Laboratory Simulation of Flamelet and Distributed Models for Premixed Turbulent Combustion Using Aqueous Autocatalytic Reactions

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Abstract—Flamelet models have been widely applied to predict premixed turbulent combustion because of their simplicity for the description of chemical features in a turbulent flow field. We had used an aqueous autocatalytic reaction which produced an irreversibly propagating front with characteristics closely matching many of those assumed by flamelet models, to simulate premixed turbulent combustion. We then studied experimentally the influence of turbulence on this reaction-diffusion propagating front. The turbulence was generated by a pair of vertically vibrating grids in a chemical tank and was found to be nearly stationary and isotropic in the core region between the two grids, as verified by laser Doppler velocimetry. Visualization of these turbulent propagating fronts in the nearly isotropic region was obtained using the chemically reacting, laser-induced fluorescence (LIF) technique. In this paper, these planar LIF images were then processed to extract the mean reaction progress variable (c), the variance, and the probability density function (pdf) of the progress variable. Markstein numbers of these chemical fronts are probably close to unity. Results of the progress variable pdf revealed a bimodal distribution at a low turbulent Karlovitz number (Ka) with nearly zero probability of intermediate values of c. At higher Ka, the pdfs seemed to show significant probability of partially reacted fluid, in support of the theoretical description proposed by Pope and Anand (1984). Values of the turbulent burning velocities (\(S_T\)) were also extracted from successive images. When the turbulent Karlovitz number is less than \(S\), measurements of front propagation rates \(U_T = S_T \rho/\rho\) as a function of the normalized turbulent intensity \(U = u'/\rho\) show a roughly linear increase of \(U_T\) with \(U\). Values of \(U_T\) are found to be much lower than those proposed by Bray (1990) and by Pope and Anand (1984), but in good agreement with a Huygens propagation model employing renormalization group analysis. At \(Ka > 10\), \(U_T\) departs from the linearity and behaviour of these propagating fronts possibly suggests that modes analogous to distributed combustion are observed. For high \(Ka\), values of \(U_T\) are compared to a classic model. These results are also compared to premixed gaseous experiments.

Key words: Premixed turbulent combustion, flamelet and distributed models, aqueous autocatalytic reactions.

1. INTRODUCTION

The study of premixed turbulent combustion is of great practical importance because of its occurrence in spark ignition engines (Heywood 1988; Bracco 1990) and its potential for reduced NO\(_x\) emissions in some gas turbine applications (Correa 1990). Furthermore, studies of premixed turbulent flames may be central to the understanding of yet more complicated combustion phenomena (Bradley 1993).

It has been recognized for some time that different modes of combustion, such as flamelet and distributed combustion, may exist in premixed turbulent flames (e.g.

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Williams 1985; Peter 1986), probably depending on two dimensionless parameters, for example a turbulent Reynolds number \(Re_T\) and a turbulent Karlovitz number \(Ka\) if Markstein number \(Ma\) were unity. An adaptation of Eq. (23) from Bradley (1993) gives:

\[
Ka = 0.157 \frac{U^2}{\sqrt{Re_T Sc}} \quad U \equiv \frac{u'}{S_L} \quad Re_T \equiv \frac{u'l_i}{v} \quad Sc \equiv \frac{v}{\nu}
\]  

(1)

where \(U\) is the ratio of the r.m.s. turbulence intensity to the laminar burning velocity, \(Sc\) is the Schmidt number, \(l_i\) is the integral length scale of turbulence, and \(v\) and \(D\) are the representative momentum and reactant mass diffusivities. Flamelet combustion occurs at values of \(Ka \ll 1\) while distributed combustion may occur at values of \(Ka \gg 1\). In the former, the chemical reaction time is much shorter than the characteristic turbulent time scale, so that there are only two types of fluid, reactant and product, separated by a thin flame front. Some models of premixed turbulent combustion employing this flamelet concept (e.g. Pope & Anand 1984; Yakhot 1988; Bray 1990; Peters 1992; Kerstein & Ashurst 1994) predict the effect of laminar burning velocity of the planar steady flame front \(S_L\) and turbulence characteristics such as the root-mean-square (rms) velocity fluctuation of the turbulent flow \(u'\) on the mean propagation rate \(S_T\) of the wrinkled (and possibly disrupted) flame front. Note that model predictions vary considerably with no consensus on any point except that \(S_T/S_L\) increases as \(u'/S_L\) increases, indicating a need for benchmark experimental or computational data for comparison. However, experiments (e.g. Abdel-Gayed et al., 1987) do not agree closely with models. This is probably because experiments have been conducted in premixed combustible gases that do not satisfy some of the simplifying assumptions the models usually require for tractability. These assumptions may include: (I) the effect of thermal expansion can be neglected (negligible density change across the flame front), (II) heat loss is absent, (III) thermodynamic and transport properties are constant, and (IV) the turbulent flow field is homogenous, isotropic, and statistically stationary. Furthermore, direct numerical simulations have only been performed for small \(u'/S_L\) (Rutland et al., 1990; Haworth & Poinset 1992) due to numerical accuracy limitations. Recently, Shy et al. (1993) applied an aqueous autocatalytic reaction that produced an irreversibly propagating (reaction-diffusion) front with characteristics nearly satisfying assumptions (I)-(IV) to simulate premixed turbulent combustion. Thus these autocatalytic reaction fronts may be more justifiably compared to these aforementioned models, as briefly summarized below.

