Analysis of homogeneous turbulent reacting flows

By A. D. Leonard[1], J. C. Hill[1], S. Mahalingam[2], AND J. H. Ferziger[2]

Full turbulence simulations at low Reynolds numbers were made for the single-step, irreversible, bimolecular reaction between non-premixed reactants in isochoric, decaying, homogeneous turbulence. Various initial conditions for the scalar field were used in the simulations to control the initial scalar dissipation length scale, and simulations were also made for temperature-dependent reaction rates and for non-stoichiometric and unequal diffusivity conditions. Joint probability density functions (pdf's), conditional pdf's, and various statistical quantities appearing in the moment equations were computed. Preliminary analysis of the results indicates that compressive strain-rate correlates better than other dynamical quantities with local reaction rate, and the locations of peak reaction rates seem to be insensitive to the scalar field initial conditions.

1. Background

Chemically reacting species that are not premixed are separated by a reaction zone, unless the time scale for mixing is much less than the time scale for reaction. The structure of the reaction zone is determined by interactions of the turbulent motion, molecular diffusion, and chemical kinetics. Molecular diffusion controls the rate of reaction when the kinetic rate is very fast and the reaction zone becomes a surface separating regions of reactants. This surface is typically a contour surface for a conserved scalar species. The reaction zone has a finite thickness for fast but finite rate kinetics. The reactants become completely mixed when the kinetic rate is very slow, and the classification of "non-premixed" is then irrelevant, because the kinetics, rather than transport processes, control the rate of reaction.

Previous simulations of inert scalars are useful in the study of reacting flows because an infinitely fast reaction can be described in terms of a conserved scalar species. Simulations of inert scalars in homogeneous turbulence show a tendency for the scalar gradient to align with the most compressive rate of strain of the velocity field and for the rate of scalar dissipation to be highest where the alignment of the scalar gradient and the direction of the compressive strain rate is greatest (Ashurst *et al.*, 1987). Previous work with reacting scalars (Leonard and Hill 1987, 1988; Hill, Leonard & Rogers 1987) show that the compressive strain rate direction tends to be perpendicular to the reaction zone, in agreement with the results of Ashurst *et al.*

1   University of Iowa
2   Stanford University

Probability density functions (pdf's) are frequently used in the statistical treatment of reacting turbulent flows, because the source terms do not need to be directly modeled. Mixing effects are seen in the statistical equations as conditional expectations of scalar dissipation. Eswaran and Pope (1988) have used direct numerical simulation to study the evolution of the pdf and the conditional scalar dissipation of an inert scalar species. The initial mixing rate was increased by a decrease in length scale, but evolution of the shape of the pdf of the scalar was not affected. The pdf was initially a "double delta" function, which would be appropriate for the conserved scalar of unmixed reactants. It evolved into a nearly Gaussian form. The conditional dissipation approached a self-similar value and became independent of the level of the scalar. The conditional dissipation needs to be modeled properly to give accurate mixing results, but current modeling efforts (*e.g.*, Pope, 1985; Kollmann, 1987) do not account for the effects of reaction rate on the mixing terms. The price to pay for treating the source terms exactly in reacting flows is the need to model all of the mixing effects, and this must be done in a higher dimensional system (Hill, 1988).

2. Problem Description

The specific problem under study is an irreversible reaction of two initially segregated species in homogeneous turbulence. The reaction mechanism can be expressed as

$$A + B \rightarrow P.$$

Full turbulence simulations were performed for the evolution of the concentration and velocity fields, taking into account the advection and Fickian diffusion of species A, B, and P. The turbulence in most cases was isotropic and decaying. The velocity field was incompressible and not affected by the concentration fields, even in the studies using a temperature-dependent reaction rate. The governing equations for the study are

$$\frac{\partial A}{\partial t} + \mathbf{u} \cdot \nabla A = \mathcal{D}_A \nabla^2 A + w_A,$$

with similar equations for the evolution of species B and P, and

$$\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{\nabla p}{\rho} + \nu \nabla^2 \mathbf{u}.$$

The concentration of species A is denoted by A and the rate of change due to reaction by w_A. Two forms of the source terms were used in the studies,

$$w_A = -kAB$$
$$w_A = -k_0 e^{-T/T_a} AB,$$

where the k's are reaction rate constants and T_a is the activation energy in the nonisothermal case.

