

Implications of Recent Experimental Results for Modeling Reactions in Turbulent Flows

James E. Broadwell and Paul E. Dimotakis
California Institute of Technology, Pasadena, California

Introduction

THE general subject of this paper is turbulent mixing, chemical reactions, and combustion in fully developed shear layers and jets. More specifically, the purpose is to review the results of a series of experiments that seems to us to have important implications for understanding and modeling of these flows, with possible implications for dealing with turbulent transport in general. A comprehensive review of all important and useful recent experiments on the subject will not be undertaken; instead, attention is focused on some experiments that have, in our opinion, revealed new features of the mixing and combustion processes in turbulent shear flows.

The recognition of the importance of the large-scale organized structure (e.g., Ref. 1) in turbulent shear flows has given new directions to research in this field. While there is no doubt that the existence of the large structures has been recognized for some time (see, for example, Refs. 2 and 3), it was believed that their main effect on turbulent transport was the convolution at low wavenumbers of the turbulent/nonturbulent interface in the flow, which could be accounted for through the notion of intermittency.^{4,5} Much of the recent work, both theoretical and experimental, has had the objective of clarifying the fluid mechanics. For the purposes of the present discussion, our interest is in the implications for turbulent transport and diffusion of scalars, where the role of the large-scale organized motions is perhaps clearer. A complete theory would, of course, deal with the momentum and scalar equations simultaneously, but since it is likely to be some time before such a theory is available, it seems worthwhile to see what can be deduced from the evidence already at hand.

Recent Experimental Results

It is convenient for discussion to divide the experiments into three sometimes overlapping sets. The first implies that molecular transport coefficients, particularly the diffusion coefficient D , influence significantly the molecular mixing rate, even at Reynolds numbers at which turbulence is considered to be fully developed. The second set emphasizes the role of the

large-scale structures in establishing the environment for the subsequent molecular mixing. That the mixing processes are essentially unsteady on the largest temporal and spatial scales of the flow is inferred from the final group of experiments.

Molecular Transport Effects

Nitric Oxide Production in Turbulent Diffusion Flames

The amount of nitric oxide (NO) produced when nitrogen-free fuel jets burn in air is, of course, of practical importance, but the interest here is in what the measurements imply with regard to mixing and combustion. Bilger and Beck⁶ have measured the amount of NO produced in axisymmetric hydrogen jets burning in air and Peters and Donnerhack⁷ in methane-air flames. Both studies yielded an unexpected dependence of the nitric oxide production on the fuel jet Reynolds number. Bilger and Beck point out that since NO is generated only at high temperature by a slow reaction, the amount should scale with the residence time in the flame u_0/d_0 , where u and d are the jet velocity and diameter and the subscript 0 denotes the initial conditions. They find instead that the NO index (ppm of NO per unit mass of fuel) appears to depend on $(d_0/u_0) Re_0^{1/2}$, where $Re_0 = d_0 u_0 / \nu_0$ is the jet Reynolds number. The symbols have their usual meanings.

Likewise, Peters and Donnerhack find a Reynolds number dependence for the methane-air flame. To quote from their paper, "The experiments show, that there exists a Reynolds and a Froude number dependence of the emission index. If one considers the Reynolds number dependence separately, it is difficult to explain. As the ratio of the molecular viscosity to the turbulent viscosity is ... of the order of $70/Re$, the molecular viscosity can be neglected in the governing equation. Thus there is no theoretical way to predict a Reynolds number dependence of the emission index on the basis of the governing equations."

After examining several alternative causes for the discrepancy between their model and the data, they conclude that the most likely explanation is an inadequacy in their turbulence model. They further speculate that the shortcoming is somehow associated with the large-scale structure.