The propagation rates of reaction-diffusion fronts using aqueous H₃AsO₃·IO₇⁻ solutions (Hanna et al., 1982) in a nearly isotropic turbulent flow field have been measured and some of these results are reported in a recent paper (Shy et al., 1996a). Since these aqueous propagating fronts have very small exothermicity (typically 1K) and the solutions are dilute, assumptions (I) and (II) are nearly satisfied, whereas typically density decreases by a factor of 7 and kinematic viscosity increases by a factor of 25 across gaseous flame front. Due to their tiny temperature rise, the aqueous fronts are unaffected by heat losses (Shy et al., 1993) so that assumption III is nearly satisfied, whereas gaseous flame fronts are strongly affected by such losses.
(Williams 1985). Thus these aqueous fronts are more appropriate for comparison with the aforementioned combustion theories than are real gaseous flames. As already discussed in Shy et al. (1996a), Markstein numbers are probably close to unity for these chemical fronts. If $Ma = 1$ then the influence of flame stretch rate can be expressed entirely in terms of a Karlovitz number, such as that given by Eq. (1). Moreover, since $Sc \approx 500$ in this aqueous autocatalytic system whereas $Sc \approx 1$ in gases, thus the Karlovitz number criterion (Eq. 1) reveals that flamelet behavior may persist to much higher values of $u'/S_L$ in the aqueous system than in gases. Furthermore, a prerequisite for turbulent combustion study, experimental or theoretical means, is to obtain (or assume) a turbulent flow field that is homogeneous and isotropic, because otherwise the statistical stationarity of flame propagation cannot be assured. We have created such a three-dimensional turbulent flow field via a pair of specially-designed, vertically-vibrated grids in a chemical tank (see Shy et al., 1996a,b), satisfying assumption (IV). Visualization of turbulent propagating fronts in the nearly stationary isotropic turbulent flow field was via the chemically reacting, LIF technique. In this paper, we will present the mean reaction progress variable, the variance, and the pdf of the progress variable which are extracted from these planar LIF front images.

Some models of premixed turbulent combustion employing the flamelet concept characterize the structure of the turbulent flame in terms of the reaction progress variable $\phi$ modeled either by an algebraic closure (e.g. the Bray-Moss-Libby, BML, model; see Bray & Libby 1994) or from a transport equation (e.g. Pope 1987). Pope & Anand (1984) showed theoretically that for flamelet combustion, the probability density function (pdf) of the progress variable was a double-delta-function distribution, while for distributed combustion the progress variable pdf showed significant probability of partially reacted fluid. At low $Ka$, Damköhler (1940) proposed that the Huygens propagation model applies, yielding predictions such as (Yakhot 1988)

$$\frac{S_T}{S_L} = \exp[\left(\frac{u'/S_L}{(S_T/S_L)^2}\right)]$$  \hspace{1cm} (2)

and (Pope & Anand 1984; Bray 1990)

$$\frac{S_T}{S_L} = 1 + C u'/S_L$$  \hspace{1cm} (3)

for large $u'/S_L$ where $C$ is a constant (to be discussed later). At high $Ka$, Damköhler (1940) proposed that the distributed reaction zone (DRZ) model applies (Ronney et al., 1995) in which the front structure may be disrupted but turbulence influences $S_T/S_L$, mainly by increasing diffusive transport inside the broadened front without influencing the reaction rate, implying $S_T/S_L \approx \sqrt{D_T/D_L} = \sqrt{(v_T/v_L)Sc_T/Sc_L}$, where subscripts $T$ and $L$ represent turbulent and laminar values. If we assume that theoretical relations for Kolmogorov turbulence (Yakhot & Orzag 1986) may apply at least for large $Re_T$, then $v_T/v_L \approx 0.061 Re_T$ and $Sc_T \approx 0.72$ whereas $Sc_L \approx 500$. The DRZ model predicts

$$\frac{S_T}{S_L} \approx 6.5 \sqrt{Re_T}.$$  \hspace{1cm} (4)
Hence, the objectives of this paper which are clearly separate from prior studies are: (1) to provide experimental data on the mean, variance and pdf of progress variable and thus compare these results with the theoretical model of Pope and Anand, (2) to actually verify whether a steady state $S_T$ can exist, and (3) to compare our experimental data on the propagation rates with theoretical models (Eqs. 2 and 4) and with other experimental results using completely different hydrodynamic disturbances (Taylor-Couette and capillary wave flows; see Ronney et al., 1995) and thus make the analogy in these cases.