The initial concentration distribution was one of two forms: alternating stripes of reactant species aligned perpendicular to one of the coordinate axes, or spatially

segregated species uniform on the largest scales of the domain. The second form of initial conditions was created using a procedure similar to that used by Eswaran and Pope (1988). The initial velocity field was chosen by randomly selecting Fourier coefficients and scaling them so the three dimensional energy spectrum fit a specified function. This initial velocity field was allowed to develop in a "pre-simulation" until the skewness of the velocity derivative had a value that agreed with experimentally measured conditions in turbulent flows.

The simulation method used an existing pseudospectral code (Rogallo, 1981) that was modified to include the rate of change of scalar values due to chemical reactions. The domain for most studies was $64\diamond3$ points. Three velocity components and eight scalar values were included in the numerical solutions.

One objective of the study was to determine the influence of local properties of the turbulence on the local reaction rate and structure of the reaction zone. This includes studies of the alignment of directions of strain rate and concentration gradients for reacting scalars for different speeds of reaction and of the influence of vorticity on reaction zone structure. The long-term goal of such a study is to model average reaction rate using strain-rate parameters.

A second objective was to determine the importance of the initial conditions in previous calculations (Leonard and Hill 1987). In these studies with a preferential alignment of the scalar fields and no variation in scalar length scales, globally-averaged scalar microscales showed little sensitivity to reaction rate.

A third objective was to determine if there is any effect of reaction rate on conditional expectations of molecular diffusion terms in a hierarchy of pdf equations. The modeling of these terms neglects any dependence on reaction rate, despite the fact that the pdf approach is widely used in the statistical treatment of reacting flows.

Additional questions concerned the behavior of reactions with non-stoichiometric conditions or with species having different mass diffusivities, as well as the local structure of the reaction zone for temperature-dependent kinetics.

2. Approach

The existing databases of homogeneous turbulent flows with chemical reaction were inadequate to answer the questions that had been raised, and so the emphasis was placed on the generation of new databases during the Summer Program, with careful consideration of physical parameters and initial conditions. Additional diagnostics were developed and added to the code to permit evaluation of the evolution of single point pdf's, single-point conditional expectations, and averages over two, rather than three, spatial dimensions during the course of the simulations.

The conditions of the studies made during the Summer Program are summarized in Tables I and II. Table I groups the simulations into 7 different studies, each study consisting of one or more simulations. Parameters for the scalar variables were varied within each study, as well as between different studies, in order to isolate effects such as initial conditions or kinetic mechanism.

Most studies have in common a decaying isotropic turbulent velocity field which has evolved from what will be called a "developed" initial field, isothermal reaction,

Table I.   Summary of Studies⋄a

Name of Study	Effects Studied	Initial Conditions	Comments
1. "Non-ideal" a) Non-stoichiometric b) Unequal diffusivity	Migration of reaction zone	Gaussian v-field (Same as '87 study) Scalar slabs	Reaction zone follows $X = 0$
2. "Slab"	Kinematics of reaction zone (Improved v-field)	Developed v-field Scalar slabs	Confirmed '87 results re: strain effects
3. "Flame"	Ignition and quenching	Developed v-field Scalar slabs Isothermal	Arrhenius rate expression Quenching not observed
4. "Stirred"	Effect of scalar field initial conditions	Developed v-field Prestirred scalar	Conditional scalar dissipation measured
5. "Stripes"	Effect of scalar dissipation scale	Developed v-field Scalar stripes (1,2,3,4)	Conditional scalar dissipation measured
6. "Forced"	Decouple reaction rate from turbulence decay	Developed v-field Scalar slabs or other	Incomplete (Compiled code untested)
7. "Shear"	Strain-rate dependence on reaction rate and scalar field structure	Top-hat v-spectrum Scalar slab in plane of shear	Incomplete (Excessive Gibbs' ringing for conditions used)

⋄a All studies were for decaying homogeneous turbulence except for #6 and 7. The term "scalar slabs" denotes one stripe of one reactant centered in the domain of non-premixed reactants, hence there are two reaction zones initially. Improved diagnostics, including various joint-pdf's and planar averages, were used for all studies except #1.

equal mass diffusivities for all species, and stoichiometric proportions of reactant concentrations. The parameters and initial conditions shared by a majority of the simulations are given in Table II. The individual features of each study that were changed are discussed in the remainder of this section, using the informal names in Table I as convenient identifiers.