James E. Broadwell is a Senior Scientist in Aeronautics. He received a B.S. degree in mechanical engineering from the Georgia Institute of Technology, an M.S. degree in aeronautics from the California Institute of Technology, and a Ph.D. in aeronautical engineering from the University of Michigan. He was associated with TRW for 26 years, the last few years being involved primarily with the development of the high energy chemical laser. His current professional interests include mixing and chemical reactions in turbulent flows and molecular gas dynamics.

Paul E. Dimotakis is a Professor of Aeronautics and Applied Physics. He has received a B.S. degree in physics, a M.S. degree in nuclear engineering, and a Ph.D. degree on superfluid helium heat transfer, from the California Institute of Technology. His research has focused on turbulent flow phenomena, combustion, feedback control of separated flows, vortex dynamics calculations, and computer-aided instruction and design.

Chemical Product Formation in Gases and Liquids

A more direct illustration of the effect of the molecular diffusion coefficient in a high Reynolds number mixing layer is given by a comparison of the results of Wallace⁸ and Mungal et al.⁹ for reactions in gases with those of Breidenthal¹⁰ and Koochesfahani¹¹ in liquids. In these experiments, the product of fast chemical reactions is measured (proportional to the temperature rise in gases) in turbulent shear layers at the comparable Reynolds numbers ($Re_\delta = \delta_{vis} \Delta U / \nu \approx 40,000$) and the same velocity ratio ($U_2/U_1 \approx 0.4$). The amount of product is measured by means of the nondimensional parameter δ_{P2}/δ , where δ is the shear layer thickness δ_{vis} ,

$$\delta_{P2} = \int_{-\infty}^{\infty} \frac{c_P(y)}{c_{O2}} dy$$

where $c_P(y)$ is the product concentration profile and c_{O2} is the reactant concentration in the low-speed stream (the subscript 2 denotes the freestream bearing the normalizing reactant concentration). The comparison is shown in Fig. 1, taken from Ref. 11, where it is seen that almost twice as much product is generated in similar flow conditions in gas-phase vs liquid-phase flows.

Reynolds Number Effect on Shear Layer Product Formation

If there is an influence of the diffusion coefficient D on the product formation rate, then, in gases in which the Schmidt number $Sc = \nu/D$ is approximately unity, the "Reynolds number" $U\delta/D$ (where δ is the local transverse scale of the flow and U is the shearing velocity difference across the turbulent region) must be a relevant parameter.

That this is the case is suggested by the direct measurements of Mungal et al.⁹ for the $H_2 + F_2$ reaction in a turbulent shear layer. Figure 2, borrowed from that paper, shows that the effect is slight. The authors warn, however, that since Reynolds number has secondary effects, such as upon the state of the splitter plate boundary layer, which is known to affect high Reynolds number shear layers,¹²⁻¹⁴ and that in those experiments the Reynolds number was raised by increasing flow velocity, possibly lowering the Damköhler number to the point where kinetics effects may come into play, this result should perhaps be considered as requiring further confirmation.

In summary, these experiments imply that molecular transport coefficients cannot be neglected even at Reynolds numbers normally considered sufficiently high for the turbulence to be fully developed. Alternatively, the "high Reynolds number limits" of diffusion phenomena appear to be functions of the Schmidt number and/or the Reynolds number.

Influence of Large Structure Organized Motions

It has long been known that in fully developed turbulent shear flow, fluctuations occur with scales equal to the largest dimensions in the flow. See Ref. 5 for pertinent references and discussions of this point. The experiments cited next, however, imply that these large-scale (inviscid) motions dominate the first step in both the shear layer and jet mixing process, the entrainment, and provide the environment for the subsequent molecular mixing.

The result is that within the structures, the mass ratio of the two fluids for the shear layer, or the ratio of the jet to reservoir fluid for the turbulent jet, is determined by the large-scale dynamics.

