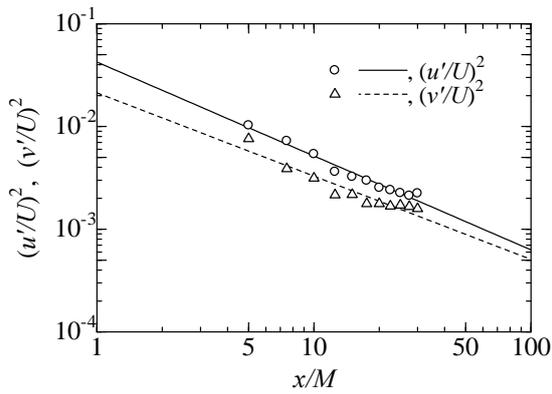


Figure 2: The decay of turbulent intensities.

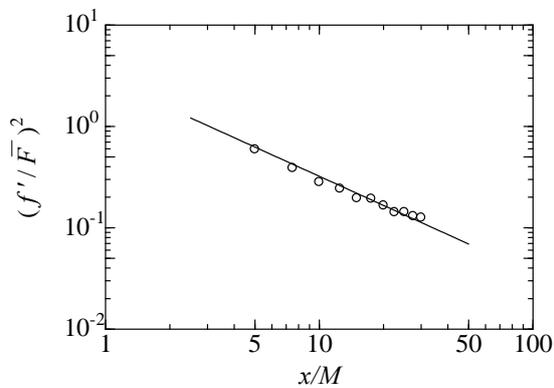
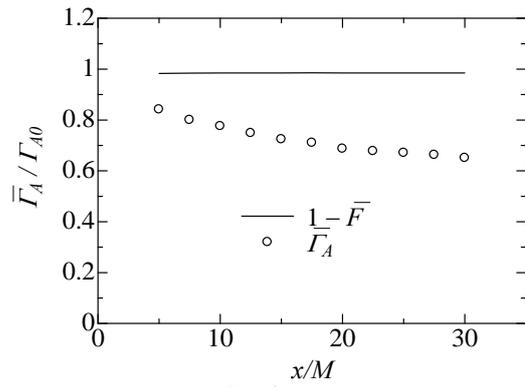
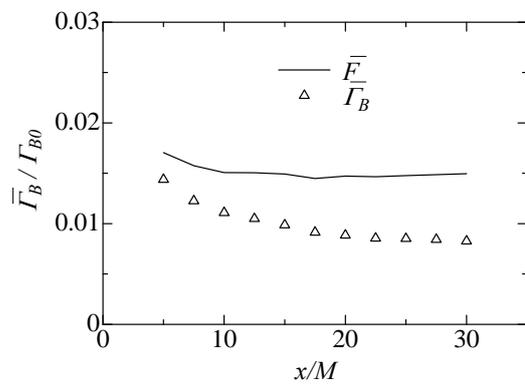


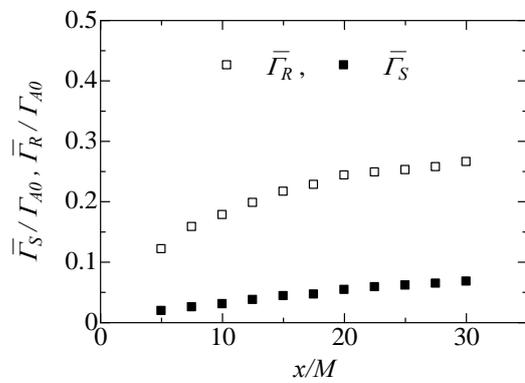
Figure 3: The decay of conserved scalar fluctuation intensity.