1. **"Non-ideal."** The non-ideal study examined the migration of the reaction zone for two different conditions. One simulation was made with stoichiometric coefficients (B_0/A_0) of 1 and 2 for two different reaction rates. The second simulation

Table II. Conditions for the Simulations

Physical parameters	
ν	0.02
$\mathcal{D} \diamond$ a	0.0286
k, k_0	various$\diamond$ b
Initial values$\diamond$ c	
u'	1.03
λ_g	0.381
Λ_f	0.844
$\bar{A}, \bar{B}$	1.0
λ_A	various

$\diamond$a $\mathcal{D}_A = \mathcal{D}_B = \mathcal{D}$ for all studies except #1b.

$\diamond$b The parameter was varied within some studies, as well as between studies.

$\diamond$c These initial conditions are for the "developed" velocity field used in studies #2–5, and for a stoichiometric proportion of reactant concentrations.

used a mass diffusivity ratio, $(\mathcal{D}_A/\mathcal{D}_B)$, of 1 and 3.5 for a stoichiometric ratio of 1. The initial reactant concentrations were the "scalar slabs," meaning one stripe of each reactant. The initial energy spectrum for the velocity field fit a Gaussian form, and the velocity field was allowed to develop for 100 time steps before the reaction began. The initial value of R_λ was about 20. The Damköhler numbers based on the diffusivity and initial mean concentration of species A and defined by

$$\mathrm{Da_I} = kA_0\Lambda_f/u'$$
$$\mathrm{Da_{II}} = kA_0\lambda_A \diamond 2/\mathcal{D}_A$$

were 0.8 and 4 for the first kind and 30 and 150 for the second kind.

2. **"Slab."** The slab study was aimed at repeating the results from the previous Summer Program with a different detailed velocity field. Fourier coefficients for the velocity field were chosen from a Gaussian distribution, subject to satisfying

the continuity equation and an initial energy spectrum $E(k) \propto k^4/(1 + (kL)^3)^2$, and allowed to develop for 100 time steps. This initial field had more energy at high wavenumbers than did the field used for the 1987 study, and so R_λ was smaller. The initial value of R_λ was 19.7. Two reactions were included in the simulation with different reaction rate coefficients. The Damköhler numbers for this study were 1.64 and 6.56 for the first kind and 62.8 and 251 for the second kind. Eight scalars were carried in the simulation: the reactant and product concentrations for each reaction and the concentrations of two inert species having different initial conditions.

3. "Flame." The "flame" study used the same initial conditions for the velocity and reactant species as in the slab case, but the reaction rate was temperature depen-dent, with an Arrhenius-type rate constant. For this case the temperature was calculated (actually, the fractional approach to the adiabatic flame temperature was used), rather than the product concentration. Simulations were performed for two different values of the Lewis number, in order to examine the possibility of strain-induced quenching, and also for two different values of the Zeldovich number. The Zeldovich number is defined here as the logarithm of the ratio of the reaction rate at the adiabatic flame temperature to the reaction rate at the initial temperature. Zeldovich numbers of 1 and 2 were used in the study. The Damköhler numbers based on the initial temperature and mean concentrations were the same as those used in the "slab" study.

4. "Stirred." Conditions for the "stirred" study were identical to the "slab" case, except for having different initial scalar distributions. A scalar field was defined in a manner similar to that for the velocity field, by scaling Fourier coefficients to fit an energy spectrum and advancing the coefficients in a "pre-simulation". In the case of the reactant concentrations, one reactant was set to be a (positive) initial value at the points were the initial scalar was positive and the other reactant was set to be a (positive) initial value at the points where the initial scalar was negative. The high wavenumbers were damped to eliminate Gibbs' ringing in the initial conditions. The reactant species were spatially segregated, but uniform on a large scale. The microscales for the reactants were much smaller than in the slab case, so mixing will tend to be faster. The Damköhler numbers of the second kind were 7.71 and 30.9, while the Damköhler numbers of the first kind were identical.

5. "Stripes." The initial values of reactant concentration in the slab and stirred cases differed both in orientation of the reaction zones and in the dissipation length-scale. The stripe study isolates the effect of the initial concentration scale from the orientation. One reaction rate coefficient is used for the calculation of reactant species for 4 reactions, each with different initial conditions. Damköhler numbers of the second kind were 13.7, 19.2, 30.4, and 62.8.