The following section reviews experimental methods used in the study, and is followed by a description of the image processing. Both are then employed to extract the mean reaction progress variable, variance, and front propagation rates, and conclusions are offered.

2. EXPERIMENTAL METHODS

2.1. A Region of Nearly Isotropic Turbulence

A flow field which is homogeneous and isotropic over many integral length scales in all three directions is the most desirable flow for studying premixed turbulent combustion. To fulfill this requirement, we introduced a vertically-vibrating-grid turbulence that has been commonly used to investigate the entrainment and mixing mechanisms across a density interface in several geophysical contexts (e.g. Turner 1973; E & Hopfinger 1986). A pair of specially-designed grids, each composed of ten rectangular bars yielding 36.9 % solidity, were concurrently vertically oscillated through the fluid to generate a region of nearly isotropic turbulence in the core region between the two grids (see Fig. 1). This double-grid turbulence generator and its corresponding flow velocities and statistics are recently described by Shy et al., (1996a,b). For completeness, we briefly summarize these results below.

Flow velocities were measured via a two-component laser velocimeter. A typical variation of the fluctuating components of flow velocity $u_i$ (subscript $i = 1, 2, 3$ representing the x, y, and z directions) along the center-line of the tank over the height $(H)$ between the two grids is displayed in Figure 2a, where $w = f S$ is the stirring velocity of the grids. It can be seen that there are two distinct flow regions: one is a near-grid flow region and the other is a nearly isotropic region. In the former region, the flow is roughly homogeneous in horizontal directions (x and y directions) but inhomogeneous in the vertical direction where the mean vertical velocities (not shown) are greater than the mean horizontal velocities. In the latter, the mean flow velocities are essentially zero and r.m.s. fluctuating velocities in all three directions are approximately equal (about 11 ~ 12% of $w$ for $H = 10.6$ cm). Other measurement points are similar. Taken as a whole, a region of roughly isotropic turbulence of about 4 cm height can be generated by concurrently oscillating the two grids with $H = 10.6$ cm, $f = 1 - 8$ Hz, and $S = 2$ cm. The variation among $u_i$ in this region is less than 15 % for such an arrangement. Also the integral length scale of turbulence, $l$, estimated from the Taylor hypothesis and the autocorrelation coefficient ($l = u' \tau$, where $\tau$ is the turbulent integral time estimated from the Eulerian autocorrelation
coefficient and $a = \sqrt{8/\pi}$ (Abdel-Gayed et al., 1987), is found to be nearly a constant about 0.3 cm, as shown in Figure 2b. The constant $l_1$ occurs because the integral time scale is found to be inversely proportional to $w = fS$ while the overall turbulence intensity is proportional to $w$. In this study, the Reynolds number based on the grid mesh size was varied from 600 to 4,800 and the ratio of $u'/S_L$ may range from about 20 to about 3,000 with an emphasis on small and moderate values of $u'/S_L$ ($< 400$) corresponding to the flamelet mode.

2.2. Chemical Apparatus

As in previous studies (Shy et al., 1993; 1996a), the chemical system we employed is the aqueous arsenous acid-iodate reaction. This reaction system consists of the
FIGURE 2. Typical characteristics of VGT apparatus. (a) r.m.s. of fluctuating velocity components $(u_1, u_2, u_3)$ and the turbulent shear stress $(-u_3w)$ along the centerline of the tank. (b) Variation of the integral length scale with the distance between the two grids where the solid and empty symbols represent the vertical and horizontal components, respectively. Both grids oscillated at $f = 6$ Hz, $S = 2$ cm, and $H = 10.6$ cm.
Dushman reaction \((\text{IO}_5^- + 5 \text{I}^- + 6 \text{H}^+ \rightarrow 3 \text{I}_2 + 3 \text{H}_2\text{O})\) followed by the more rapid Roebuck reaction \((\text{H}_3\text{AsO}_3 + \text{I}_2 + \text{H}_2\text{O} \rightarrow \text{H}_3\text{AsO}_4 + 2 \text{I}^- + 2 \text{H}^+)\). If the arsenous acid is in stoichiometric excess \(([\text{H}_3\text{AsO}_3]_0 > 3[\text{IO}_5^-]_0)\); subscript 0 represents the initial concentration value, the net reaction is Dushman reaction + 3 • Roebuck reaction or \(3 \text{H}_3\text{AsO}_3 + \text{IO}_5^- \rightarrow 3 \text{H}_3\text{AsO}_4 + \text{I}^-\). Such a reaction is autocatalytic in iodide \((\text{I}^-)\) because it catalyses the Dushman reaction. By electrochemically initiating this reaction system, propagating fronts with constant \(S_L\) can be obtained (Hanna et al., 1982).