Mixed Fluid Composition in a Two-Dimensional Shear Layer

In his measurements of the temperature in a slightly heated two-dimensional shear layer, Fiedler¹⁵ identifies the structures on his traces and notes that there is little lateral variation in the temperature distribution within them. This observation is compatible with the direct measurements of the probability density function (pdf), shown in Fig. 3, of the high-speed fluid

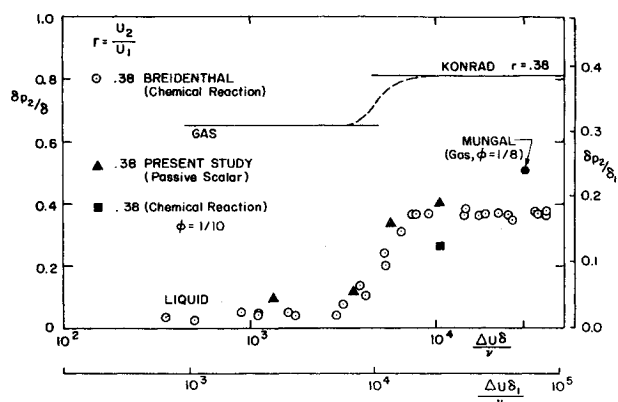


Fig. 1 Normalized chemical product thickness data for gas- and liquid-phase turbulent shear layers (from Ref. 11).

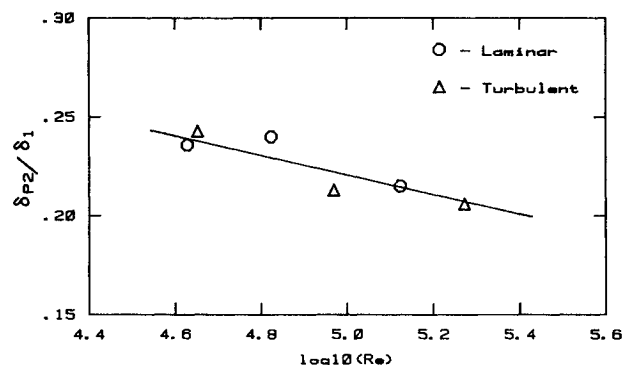


Fig. 2 Normalized product thickness Reynolds number for gas-phase turbulent shear layer (from Ref. 9).

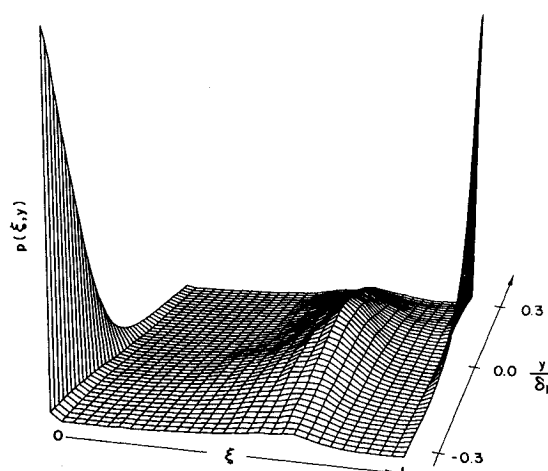


Fig. 3 High-speed fluid mixture fraction ξ probability density function for a (liquid) turbulent shear layer, as a function of the transverse coordinate y across the shear layer (from Ref. 16).

fraction of an inert scalar in a shear layer in water by Koochesfahani and Dimotakis.¹⁶ Note that the most likely mixture ratio is independent of the lateral layer coordinate y . Earlier results obtained by Konrad¹⁷ in a gas-phase shear layer passive mixing experiment are qualitatively similar, even though the resolution in Konrad's measurements was not as high.