6. "Forced." Reactions in decaying homogeneous turbulence are unduly influenced by the amount of mixing in the early part of the simulation and controlled by diffusion in the later parts of the simulation. A forcing mechanism was devised to maintain a constant energy level but has not been implemented at this point.

7. **"Shear."** The same scalar field initial conditions of the "slab" case were used in the shear case, with the initial reaction zones lying in the plane of shear, but with a mean velocity gradient imposed on the flow. The initial velocity field was defined using the top-hat energy spectrum $E(k)$ = constant, for $k_1 < k < k_2$. The resolution was doubled in the direction of the mean flow, using a wavenumber grid $64 \times 64 \times 128$. A preliminary run was made—at too high a Damköhler number, apparently—but discontinued because of excessive Gibbs' ringing. Intensification of vorticity that occurs in the shear case clearly imposes limitations on the Damköhler number range that can be simulated.

3. Preliminary Results

Because most of the effort at the Workshop was spent in developing new databases for subsequent analysis, very little analysis was accomplished at the Workshop itself, and the preliminary results discussed here were obtained by cursory examination of some representative save-fields. Interpretation of the statistical results, especially the dependence of conditional expectations of scalar dissipation rate on the Damköhler number (dimensionless reaction rate constant), is still in progress, as is a more detailed examination of the saved fields. The principal results obtained so far deal with the kinematics of the reaction zone and with the influence of initial concentration length scales on the reaction (Hill & Leonard, 1988; Leonard *et al.*, 1988), and some of those results are summarized below.

The main observation from the "Non-ideal" study was the movement of the reaction zone for both the non-stoichiometric and the unequal diffusivity cases. The maximum reaction rate for the non-stoichiometric case occurred at the same point as the value of the conserved scalar corresponding to stoichiometric conditions ($X \equiv A - B = 0$). The conserved scalar treatment is not valid for the case of reactants with unequal diffusivities, but the behavior is qualitatively the same for the two cases. The mass flux of reactants to the reaction zone must be the same for both reactants, but the fluxes are unequal in the non-ideal cases, either because of a difference in diffusivity or in concentration gradient, and so the reaction front must move.

"Slab" initial conditions are used to study the structure of the reaction zone because two distinct fronts are present in the simulation. In the present study the initial reaction zones are perpendicular to the y axis and located at $y = \pi/2$ and $y = 3\pi/2$. The average positions of the fronts will not change in isotropic turbulence for reactions between stoichiometric proportions of reactants with equal diffusivities.

The scalar field has a preferential alignment with this initial condition. The gradients of reactant concentrations are in the direction of the y axis. The initial velocity condition is isotropic, so the probability of the cosine of the angle between concentration gradient and any of the eigenvectors of the rate of strain tensor is uniformly distributed. The concentration gradients become aligned with the direction of most compressive strain. Figure 1 shows the pdf's of the angles between the gradient of concentration for species A and eigenvectors of the rate of strain tensor for the slab
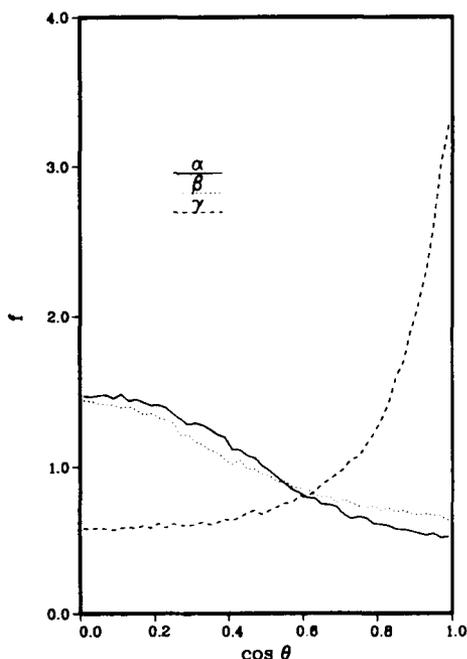
FIGURE 1.   Probability density function $f(\cos\theta)$ of the alignment of the gradient of reactant species concentration and the eigenvalues of the strain-rate tensor, for a *slab* case at $t = 2.75$.

case at a dimensionless time of 2.75. The pdf is sharply peaked at $\cos\theta_\gamma = 1$ (where θ_γ is the angle between the gradient of species A and the eigenvector corresponding to the most compressive strain rate) after only 100 steps or $t = 0.4$. The alignment is in agreement with the results of Ashurst *et al.* (1987), but the present results show how rapidly it develops.