All solutions were prepared using reagent-grade chemicals and deionized water. In this work experiments were conducted at conditions corresponding to values of \(\text{Ka}\) ranging from about 0.1 to 300, with an emphasis on values of \(\text{Ka} < 10\) because the apparent limit for flamelet behavior in these aqueous autocatalytic fronts is \(\text{Ka} \approx 5\) (Shy et al., 1993, 1996a; Ronney et al., 1995). For the aqueous reaction system, the conventional definition of Karlovitz number \(\text{(Ka)}\) requires some modification. As discussed by Shy et al. (1993), in general \(\text{Ka}\) is defined as the ratio of the mean turbulent strain rate \(\langle \text{u'/}\rangle\text{Re}_T^{1/2}\) (which can readily be shown to be equal to the strain rate at the Kolmogorov scale) to the mean chemical reaction rate \(\text{S}_L^2/\text{D}\).

This ratio can be rearranged to obtain \(\text{Ka} \approx \langle \text{u'/}\text{s}_L\rangle^2\text{Re}_T^{-1/2}\text{Sc}^{-1}\). For gaseous flames \(\text{Sc} \approx 1\) and thus the Schmidt number has been ignored in prior definitions of \(\text{Ka}\). For consistency with the definition proposed by the Leeds group, we incorporate a factor of 0.157 in the definition of \(\text{Ka}\) (Eq. 1). Alternatively, \(\text{Ka}\) is sometimes defined in terms of the ratio of the laminar flame thickness \(\sim D/S_L\) to the Kolmogorov length scale \(\sim l_\kappa \sim \text{Sc}^{-3/4}\) which can be expressed as \(\langle \text{u'/}\text{s}_L\rangle\text{Re}_T^{-1/4}\text{Sc}^{-1}\).

Thus for gaseous flames with \(\text{Sc} \approx 1\), the two definitions are equivalent if \(\text{Ka}\) is defined as the square of the ratio of the laminar flame thickness to the Kolmogorov length scale. For aqueous systems with \(\text{Sc} \gg 1\), the definition based on length scales results in a much lower \(\text{Ka}\), by a factor of \(\text{Sc}^{-1}\), than the definition based on the ratio of strain rate to chemical rate. In our studies of aqueous front propagation we have chosen to employ the definition based on rates because it would seem that flamelet-like behavior would cease if either the mean strain rate were higher than the mean chemical rate or the flame thickness were larger than the Kolmogorov length scale.

The front thickness, \(\delta\), can be estimated (Williams 1985) as \(D/S_L\) with \(D \approx 2.0 \times 10^{-5}\) \(\text{cm}^2/\text{s}\) for \(\text{I}^-\) in water (Hanna et al., 1982). Reactive solutions of \([\text{H}_3\text{AsO}_3]_0 = 0.1 \sim 0.15\ \text{M}\) and \([\text{IO}_5^-]_0 = 0.005 \sim 0.03\ \text{M}\), at a pH value of 6.8 and an initial temperature of 25°C, were used for these experiments. This arrangement provided values of \(S_L\) ranging from about 7 to \(75 \times 10^{-3}\) \(\text{mm/s}\). Thus the estimated \(\delta\) may range from about 27 to 286 \(\mu\text{m}\) which was generally smaller than the laser sheet thickness \((\approx 250\ \mu\text{m})\). As sketched in Figure 1, a 5 mm argon-ion laser beam (Coherent Innova 90–5W) was directed into the tank by means of two specially coated mirrors and a combination of spherical and cylindrical lenses, to ensure that there was little change in the thickness of the laser sheet with distance from the lenses, using similar optical lenses combination as Prasad and Sreenivasan (1990). Thus, the main limitation on the image resolution is probably the laser sheet thickness rather than the chemical front thickness.

