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Coalescence/Dispersion Modeling of Turbulent Combustion in a Jet-Stirred Reactor

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The results of a direct simulation of high-intensity methane/air combustion are compared to previously reported measurements of OH and O atom concentrations in a jet-stirred reactor. The simulation uses a zero-dimensional coalescence/dispersion model that requires only the initial reactor feed conditions, an appropriate chemical rate mechanism, and estimates of the fluid mixing time scales. Empirical time scales, determined by forcing agreement between simulation and experiment, are of the same order of magnitude as those estimated from either global fluid dissipation length and velocity scales or inviscid arguments based on reactor length and velocity scales. Simulations using these estimates satisfactorily represent the measured temperature and species profiles over a wide range of fuel-air equivalence ratios and, in direct contrast to perfectly stirred reactor predictions, show close agreement with the measured values near stoichiometric conditions.

Nomenclature

c = concentration
C/D = coalescence dispersion
L = reactor length scale
M = total mass flow rate
N = number of Monte Carlo particles
$\dot{N}$ = time rate of change of N
N_s = number of time series samples
$P(c)$ = concentration probability density function
$P_0(c)$ = reactor inlet stream concentration probability density function
pdf = probability density function
PSR = perfectly stirred reactor
r = net rate of chemical reaction
R_e = Reynolds number based on mean reactor conditions
T = mean reactor temperature
t_d = dissipation time scale (based on reactor conditions, see Table 1)
t_e = eddy roll-up time (based on inviscid arguments, see Table 1)
t_m = macrotime scale (based on global length and velocity scales, see Table 1)
t_0 = microtime scale (based on Kolmogorov length scale, see Table 1)
U = mean inlet jet velocity
V = reactor volume
α = total shear layer growth angle
β = rate of binary encounters normalized by the number of Monte Carlo particles (N), coalescence/dispersion mixing frequency
ϕ = fuel-air equivalence ratio
ρ = mean reactor density
θ = angular divergence of reactor walls

Introduction

HIGH-INTENSITY combustion processes may be generally characterized by a high fluid turbulence level and, for most practical fuels, rapid chemical reaction rates. In fact, a continuous and stable ignition of common exothermic fuels is often achieved by "backmixing" the products of combustion with the incoming fresh fuel/air mixture. The hot products provide not only local sensible energy, but also reaction intermediates that further enhance the fast reaction kinetics. In such situations, the rates of fluid mechanical mixing and chemical species production may be comparable, so that both processes and their mutual interaction must be modeled to satisfactorily predict combustor performance. For these reasons, significant research efforts have been directed toward the investigation of turbulent combustion and the strongly coupled interaction of mixing and chemical processes.

Most of the available reactor models assume that the reactants are either mixed instantaneously at the molecular level (micromixed) or are mixed as independent segregated elements (macromixed), thus avoiding complex fluid dynamic calculations. Each approach provides a description of a useful limiting case, but neither adequately predicts combustion performance over a wide range of operating conditions. Furthermore, while detailed numerical treatment of the coupled chemistry and fluid mechanics may now be computationally possible, it is often not economically feasible.

An alternative method that has shown promise not only because of its straightforward implementation, but also because of its physically plausible basis, is the *C/D* formulation first introduced by Curl[1] to describe the concentration probability density distribution function of liquid droplets mixing in an emulsion,

$$\frac{\partial}{\partial t}P(c)=\frac{1}{t_s}[P_0(c)-P(c)]-\frac{\partial}{\partial t}[rP(c)]+8\beta\int_0^c P(c')P(2c-c')\mathrm{d}c'-2\beta P(c) \tag{1}$$

In order to demonstrate the effects of imperfect mixing in gas phase combustion, Evangelista et al.[2] applied perturbation methods to solve Eq. (1) as the ratio of fluid mixing rate to inlet mass flow rate becomes large. Spielman and Levenspiel[3] first applied Monte Carlo techniques to the solution of Eq. (1) and the natural coupling of Curl's mixing model with the random stochastic nature of Monte Carlo methods has subsequently received widespread use.[4,5] Other researchers[6-10] have recently solved the pdf equations for turbulent flows and have suggested improvements in Curl's formulation in order to satisfactorily model the molecular mixing terms.

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In this work, the results of a *direct simulation* which utilizes Curl's *C/D* approach are compared to previously reported measurements of OH and O atom concentrations in high-intensity methane/air combustion.[11,12] Malte et al.[12] have shown that a fully micromixed, PSR model poorly predicted experimental results near $\phi = 1$. In order to explain this discrepancy, they analytically examined the influence of finite-rate mixing in the experimental jet-stirred reactor. By using a first-order kinetics, constant properties PSR analysis,[13] fluid mixing rates were shown to have significantly influenced the predicted concentrations near $\phi = 1$. The conflict between the PSR model and measurement thus provides an appropriate test of the finite-rate mixing, finite-rate chemistry *C/D* simulation. A synopsis of the experimental setup used in Refs. 11 and 12 is presented below. It is then followed directly by a discussion of the numerical approach, a comparison of the *C/D* simulation results and measurements, and the conclusions of this study.

Experimental Setup

Measurements of OH and O atom have been made for a range of fuel-air equivalence ratios in the jet-stirred reactor sketched in Fig. 1.[11,12] Similar measurements of primary combustion species (CH_4, CO_2, CO, and H_2O) were not reported. Although the details of the reactor design, fuels, flow conditions, and experimental techniques are fully described in the referenced publications, pertinent aspects of the studies will be briefly discussed.