A cross section of the domain in the slab case, either in an $x-y$ or a $y-z$ plane, shows two distinct, distorted reaction zones, as seen in Figure 2. The degree of alignment with the eigenvector corresponding to γ, the least principal strain rate, is shown in Figure 3. Concentration gradient alignment with compressive strain is important because the strain increases the magnitude of the gradient, which increases the mass transport of reactant species to the reaction zone, which increases rate of reaction. This is shown by the set of equations developed from the conservation equations for the reactants in the case of an isothermal reaction,

$$\frac{d\bar{A}}{dt} = -k\overline{AB},$$

$$\frac{d\overline{AB}}{dt} = -2\mathcal{D}\overline{\nabla A \cdot \nabla B} - k\overline{AB(A+B)},$$

$$\frac{d\overline{\nabla A \cdot \nabla B}}{dt} = -2\mathcal{D}\overline{\nabla\nabla A : \nabla\nabla B} - 2\overline{\nabla A \cdot \mathbf{e} \cdot \nabla B}$$

$$- k\left[\overline{(A+B)\nabla A \cdot \nabla B} + \overline{B\nabla A \cdot \nabla A} + \overline{A\nabla B \cdot \nabla B}\right].$$

We refer to the term $\nabla A \cdot e \cdot \nabla B$ as "gradient compression", because the gradients are being amplified by the tendency of turbulence to move material surfaces together. When the scalar gradients are aligned with γ the gradient compression becomes $-\gamma |\nabla A||\nabla B|$. This term, which is positive because the gradients of A and B are in opposite directions and $\gamma < 0$, increases the magnitude of $\nabla A \cdot \nabla B$ at a rate $e \diamond -2\gamma t$ in the absence of diffusion or reaction. Contours of AB, $|\nabla A \cdot \nabla B|$, and $\nabla A \cdot e \cdot \nabla B$ all show peak values in the same regions.

A second consequence of the scalar gradient alignment is that the location of peak reaction rates seems not to be very sensitive to the initial conditions of the scalar field. The initial values of the "Stirred" case were determined randomly, while the initial values of the "slab" case were chosen deterministically. The velocity field was the same for the two cases. A cross section of the slab case in a $x-z$ plane, as well as cross sections of the stirred case in any plane, show a more random structure than that in Figure 2. Regions of intense reaction rate for the slab and stirred cases coincide in a plane that was the center of one of the initial reaction zones for the slab case (Figs. 4 and 5), even though the initial concentration fields are different.

Since the reaction rate is high in the same locations for completely different initial conditions, it may be possible to predict the structure of the reaction zones from a knowledge of velocity field. One parameter that must be used is the rate of strain. Criteria were proposed by J. Hunt and A. Wray at the CTR Summer Program to determine kinematic structures of the flow field. These structures are identified as streams, eddies, and convergence zones, based on the strain rate invariants, vorticity, kinetic energy, and pressure. Hunt and Wray suggest that the reaction rate will be highest in the convergence zones. Simulation results, however, show that regions where the reaction rate is high are not necessarily convergence zones. The gradient compression term appears to be the best marker of regions of high reaction rate.

Only a few remarks will be made here about the remaining studies. First, the "Flame" study. The reaction rate has been found to be higher when the magnitude of γ is large, because of the gradient compression. Gradient compression is believed to be responsible for quenching in flames, because the transport of heat away from the reaction zone becomes more important than the transport of reactants to the reaction zone for Arrhenius kinetics. Simulations with temperature-dependent reaction rate do not show any reduction of reaction rate where the strain rates are high. The only qualitative difference between the temperature-dependent case and the isothermal case is that the reaction zones are initially thinner in the former case, an apparent ignition effect.

Comparison of results from the "Stripes" study with those of the "stirred" and "slab" studies shows that as one increases the scalar dissipation scale, mixing becomes more important, and the reactants decay faster. (The fraction of the domain occupied by scalar dissipation zones, and hence reaction zones, is increased.) In the case of several stripes, interference effects clearly become important after several hundred time steps, and the reaction zones lose their identity.