The propagating front was detected by chemically reacting, laser-induced fluorescence (LIF) technique. Initially, a small amount of disodium fluorescein (typically
less than $10^{-5}$ M) was added and mixed in the autocatalytic solution. When excited by an argon-ion laser sheet, the dye fluoresced if the pH value of the solution was greater than 4, otherwise no fluorescence occurred. Thus the reactants (pH $\approx$ 6.8) fluoresced, and upon autocatalytic reaction, the bright reactants were converted into the dark products (pH $\approx$ 2.3) (see Fig. 1). Note that the fluorescence transition for the laser dye was occurred over a very short time scale (on the order of tens of nanoseconds; Koochesfahani 1984).

2.3. Experimental Procedures

A run began by initiating reactants at the top surface of solution using potential differences between Pt electrodes. The autocatalytic reaction then spread uniformly over the entire surface of the solution and developed into a stable downward propagating front with a constant speed. With sufficient products generated, the twin grids were then vertically oscillated in opposite phases. The front propagated downward and converted reactants into products. In the region of interest, i.e. the region of nearly isotropic turbulence, we recorded the evolution and spatial structures of the turbulent propagating front with a super VHS video system and a motor driven 35 mm camera.

2.4. Image Processing

The LIF images were digitized into $512 \times 256$ pixel arrays, with each pixel being assigned a value of brightness between 0 and 255 (8-bit). The analyzed zone corresponded to a rectangular region of 12 cm long and 6 cm high which was centered at the core region between the two grids. Each pixel had a physical space of 234 $\mu$m $\times$ 234 $\mu$m. Finer image resolution would not have been useful because the laser sheet thickness was about 250 $\mu$m.

Figure 3a displays the brightness profile along a line of pixels across the mean position of the wrinkled front at conditions of $u'/S_L \approx 47$ and $Ka \approx 0.2$. It can be seen (Fig. 3a) that the interface between reactants (high brightness) and products (low brightness) is quite sharp. At column 195 (Fig. 3a), there is a discontinuity indicating that the very sharp front is perpendicular to the line of pixels. Such a flamelet-like behaviour of the front can be sustained for values of $Ka$ up to at least 5 in this aqueous autocatalytic system (Ronney et al., 1995; Shy et al., 1996a). The histogram of pixel brightness for an entire image corresponding to the case of Figure 3a is shown in Figure 3b. The images are then digitized with $c = 0$ for reactants and $c = 1$ for products by selecting a digitized threshold value which in flamelet-like cases can be easily determined without any ambiguity. However, we agree that the appearance of sharp fronts based on the LIF image is a necessary but not sufficient condition for flamelet behavior. In comparison, Figure 4 shows the result for a higher value of $Ka$ ($\approx 18$, where $u'/S_L \approx 662$). Clearly, the interface is no longer sharp (i.e. front structures reminiscent of distributed combustion are observed), causing some doubts in choosing the digitized threshold value which in this case is selected as the brightness value at the well of the histogram ($= 64$; Fig. 4b). By thresholding, pixels with values of brightness less than 64 are equal to unity ($c = 1$)
FIGURE 3  Typical characteristics of the brightness of the LIF image for a low Karlovitz number propagating front in nearly isotropic turbulence region. (a) Horizontal profile. (b) Histogram.
FIGURE 4  Similar to Figure 3 but for a high Karlovitz number propagating front.
corresponding to products while those greater than or equal to 64 are zero \((c = 0)\) corresponding to reactants.

However, this criterion is somewhat arbitrary (the sensitivity of the threshold value will be discussed in the next section), and so far larger Ka it is expected that models of turbulent combustion developed assuming flamelet behaviour may not apply.

3. RESULTS AND DISCUSSION

3.1. Turbulent Propagating Fronts After Thresholding

As shown in Figure 3b and 4b, the probability distribution of the light intensity (the histogram of pixel brightness) for these LIF images, which looks similar to the probability density function of progress variables, exhibits a nearly double-delta function distribution for values of Ka < 5 while for Ka \(\approx 18\) there is significant probability of intermediate distribution (partially reacted fluid). These results support the theoretical description of flamelet and distribution combustion proposed by Pope and Anand (1984) and confirmed experimentally by Yoshida (1990). However, since the light intensity may not be simply related to the progress variable, the results for high Ka must be viewed at most qualitatively and with caution.

Four instantaneous, 2-D images obtained after thresholding procedures for varying values of \(u/S_L\) and/or Ka are shown in Figure 5. It can be seen that the propagating front becomes more and more wrinkled as Ka increases. At Ka \(\approx 2\), it is possible to observe some islands or pockets of unreacted material (the reactant), where the propagating front still remains sharp. As Ka increases further (above 10, shown in Fig. 5d), many islands of reactants as well as pockets of products can be observed in the 2-D image. This seems to be consistent with the observation (Shy et al., 1993) that only at high Ka does local quenching occur and permit islands of products to form. As explained by Ronney (1995), only when local front quench occurs can islands of products occur. However, in order to truly confirm the above island formation, observation images in the third direction have to be obtained. This is currently being studied (by the first author) using a 3-D successive planar LIF imaging of a synchronized raster swept laser beam, combined with a very fast data acquisition system, and these results will be reported elsewhere in the near future.