Reacting Two-Dimensional Shear Layers

The measurements¹⁸ of the temperature rise produced by the $H_2 + F_2$ reaction in a two-dimensional shear layer were mentioned above. The structures can be identified in the time

traces in Fig. 4, taken from that paper. As in the observations of Fiedler,¹⁵ it can be ascertained that there is little lateral variation in the temperature traces within the structures. Note also that cool tongues of unreacted fluid penetrate deep into the layers. These same characteristics may also be observed in the photographs from the studies of Dimotakis and Brown,¹⁹ the data of Koochesfahani and Dimotakis¹⁶, and Breidenthal,¹⁰ who studied fast chemical reactions in two-dimensional shear layers in water. It should be noted that these experiments span a Reynolds number range of $10^4 \leq Re_\delta \leq 10^6$.

Asymmetric Entrainment into Shear Layers

In his gas phase shear layer dilution experiments between freestreams of both equal and unequal density, Konrad¹⁷ concluded that the shear layer entrains unequal amounts of fluid from the freestreams. The surprising result was that, even in the case of the equal density streams, the shear layer was found to entrain more high-speed fluid. Konrad's measurements indicate that when the densities of the two streams are equal, the ratio of high- to low-speed fluid entrained is about 1.3 (for a velocity ratio of $U_2/U_1 \approx 0.4$). Hence, in this case, there is a small difference in the amount of product, depending on which stream carried the lean reactant. If, however, the densities are different, the entrainment ratio may be large. In this case, there would be a large difference in the expected product formation, depending on which stream the lean reactant is carried.

Results from "flip" experiments¹⁸ have confirmed this in the case of the $H_2 + F_2$ shear layer reaction. See Fig. 5. In those experiments, the concentrations of the dilute reactants in a high-speed helium and low-speed nitrogen stream were exchanged, i.e., 2% H_2 in a high-speed He diluent and 1% F_2 in a low-speed N_2 diluent ($\phi = 1/2$) vs 1% H_2 in He and 2% F_2 in N_2 ($\phi = 2$). The resulting mean temperature rise from these two cases is compared to the equal concentration case ($\phi = 1$), in which both reactants were carried at a 1% concentration. The speed ratio is the same in all three cases ($U_2/U_1 \sim 0.5$). It can be seen that increasing the H_2 concentration from 1 to 2% in the already hydrogen-rich mixture in the layer (from $\phi = 1$ to $1/2$) has little effect, whereas a similar increase in the fluorine concentration from 1 to 2% (from $\phi = 1$ to 2) results in a substantial increase in the heat release.

Relevant experiments were also performed at lower Reynolds numbers by Koochesfahani et al.²⁰ in a liquid reacting layer in which it was possible to choose conditions that resulted in very large "flip" asymmetries in the formation of product.

It may be remarked that a practical application of these results may be to the flow behind flame holders in premixed streams where there is a large density difference between the

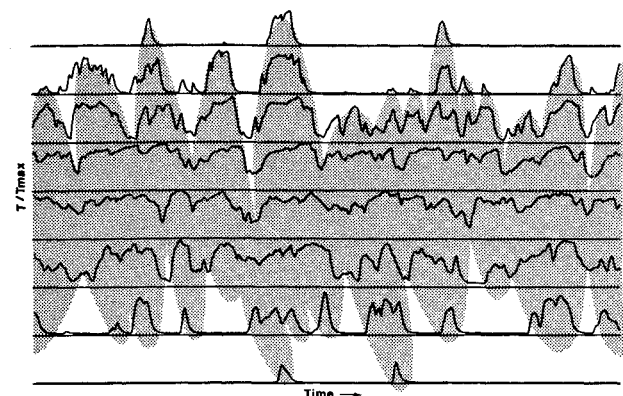


Fig. 4 Temperature time traces at 8 stations across a chemically reacting shear layer (from Ref. 18). All traces are normalized to the same maximum temperature rise ($T_{\max} = 81$ K, $T_{\text{flm}} = 93$ K).

