A "Flip" Experiment in a Chemically Reacting Turbulent Mixing Layer

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An experimental investigation of entrainment and mixing in a reacting, uniform density, liquid plane shear layer has been carried out using laser-induced fluorescence diagnostics. Results indicate that the reactants mix on a molecular level and react at a composition that is nearly uniform across the transverse extent of the layer. The composition of the mixed fluid is found to be asymmetric with an excess of high-speed fluid, suggesting that entrainment into the shear layer is also asymmetric. These results are at variance with predictions of conventional models of turbulent transport and mixing.

Introduction

In recent years,[1-13] experimental results have suggested that entrainment and mixing processes in a turbulent two-dimensional shear layer are dominated by the dynamics of large-scale vortical structures. The evidence indicates that pure fluid entrained from the freestreams can be transported unmixed across the entire transverse extent of the shear layer and, in the case of chemical reactions between the entrained fluids, the chemical product is found in regions associated with the large vortical structures. This view of turbulent entrainment and mixing processes is in contrast with classical descriptions of these phenomena based on the concepts of gradient transport and eddy diffusivity. In the classical descriptions, turbulent species transport, for example, is commonly modeled as a diffusive process, whose flux is linearly proportional to the local mean concentration gradient.[14,15] The issue of mixing and turbulent fluctuations of concentration involves the distribution of values, or higher moments, of the species concentration. The methods used to address this issue generally require gradient transport assumptions of one form or another (e.g., in the classical moment approach),[15,16] or additional closure assumptions (e.g., modeling the mixing term in the probability density function approach).[17]

In order to investigate these phenomena, the mixing and chemical reaction processes in a two-dimensional shear layer were studied. Employing laser-induced fluorescence techniques and a pH-sensitive fluorescent dye, the local chemical environment was monitored in a shear layer formed between acid and base-carrying freestreams. The investigation consisted of two sets of experiments performed under identical conditions (i.e., geometry, flow parameters, chemicals, optics, etc.), with the only distinguishing feature being the exchange of the chemical composition of the freestreams between the two sets. Such experiments have been termed "flip" experiments.

Experimental Facility and Measurements

Shear Layer Apparatus

The shear layer was produced in a small blow-down facility, in which the chemical reactants in each of the two freestreams could be premixed to any concentration. A gaseous nitrogen supply system, regulated through two separate sonic orifice metering valves, one for each stream, maintained the freestream flow velocities nearly constant. Transients and possible U-tube oscillations were minimized by placing two large vessels in the gas supply route. The effective capacitance of these vessels, in conjunction with the resistance in the supply tubes, could be adjusted to give adequate damping of the whole system (see Fig. 1). The dimensions of the test section were 3.5 cm in width and 23.5 cm in span.

Experimental Technique

An acid-base reaction between sulfuric acid (H_2SO_4) and sodium hydroxide (NaOH) was used, with the fluorescent dye premixed with the acid solution. The dye was the water-soluble organic dye disodium fluorescein which fluoresces efficiently when excited by an argon ion laser, provided it is in a chemical environment whose pH is above a certain threshold. Generally, the fluorescence is off (suppressed) when the pH of the solution is below approximately 4. The volume of base, v_b, required to titrate a unit volume, v_a, of the acid/dye solution and cross the pH threshold for fluorescence could be adjusted by the proper choice of the acid and base concentrations. Figure 2 illustrates the resulting fluorescence intensity from the titration of a fixed volume of acid/dye solution. As we start adding the basic solution to the acid/dye solution, the fluorescence intensity is close to zero since the solution is very acidic. With the addition of more base, a point is reached where the pH threshold is crossed and the fluorescence "turns on." Beyond this mixture ratio, which was set to $v_b/v_a = 1.8$ in the present experiments, addition of more based merely dilutes the dye. Now the fluorescence intensity is only proportional to the dye concentration and, hence, proportional to the acid volume fraction. The fluorescence on/off transition can be made very sharp. This transition is reversible in the sense that the fluorescence intensity can be switched on and off by adding sufficient amounts of base or acid required to cross the pH threshold. Note that, since the fluorescence behavior of this dye depends on its local chemical environment, mixing can be monitored down to the molecular scale. Further details of this technique can be found in Ref. 18.