3.2. Mean Reaction Progress Variable and Variance

Figure 6 displays the profiles of the mean and variance of the progress variable \(c\) for four different values of Ka and \(u/S_L\) corresponding to those in Figure 5. Here the coordinate \(z\) is measured from the top of the binarized image (Fig. 5) and normalized by \(l_i\) (Fig. 2b). For all values of Ka studied here, the fronts propagate to the right into reactants \((c = 0)\), leaving products \((c = 1)\) behind it. The variance \((c - \bar{c})^2\) is zero outside these propagating fronts where \(\bar{c}\) is either zero (reactants) or unity (products); the overbar represents the ensemble averaged. The variance reaches a peak close to where \(\bar{c} = 0.5\). If the front were infinitely thin, then the maximum value of the variance would be 0.25 at \(\bar{c} = 0.5\) (Fig. 6).
FIGURE 5  Typical images of turbulent propagating fronts after thresholding. Black represents products and white is reactants, with 60 mm × 120 mm field of view. (a) $u'/S_c \approx 47$ and $Ka \approx 0.2$; (b) $u'/S_c \approx 116$ and $Ka \approx 1.0$; (c) $u'/S_c \approx 143$ and $Ka \approx 2.0$; (d) $u'/S_c \approx 662$ and $Ka \approx 18$. 
FIGURE 6

(a) $\text{Ka} = 0.2$

(b) $\text{Ka} = 1.0$
FIGURE 6 Mean and variance of progress variable against normalized distance for four different values of $Ka$, corresponding to Figure 5.
As can be partly seen in Figure 6, the span of the mean and variance of the reaction progress variable, which can be viewed as an indication of the flame brush thickness ($l_{FB}$) defined as the width of the variance at 0.125 (one half of the maximum value of the variance), increases rapidly from about 1.6 integral length scale of turbulence ($l_i$) to about 4.8 $l_i$ as Ka increases from 0.2 to 4. When Ka increases up to 5 or above, $l_{FB}$ only increases slightly and reaches to about 5.2 $l_i$ at Ka $\approx 18$ (Fig. 6d). For the flamelet mode (Ka < 5), the profiles of $\bar{c}$ and $(c - \bar{c})^2$ look roughly symmetric with respect to the location where the variance is at its maximum value. However, for large values of Ka (such as Ka $\approx 18$; Fig. 6d), the profiles of $\bar{c}$ and $(c - \bar{c})^2$ depart from the symmetry, rapidly approach their boundary values on the product side ($\bar{c} = 1$ and $(c - \bar{c})^2 = 0$), and act slowly on the reactant side ($\bar{c} = 0$ and $(c - \bar{c})^2 = 0$). This result is in good agreement with the behaviour proposed theoretically by Pope and Anand (1984). Again we note that the current aqueous chemical system is not an Arrhenius reaction system with high activation energy, so that results for higher values of Ka (> 5) have to viewed at most qualitatively and with caution. Also $\bar{c}$ and $(1 - \bar{c})$ is essentially equal to the variance, $(c - \bar{c})^2$, within experimental uncertainties, because the front interface is assumed to be infinitely thin. Thus progress variable variance against mean reveals a parabolic curve with the maximum value of the variance $(c - \bar{c})^2 = 0.25$ at $\bar{c} = 0.5$, in excellent agreement with the theoretical models proposed by Pope and Anand (1984).

**FIGURE 7** Mean progress variable against normalized distance for three different thresholding values in mode similar to distributed behavior (Ka $\approx 18$), indicating the sensitivity of thresholding values.
3.3. The Sensitivity of the Threshold Value

In order to test the sensitivity of these results to threshold values, the effect of smaller or larger values of the brightness threshold were tested (60 or 68) (see Fig. 4b). Figure 7 displays the mean progress variable against the normalized distance for three different threshold values (60, 64 and 68; see Fig. 4b). It is found that changing the threshold value, either close to the reactant side or close to the product side, does not alter the primary shape of \( \tilde{c} \), indicating the insensitivity of the results to the threshold value even for the value of \( \text{Ka} \) as large as 18.