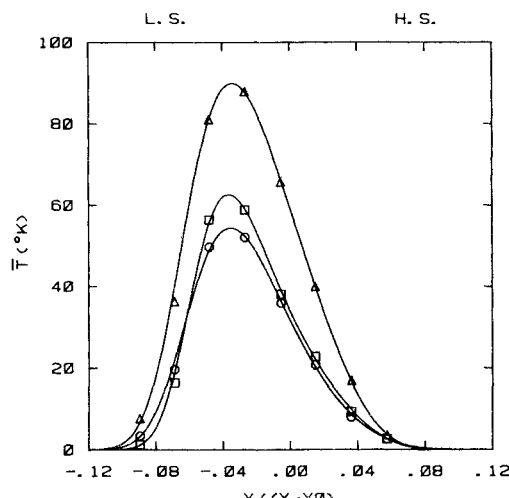
hot recirculating gases and the cool reactants, and in the experiments reported by Pitz and Daily²¹ and Ganji and Sawyer,²² as well as in the experiments of Keller and Daily²³ in a combustor shear layer formed between cool premixed reactants and hot combustion products at a velocity ratio $U_2/U_1 = 1/3$. These authors have noted the presence of the large-scale structures in these flows.

To reiterate, the conclusion we draw from the results summarized above is that the large-scale motion in the organized structures plays a dominant role in setting the environment for the molecular mixing and chemical reaction in shear layers.

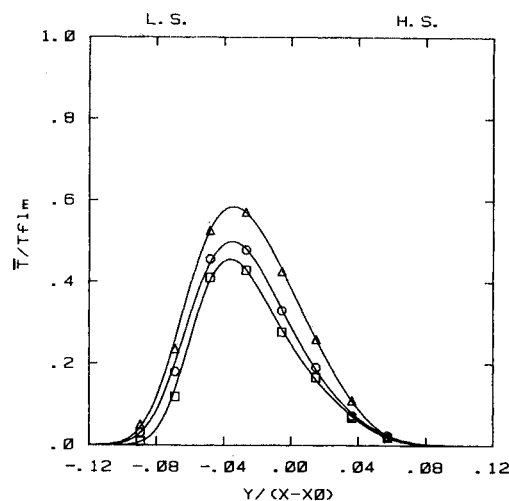
Turbulent Jets

While perhaps not satisfying the criterion in the title "recents," the experiments of Uberoi and Singh²⁴ on two-dimensional, slightly heated turbulent jets are pertinent to the present discussion. They projected hot wires across the jet at several axial locations to obtain virtually instantaneous lateral temperature profiles, which, as in the shear layer, are characterized by large regions of near-uniform composition. While it has not been established that these characteristics are the consequence of large-scale motion, they are consistent with the notion.

The experiments in which laser-induced fluorescence is used to visualize the structure in thin illuminated sheets are dis-



a) Actual temperature profiles.



b) Temperatures normalized by adiabatic flame temperature rise.

Fig. 5 Mean temperature profiles in a reacting shear layer¹⁸ for a stoichiometric mixture fraction of $\phi = 1/2$ (\square $T_{\text{flm}} = 138$ K), $\phi = 1$ (\circ $T_{\text{flm}} = 109$ K), $\phi = 2$ (\triangle $T_{\text{flm}} = 154$ K).

cussed next. The photograph made by Dimotakis et al.²⁵ (reproduced in larger format in Ref. 26) shows jet fluid interspersed with unmixed reservoir fluid throughout the jet. The still and motion picture data of Dahm et al.,²⁷ in which the same technique was employed, show that reservoir fluid is drawn deeply into the jet and that fluid from within the jet is carried outward to the edges. These data also show that, when the jet fluid reacts (reaches a certain composition with respect to the reservoir fluid), the "flame" end fluctuates in length and with a frequency that scales with $u(x)/d(x)$, the local velocity and jet diameter, i.e., large parts of the flame burn out simultaneously. More recent turbulent jet experiments²⁸ have confirmed these earlier results and also yielded instantaneous radial concentration profiles of the mixed fluid concentration in the jet that are similar to the Uberoi and Singh two-dimensional jet data.²⁴

This section is concluded by restating the second implication suggested by this set of experiments: namely, in both jets and shear layers, it is the inviscid large-scale organized motions that are responsible for the transport and that dictate the distributions of mixed fluid or chemical products within the mixing region of these flows.