Optical Setup

The schematic of the optical setup is shown in Fig. 3. The beam of a 3-W argon ion laser (Coherent Radiation CR-3) was passed through a combination of spherical and cylindrical lenses to produce a sheet, about 1 mm thick in this experiment, illuminating a portion of the test section at the

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Presented as Paper 83-0475 at the AIAA 21st Aerospace Sciences Meeting, Reno, Nev., Jan. 10-13, 1983; received May 31, 1984.

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midspan position. A motor-driven 35-mm camera was used to photograph the resulting fluorescent chemical product on normally developed Kodak TRI-X film. Exposures were adjusted to keep the film within its latitude limits (away from saturation).

Measurements

For the experiments reported herein, the freestream velocities were set with $U_1 = 42$ cm/s on the high-speed side, and a low-speed side corresponding to a velocity ratio $U_2/U_1 = 0.45$. The flow duration was about 4 s under these conditions. This duration corresponded to the passage of approximately 70 and 25 structures at the upstream and downstream stations (as indicated by the left and right boundaries of the photographs in Fig. 5), respectively, visible on the photographs. Concentrations of the acid and base solutions in the freestreams were such that it required mixing 1.8 volumes of base to 1 volume of acid/dye to reach and cross the fluorescence threshold. The value 1.8 was chosen based on Konrad's[13] data in anticipation of maximizing the effect upon "flipping" the chemicals. On the basis of Fig. 9b in Ref. 13, it was estimated that very little of the mixed fluid in the layer would have a composition at a ratio of high- to low-speed fluid in excess of 1.8:1. The sharpness of the fluorescence on/off transition was on the order of few parts per mil, based on the mixture volume ratio. The dye concentration in the acid solution was about 2.5×10^{-6} M (molar concentration). Both solutions were sufficiently dilute, so that the resulting densities were essentially equal to that of water.

Two sets of runs were made under identical flow conditions and optical setup. In the first set, the acid solution was placed on the high-speed stream with the base on the low-speed stream. For the second set, the chemicals were reversed. Depending on which chemical is on the high-speed stream, the variation of the fluorescence intensity with the high-speed fluid volume fraction ξ can simply be obtained from Fig. 2 (see Fig. 4). The high-speed fluid volume fraction ξ is defined by the ratio

$$\xi = \frac{v_1}{v_1 + v_2} = \frac{v_1/v_2}{1 + v_1/v_2}$$

where v_1 and v_2 are the volumes of the high- and low-speed stream fluids, respectively. Therefore, in the first run, any mixed fluid at a composition

$$\xi < \frac{1/1.8}{1 + 1/1.8} = 0.36$$

will be visible, whereas, in the second run, only mixed fluid of composition

$$\xi > \frac{1.8}{1 + 1.8} = 0.64$$

will be observed. Typical results are shown in Fig. 5. The visible portion of the apparatus in the photographs corresponds to a downstream region of 10.5 cm$<x<$22.5 cm, as measured from the tip of the shear layer splitter plate, and a Reynolds number range of $2130 < Re_{\delta_{\rm vis}} < 6100$, based on the velocity difference ΔU across the layer and the local visual thickness $\delta_{\rm vis}$. It should be noted that, in this range of Reynolds numbers, the flow is in the early stages of the mixing transition[5,19] (i.e., not fully developed).

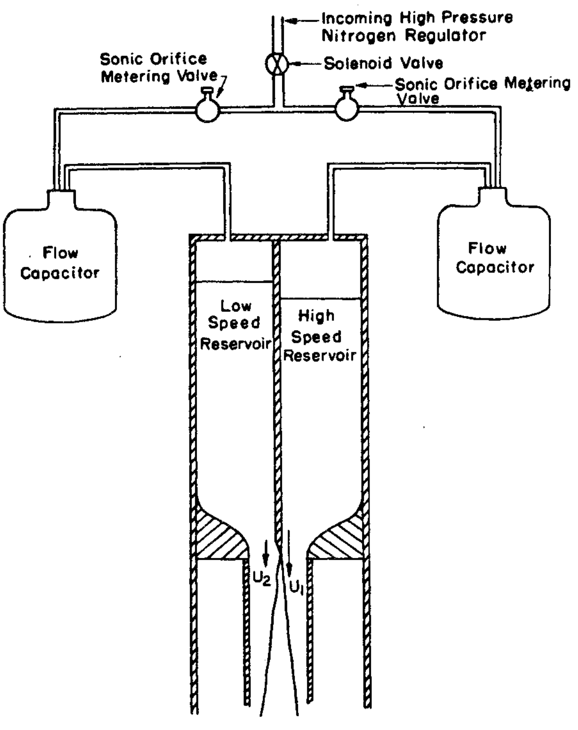


Fig. 1 Mixing layer flow facility.