The studies of chemical reactions in "Forced" and "Shear" turbulence were not carried out during the Workshop (except for coding of the forcing algorithm in
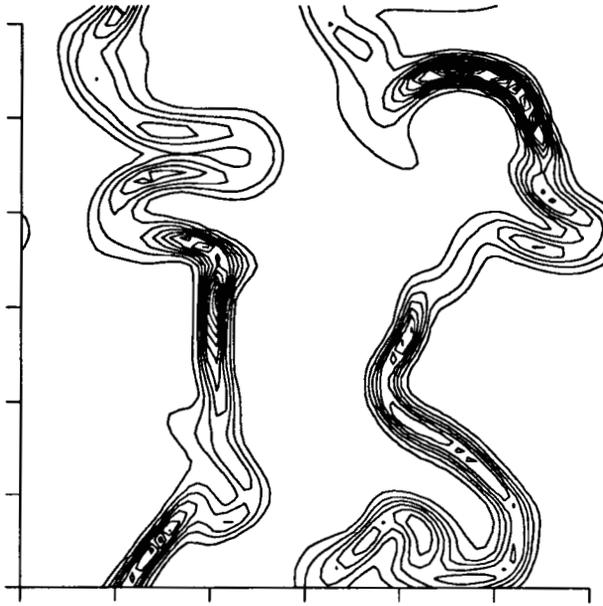
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FIGURE 2.   Contours of reaction rate, kAB, for a *slab* case at $t = 2.75$ in the plane $x = 0$.
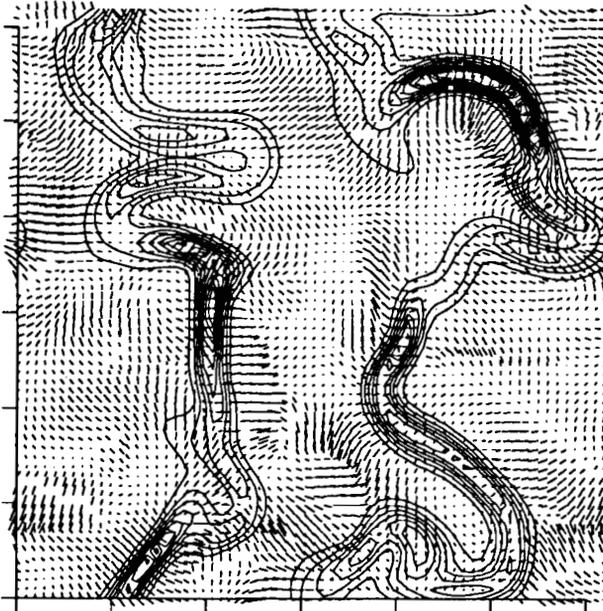

FIGURE 3.   Eigenvectors of γ, the least principal strain rate, superimposed on the contours of reaction rate from Figure 2.
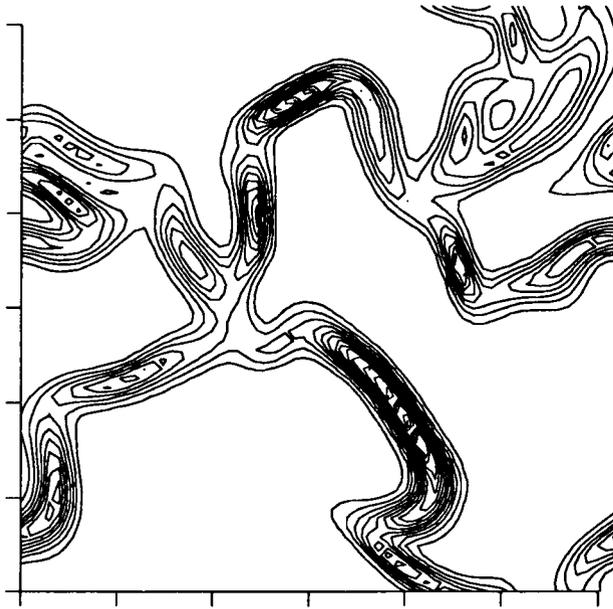
FIGURE 4. Contours of reaction rate, kAB, for a *slab* case at $t = 2.75$ in the plane of the original reaction zone, $y = \pi/2$. These data are for the same time and the same run as figure 3; only the planes observed are different.
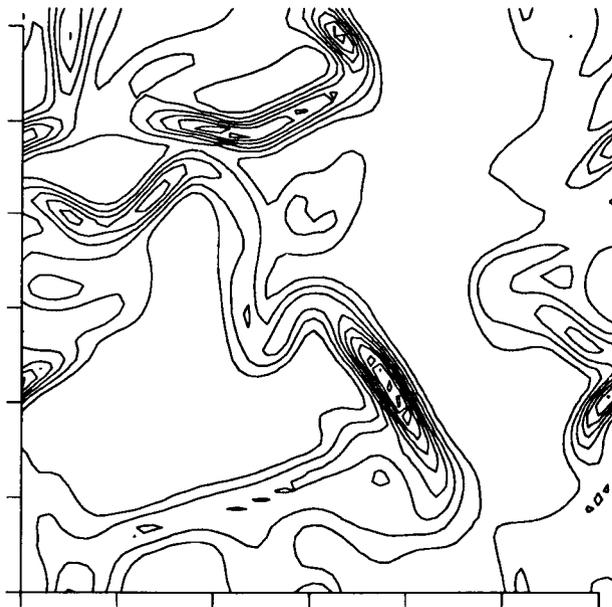