Large-Scale Unsteadiness in the Flow

Turbulent flows are by their nature (or by definition) unsteady. The results discussed in this section suggest that, at least for shear layers and jets, there is unsteadiness at the largest local scale in the flow and that the fluctuation frequency scales similarly with the largest local dimension and mean velocity.

Brown and Roshko,²⁹ Winant and Browand,³⁰ and others have noted that the large structures in the shear layer have relatively long lifetimes. Two or more structures merge as they move down the layer. In the jet, the unsteadiness is made more apparent by the fluctuating "flame" lengths observed by Dahm et al.^{27,28} That the length fluctuates may, of course, be of little practical interest. The point here is that the motion indicates an inherent, large-scale ($\Delta L/\bar{L} \sim 0.4$) unsteadiness in the entrainment and mixing process, which is necessary in the correct visualization of the flow and may be essential to include in the modeling of some aspects of these flows.

Implications for Modeling

It seems a clear implication of the data cited above that any model which is to succeed in providing a unified description of the results must incorporate, either explicitly or implicitly, the large-scale motions; the evidence is that, to a large measure, many of the effects are the direct result of these motions. It is worth noting again that eddy viscosity and diffusion models are incompatible with such large scale motions, as Corrsin³¹ has argued and Sreenivasan et al.³² have directly addressed in an experimental investigation. It may also be interesting to note that the analysis by Dimotakis³³ of the asymmetrical entrainment ratio in shear layers follows from the recognition of the large-scale motion of the flow. In the context of that analysis, at least for equal freestream densities, the assertion can be made that if the largest flow structures were small, there would be no significant entrainment asymmetry.

Analyses to date that we are aware of, in which the unsteady large-scale motions emerge naturally, include those based on vortex dynamics calculations³⁴ and large-eddy simulations in wall-bounded flows³⁵⁻³⁷ and in temporally developing shear layers.³⁸⁻⁴⁰ These studies are part of the continuing effort to obtain reliable numerical solutions of the Navier-Stokes equations at Reynolds numbers small enough so that the relevant scales can still be resolved numerically, but large enough so that the solution contains the essential features of turbulent flow.

Even with the limitations on the values of the Reynolds numbers that must be imposed on such investigations—both now and in the foreseeable future—these studies, as has already been noted, involve extensive computation and many

idealizations and are not yet suitable for routine engineering use. For this purpose, considerably simplified models are still needed; a possible approach is outlined by Broadwell and Breidenthal.⁴¹ It nevertheless seems reasonable to expect that the results obtained in these computational studies will, together with the experiments, provide the basis for the formulation of such models.

With regard to Reynolds and Schmidt number effects in fully developed turbulence, more experiments are needed. Nevertheless, if the present results are accepted, it is no longer satisfactory to consider the molecular transport processes as overwhelmed by the turbulent ones, and thus ignored. On the issue of Reynolds number, we should also point out that, for many of the experiments cited, the Reynolds numbers were quite high, so care must be exercised in comparing these results with those of small-scale, high-temperature combustion experiments for which the appropriate Reynolds numbers may be low.

Conclusions

The objective of this discussion was to present the results of a set of experiments that illuminate several critical features of turbulent mixing in fully developed shear flows. The inferences drawn from these results have been stated in the text and hence need only be summarized briefly here, namely:

- 1) Molecular transport coefficients play a significant role in turbulent mixing phenomena, even in fully developed turbulent flows.
- 2) The large-scale structure dynamics dictate the entrainment, which in turn provides the environment for the subsequent turbulent cascade and mixing processes.
- 3) Turbulent shear layers and jets are essentially unsteady at the largest temporal and spatial scales in the flow.

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