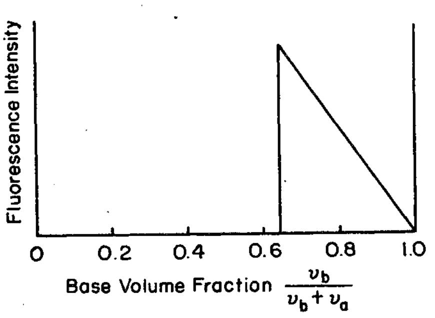


Fig. 2 Fluorescence intensity vs base volume fraction.

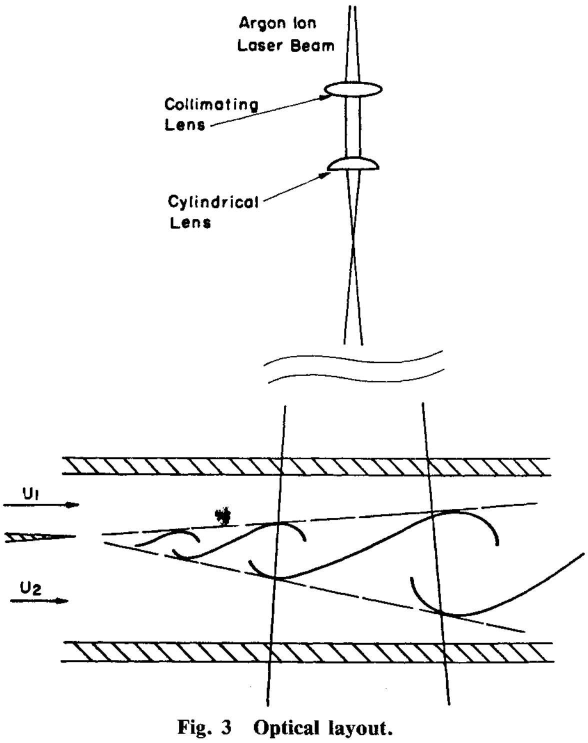
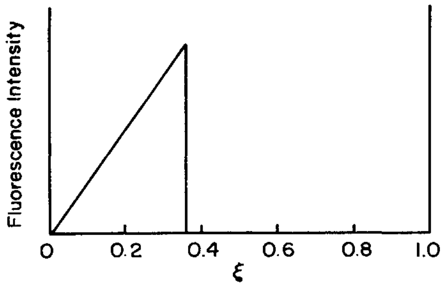
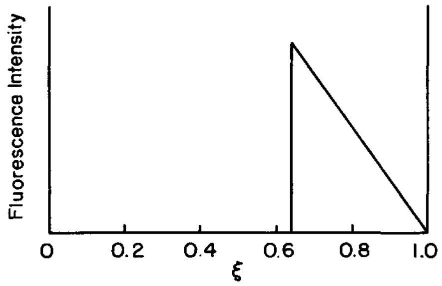


Fig. 3 Optical layout.



(a) Acid + Dye On The High Speed Side
Base On The Low Speed Side



(b) Base On The High Speed Side
Acid + Dye On The Low Speed Side

Fig. 4 Fluorescence intensity vs high-speed fluid volume fraction.