FIGURE 5. Contours of reaction rate, kAB, for a *stirred* case at $t = 3.12$ in the plane $y = \pi/2$. Coincidence, at nearly the same time, of the locations of the highest reaction rates as for the slab case in figure 4 should be noticed.

the former case and making a preliminary run at too high a Damköhler number in the latter), and those studies will be continued.

4. Conclusions

The local rate of strain in the fluid appears to be the feature of turbulence that has the most direct influence on the course of a chemical reaction, indicating that rate of strain should be incorporated into models for reacting flows. Reaction zones are aligned perpendicular to the direction of compressive strain rate, and the magnitude of the reaction rate is greatest when the magnitude of the strain rate is highest. The straining motion of the fluid compresses concentration gradients to enhance molecular diffusion, and thereby reaction rate. These observations support the simple one-dimensional model by Gibson and Libby (1972) of the reaction zone, in which the reaction surface is aligned with the most compressive strain rate, and the fluid motion is approximated by $u = -\gamma x$ in a coordinate system fixed to the reaction zone.

Preliminary examination of the saved fields suggests furthermore that the alignment of scalar gradient and strain rate directions is independent of initial conditions, and that the highest reaction rates tend to occur at the same points in the flow for reactions with different initial reactant distributions. Changing the initial scalar microscale, and hence changing the fraction of the domain occupied by reaction zones, affects the overall reaction rate and scalar dissipation rates.

The analysis of the data generated during the Summer Program is continuing, and additional simulations are being made where necessary. Further studies of non-isothermal cases are planned, as well as non-decaying turbulent flows.

REFERENCES

ASHURST, W. T., KERSTEIN, A. R., KERR, R. M., & GIBSON, C. H. 1987 *Phys. Fluids* **30**, 2343–2353.

ESWARAN, V. & POPE, S. B. 1988 *Phys. Fluids* **31**, 506–520.

GIBSON, C. H. & LIBBY, P. A. 1972 *Combust. Sci. and Technol.* **6**, 29–35.

HILL, J. C. 1988 Tutorial, 1988 CTR Summer Program.

HILL, J. C. & LEONARD, A. D. 1988 *APS Bull.* **33**, 2256.

HILL, J. C., LEONARD, A. D., & ROGERS, M. M. 1987 *APS Bull.* **32**, 2120.

KOLLMAN, W. 1987 Tutorial, 1987 CTR Summer Program.

LEONARD, A. D. & HILL, J. C. 1987 *Proc. 1987 CTR Summer Program.*

LEONARD, A. D. & HILL, J. C. 1988 *J. Scient. Computing* **3**, 25–43.

LEONARD, A. D., HILL, J. C., MAHALINGAM, S., & FERZIGER, J. H. 1988 *APS Bull.* **33**, 2257.

POPE, S. B. 1985 *Prog. Energy Combust. Sci.* **11**, 119–92.

ROGALLO, R. S. 1981 *NASA TM 81315.*