3.4. Verification of a Steady-State \( S_T \)

Typical digitized images of the turbulent front propagation through the nearly homogeneous isotropic turbulence region, the region in which \( S_T \) was inferred, are shown in Figure 8 for \( \text{Ka} \approx 0.2 \). In order to verify whether a steady state \( S_T \) can exist in this chemical system, we measured a mean product creation height \( (h) \) which was the products (dark) area per unit tank width measured from the middle plane of the upper grid as a function of time using a simple binarized area counting program. \( S_T \) was then defined as the mean vertical propagation rate of product \( (dh/dt) \), similar to the conventional definition of \( S_T \), since \( S_T \) multiplied by the cross-sectional area of the tank represented the volumetric rate of product creation. In Figure 8, \( t_0 \) is the time period measured from starting the vibrating grids to the mean front just reaching the upper boundary of nearly isotropic turbulence region. Each binarized

\[ z \text{ (cm)} \]

\[ t \] The middle position of the upper grid

\[ 0 \]

\[ 2.3 \]

\[ 8.3 \]

\[ 12 \text{ cm} \]

\[ 4 \text{ cm} \]

\[ 4.16 \text{ s} \]

\[ 9.37 \text{ s} \]

\[ 17.46 \text{ s} \]

\[ 24.27 \text{ s} \]

**Figure 8** Typical binarized image sequence of propagating front in nearly-isotropic turbulence for \( \text{Ka} \approx 0.2 \) and \( u/S_x \approx 47 \). Field of view 60 mm \( \times \) 120 mm.
image was cut in two from the center of the image and estimated $S_T$ separately to test the symmetry of front propagation. Figure 9 shows that an essentially steady state $S_T$ does exist in this chemical system and that the front propagation is almost symmetric with respect to the centerline of the chemical tank.

3.5. Front Propagation Rates

The effect of $u'/S_L$ on $S_T/S_L$ was measured for a 3-decade range of $Ka$, corresponding to values of $u'/S_L$ between 38 to 2849. It was found that at moderate $Ka$ ($<5$), the determination of $S_T$ is quite accurate because the front interface is very sharp, but at higher $Ka$ ($Ka > 10$) the front interface becomes diffuse and “fuzzy”, suggesting a transition from flamelet to distributed behavior. Typically at large values of $Ka$, there are two possible binarized threshold values to be chosen, corresponding to the upper and lower boundaries of the fuzzy interface. The variation of $S_T$ using the above two different threshold values are found to be no more than 5%. This indicates that the average thickness of the broadened front may remain roughly constant at a given $Re_T$ and $Ka$ (Shy et al., 1996a).

Figure 10a shows values of $S_T/S_L = U_T$ as a function of $u'/S_L = U$ and compares experimental results for $Ka < 5$ with analogous theoretical results (Pope & Anand

![Image](image_url)
1984; Bray 1990; Yakhot 1988; Shy et al., 1993). Two features of these data should be noted. First, the experimental data on $U_T$ are in good agreement with Eq. (2), the prediction of Yakhot's renormalization group theory. This agreement is very good for mixtures with higher $S_L$ at lower $u'/S_L$, but less satisfactory with lower $S_L$ at higher $u'/S_L$, suggesting a strain rate (Karlovitz number) effect on $S_L$. Second, measurements of $U_T$ are much higher than that predicted by Shy et al., (1993) for front propagation through an array of singlescale vortices generated by a Taylor-Couette flow, while much lower than those by the presumed-pdf method of Bray (1990) and the joint-pdf theory of Pope and Anand (1984). The latter point is probably due to the fact that these theories incorporated certain assumptions regarding the statistical properties of the flow or some empirical constants taken from gas combustion experiments which naturally exhibited specific velocity spectra. As values of $K_a$ increase up to 10, values of $U_T$ tend to bend down from the Yakhot's prediction. Such a departure of the linearity of $U_T$ as a function of $U$ is even more pronounced for $K_a > 10$ (the small diagram in Fig. 10a; see Shy et al., 1996a).

Figure 10b displays the ratio of experimental values of $U_T$ to the theoretical predictions of Eq. (2) as a function of $K_a$. Values of $U_T$ for $K_a < 10$ are in good agreement with the predictions of Eq. (2), and the ratio of experimental to theoretical values has a mean of 1.13 and rms deviation of 0.36. This good agreement at low $K_a$ (flamelet mode) is probably because Eq. (2) is based on an exact equation for Huygens propagation model (Kerstein et al., 1988) and does not require empirical