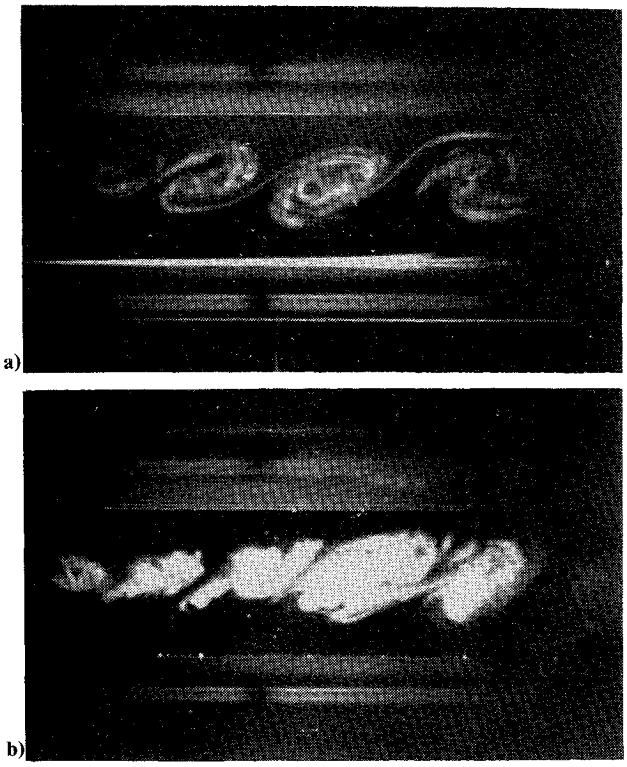


Fig. 5 Chemical reaction in a shear layer ($f/1.8$, 2-ms exposure), high-speed stream on top with $U_1 = 42$ cm/s and $U_2/U_1 = 0.45$, flow from left to right. a) Acid on the high-speed side and base on the low-speed side. b) Base on the high-speed side and acid on the low-speed side.

Results and Discussion

Figure 5 shows the regions of mixing and chemical reaction in a thin slice at the midspan of the shear layer. The fading of the fluorescence intensity toward the left and right edges of the photographs is due to the Gaussian distribution of intensity in the laser sheet. With the base on the low-speed side (Fig. 5a), any fluid that is mixed to a high-speed fluid volume fraction of $\xi < 0.36$ is visible through its fluorescence (see Fig. 4a). When the base is on the high-speed side (Fig. 5b), the mixed fluid in the range $\xi > 0.64$ fluoresces (see Fig. 4b). One conclusion can readily be made: There is a much higher probability of finding mixed fluid of composition $\xi > 0.64$, than $\xi < 0.36$. The composition of the mixed fluid is asymmetric and biased in favor of the high-speed stream.

It can be seen that the fluorescent chemical product is found in regions associated with the large-scale structures, in agreement with previous observations in chemically reacting shear layers.[4-10] Furthermore, there does not seem to be any significant systematic variation of the fluorescence intensity across the width of the mixing layer in Fig. 5b, which suggests that the mixed fluid composition is fairly uniform across the layer. This conclusion, which was recently confirmed by a direct measurement of the composition field in such a flow,[11,18] is consistent with previous findings.[12]

Based on the properties of the TRI-X film (approximately two decades of latitude for normal development), the amount of chemical product, defined here as the fraction of the fluid whose pH is in excess of the fluorescence threshold, can be estimated. The difference in the amount of product between the two cases is on the order of two orders of magnitude. It should be kept in mind, however, that this is not the amount of product in the usual sense because only the product corresponding to the mixed fluid in a selected range of compositions has been examined (see Fig. 4). This result, that the amount of chemical product, in the same flow, depends on which stream the reactants are carried in, is a direct consequence of the asymmetry of the mixed fluid composition. This effect has also been observed by Mungal[9] and Mungal et al.[10] in a gaseous layer.

It should be pointed out that a conventional gradient diffusion model of turbulent transport cannot account for this very large difference in behavior as a result of a simple exchange of the chemicals. Figure 6 is a sketch of the expected results for the present experiment, based on a gradient diffusion transport model. The mean concentration profiles of the two reactants (in the absence of a chemical reaction) and the product are indicated. Note that the product mean concentration profile has been drawn for an acid-base chemical reaction that proceeds only when the reactants are mixed with an excess of base (the experiment requires a mixture of at least 1.8 volumes of base to 1 volume of acid to yield fluorescent product, see Fig. 2). Strictly speaking, if the model does not accommodate fluctuations in the concentration of the reacting species, the resulting product profile would possess a sharp cutoff. If we assume that the range of fluctuations predicted by such a model (e.g., Ref. 16) is, at each cross-stream station y, centered around the local value of the mean (consistent with Fig. 8 of Ref. 17), then the effect of fluctuations would be to round off these sharp edges, which is indicated symbolically in Fig. 6. Note that the actual shape of the predicted product profile may vary depending on the extent of the fluctuations and the details of the mixing mechanism assumed in the model. It can be seen that flipping the freestream chemicals would result in a flip of all of the mean profiles. Consequently, these models would predict approximately the same amount of product for the two cases in our experiment. Furthermore, they predict that the reaction zone should be driven from one side of the layer to the other upon exchange of the chemicals.