(a) Species A

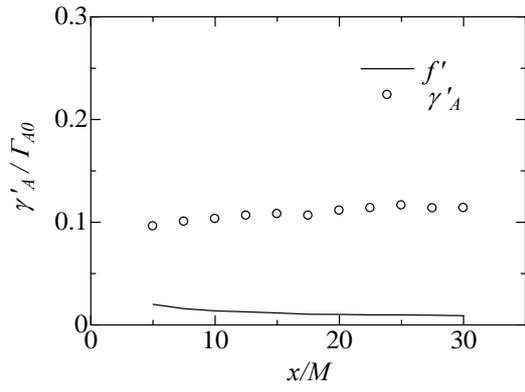


(b) Species B

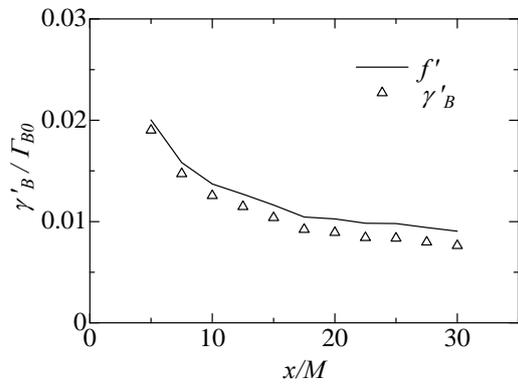


(c) Species R and S

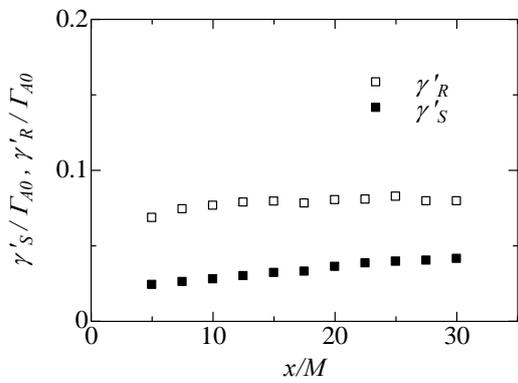
Figure 4: Downstream variations of the mean concentrations of reactive species.



(a) Species A



(b) Species B



(c) Species R and S

Figure 5: Downstream variations of the r.m.s. values of concentration fluctuation of reactive species.

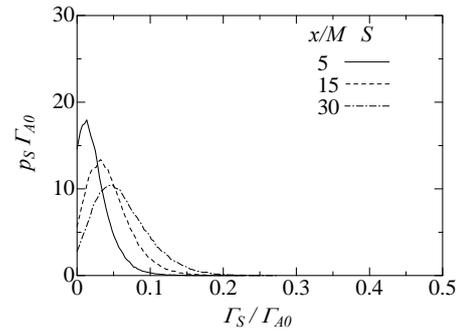
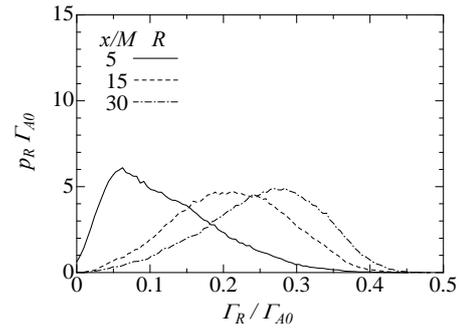
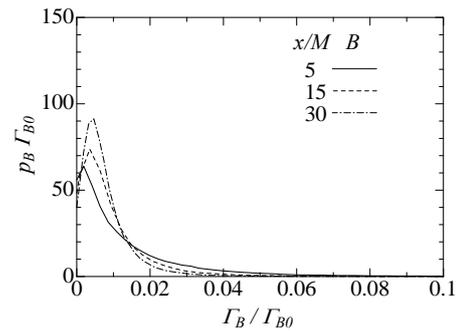
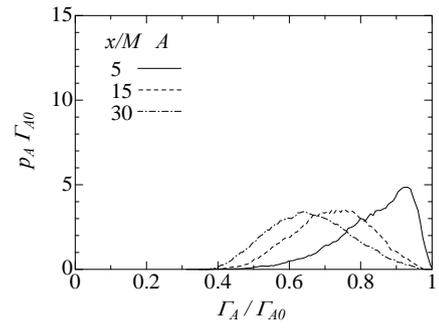


Figure 6: Downstream changes of concentration PDFs.

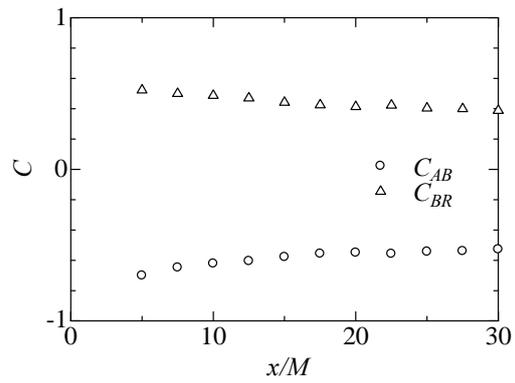


Figure 7: Downstream variations of the concentration correlation coefficients.

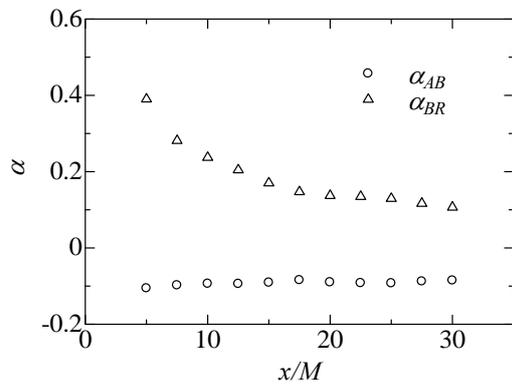


Figure 8: Downstream variations of the segregation factors.