![Figure 10](image-url)
FIGURE 10  (a) Effect of turbulence intensity on the simulated turbulent burning velocity and comparison with predictions of some flamelet models of turbulent combustion and a model of front propagation in a one-scale flow (Shy et al., 1993). These dark circles represent previous results (Shy et al., 1995a) while these white circles are the new results. (b) Comparison of measured $S_{T}/S_{L}$ values to a Huygens propagation model (Eq. 2). (c) Comparison of measured $S_{T}/S_{L}$ values to a distributed reaction zone model (Eq. 4).
constants extracted from gaseous combustive experiments that may not satisfy assumptions (I)-(IV). In an independent, separated study using completely different stirring methods, Ronney et al., (1995) found the similar results, suggesting that velocity spectra may have only a weak effect on \( U_T \) as long as the flow has broad enough spectra of flow scales. While it is not yet known how broad a range of scales is required for scale-independence, Ronney (1995) has proposed a speculative criterion based on the slope of the conventional kinetic energy density vs. wavenumber plot. It was estimated that a rather steep slope of \(-4\), compared to \(-5/3\) for classical Kolmogorov turbulence, would be required before the range of scales would be narrow enough that \( S_T \) would exhibit an appreciable scale-dependence. To date no experimental or numerical studies have been performed that would provide a test of this proposition.

Figure 10c shows the ratio of experimental data of \( U_T \) to the distributed reaction zone (DRZ) prediction (Eq. 4) for \( Ka > 10 \). Experimental data are roughly consistent with DRZ predictions at \( Ka > 10 \) except a factor of 5 higher; the ratio of experimental values of \( U_T \) to theoretical predictions has a mean of 5.14 and rms deviation 0.68. In comparison, there is no evidence from gaseous combustible flame fronts that Eq. (4) is valid. This might be due to the factor that Eq. (4) was derived by assuming that the chemical reaction is not influenced by turbulence. On the other hand, local reactions of gaseous flames with large activation energy are sensitive to small fluctuations in local temperature due to turbulence which may result in a largely nonlinear change in the local reaction rate and thus affect its mean value (Ronney et al., 1995). Such a strong nonlinearity is absent in the aqueous \( H_3AsO_3\cdot1O_7 \) system (Hanna et al., 1982).

Because of the absence of heat loss influences and the lack of strong nonlinearities in reaction rates along together with constant density and large ranges of \( Ka \) and \( U \) that we are able to employ, our experiments might be the first that could be directly compared to the DRZ model proposed by Damköhler (1940). In different turbulent flows (both Taylor-Couette and capillary wave flows), Ronney et al., (1995) found that their experimental results were in good agreement with Eq. (4). Thus the factor of 5 discrepancy between our high-\( Ka \) experimental data and the DRZ model in \( U_T \) (Fig. 10c) is surprising and the reason for such a discrepancy is not clear at this moment. However, we anticipate three possibilities: (1) The DRZ model, like all models, is speculative, especially the factor 6.5 in Eq. (4) that was obtained from the assumptions of \( \nu_T/\nu_L \approx 0.061 \) \( Re_T \) and \( Sc_T \approx 0.72 \) (see Yakhont and Orszag 1986). (2) Due to the practical limit of the size of the tank, the turbulent Reynolds number based on the integral length scale of turbulence may be not large enough. (3) While \( l_i \) was measured in the same manner as employed in prior studies of turbulent premixed flames (see section 2.1), the resulting value of \( l_i \approx 0.3 \) cm is considerably smaller than the maximum size of flame wrinkling (\( \approx 3 \) cm) seen in Figure 5. Some studies in gaseous flames (Mantzaras et al., 1988; North and Santavicca 1990) and autocatalytic fronts (Haslam and Ronney 1995) have suggested that \( l_i \) is the outer cutoff scale for front wrinkling. If \( l_i \approx 3 \) cm were tentatively used to estimate \( Re_T \) and thus \( S_T/\delta_T \) in the DRZ regime, the agreement between model and experiment would be considerably improved at which the mean of the ratio of experimental to theoretical values would drop from 5.14 to 1.63. Detailed measurement of front
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wrinkling scales relative to turbulence scales is deferred to a future study. Nevertheless, Figure 10c is presented here in the hope that it may serve a constructive purpose by stimulating additional research and discussion.

CONCLUDING REMARKS

Experiments of these aqueous autocatalytic fronts propagating in a nearly isotropic turbulent flow field have been conducted to obtain velocity and front surface property data, thereby enabling comparison with flamelet models. Our concluding remarks are: (1) The present work supports the theoretical view of flamelet and distributed combustion proposed by Pope and Anand based on a mean progress variable; (2) For flamelet mode of combustion, our results indicate that the velocity spectrum has an effect on ST/SL', even at a fixed U'/SL', and values of ST/SL are in good agreement with Yakhot's prediction; (3) Values of U'/SL can be obtained much higher than those attainable in gaseous combustion experiments, possibly suggesting the resilience of wrinkled flamelets to turbulence and the importance of heat loss.

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