The present results can be understood in part in terms of a simple mixing model proposed by Broadwell and Breidenthal.[20] According to this model, the shear layer entrains a certain ratio of high-speed fluid to low-speed fluid, which, as suggested by the work of Konrad,[13] need not be equal to unity (see also Ref. 21). The entrained fluid from the two sides of the shear layer mixes and homogenizes at a composition corresponding to this entrainment ratio. In addition, there are contributions from the strained "flame sheets"

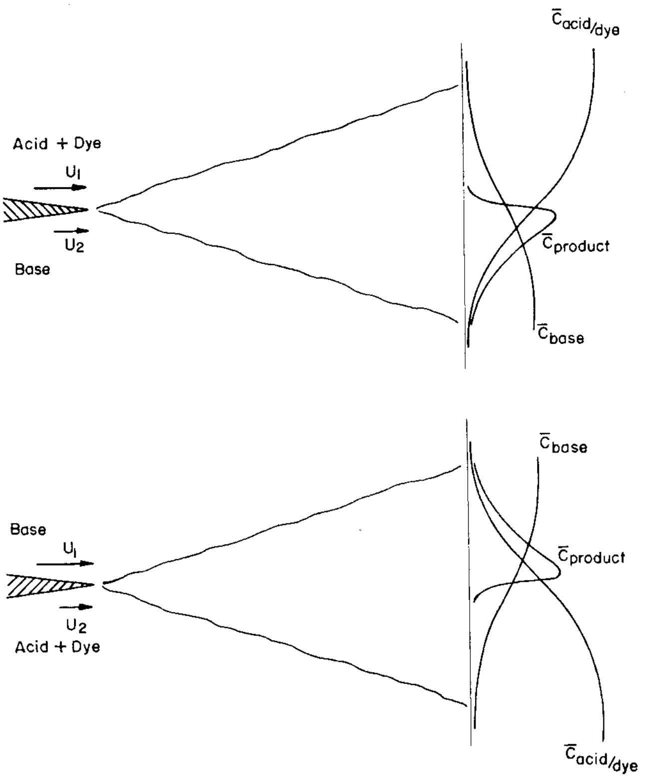


Fig. 6 Schematic of the expected mean product concentration profile based on a gradient diffusion model.

betweeen the two fluids, which, in the case of liquids (high Schmidt number), would be small. In our experiment, the predominant fraction of the mixed fluid has a composition asymmetric in favor of the high-speed fluid. This is consistent with the asymmetric entrainment into the layer described by Konrad.[13] Note also that the reaction occurs more or less uniformly across the shear layer, suggesting a transport mechanism in which large-scale motion plays an important role.

It should be emphasized, however, that the operating conditions (e.g., the Reynolds number, the mixture ratio required for fluorescence "turn-on") do influence the outcome of a "flip" experiment. These parameters were selected, in these experiments, in such a way as to accentuate the composition asymmetry in the layer. The difference, upon flipping the chemicals, may not be as large as that observed herein for different operating conditions.

Conclusions

Investigations into the mechanism of transport, mixing and chemical reaction in a plane shear layer suggest that these processes are not adequately described by the commonly used models of turbulent transport and mixing. The composition of the mixed fluid was found to be asymmetric and biased in favor of the high-speed stream, implying that the entrainment into the layer is also asymmetric. Results support the idea that this composition is nearly uniform across the width of the shear layer.

Acknowledgments

The authors would like to acknowledge the support by the Air Force Office of Scientific Research, Contract F49620-79-C-0159, for this work. The authors would also like to acknowledge helpful discussions with Professor William Reynolds.

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