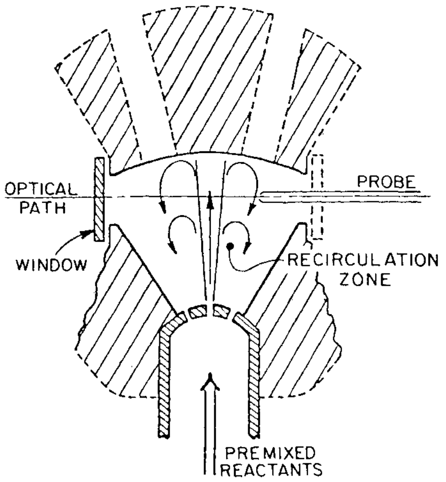

Fig. 1 Jet-stirred reactor (adapted from Ref. 12).

The experimental reactor had entrance to turning wall distance of approximately 4 cm, seven inlet orifices with equal diameters of 0.074 cm, and wall divergence of 30 deg. The translational temperature of the recirculation region was measured with a coated Pt/Rh thermocouple and found to be nearly uniform across the reactor. The uncorrected thermocouple temperature was less than the measured OH rotational temperature by approximately 4% for fuel-lean conditions and approximately 6% for fuel-rich conditions. Time-averaged OH concentrations were determined using multipath absorption techniques in the near-homogeneous recirculation regions of the reactor. The relative O atom concentrations were inferred from simultaneous gas sampling measurements of CO and the continuum chemiluminescense resulting from the reaction $CO + O \rightarrow CO_2 + hv$.[14,15] One of the major experimental uncertainties associated with these measurements was the concentration nonuniformity introduced by the feed jets. Although their fractional volume was small, numerical predictions of a similar flowfield[16] have shown that significant concentrations of CO and OH within the jets could have adversely affected the line-of-sight measurements. Assuming homogeneous conditions, Malte et al.[11] estimated a measurement uncertainty of ±30% and ±60% for the OH and O atom data, respectively.

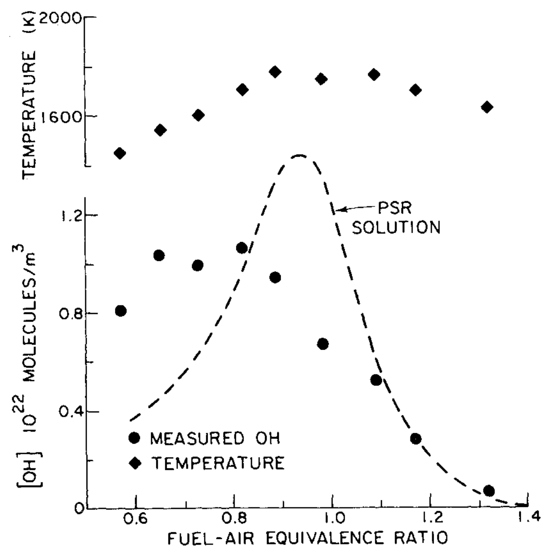

Fig. 2 Measured and predicted [OH] and uncorrected thermocouple temperature (adapted from Ref. 12).

Absorption measurements of OH and uncorrected thermocouple temperature for a range of ϕ at a reactor loading of 19 kg/m^3·s are reproduced in Fig. 2. The rotational temperature was taken to be the more accurate estimate of the actual reactor temperature and was used in the PSR calculations (dashed line). Figure 3 displays the measured O atom concentration for the same reactor conditions. The solid curve was calculated assuming partial equilibrium for the reaction $H_2O + O \Leftrightarrow OH + OH$, while the dashed line once again indicates PSR values. The "triangles" represent direct chemiluminescence data, while the "squares" show data obtained by the "slope method" (doping the inlet flow with CO and observing the change in chemiluminescence).[11,12] The solid and open symbols indicate the presence or absence of windows, respectively. The large scatter in the O atom data for $\phi < 0.8$ was attributed to a strong sensitivity of the measurement to reactor temperature fluctuations. More importantly, the influence of finite-rate mixing near $\phi = 1$ is clearly evident. When compared with the PSR predictions, the measured OH and O profiles exhibit significantly reduced maximums at lower values of ϕ.

Numerical Approach

The *C/D* technique used in this work employs a Monte Carlo mixing model to simulate turbulent mixing processes in a constant-pressure jet-stirred reactor. The numerical approach is in the spirit of a *direct simulation* of the reactor physics. The mass flow rate of fresh reactants is assumed constant and is represented by a discrete number flow rate of equal mass "particles" given by

$$\dot{N} = \dot{M}N/\rho V = N/t_s \tag{2}$$

During their transit through the reactor volume, the particles undergo continuous adiabatic chemical reaction and mix (coalesce and disperse) according to Curl's model.[1] In this model, mixing pairs are selected at specified time intervals with uniform probability (no age bias). During each *C/D* event, the extensive properties of each particle pair are simply averaged, while between *C/D* encounters, the physical properties of each particle are determined using the batch integrator code CREKID[17] and the full CH_4 kinetics description employed by Malte et al.[11,12,18-20] Assuming constant-pressure combustion, the particle removal rate is determined by the instantaneous reactor density. As with the mixing process, the exiting particles are chosen without age bias. Thus, for a given chemical mechanism and initial conditions, only the *C/D* mixing frequency β need by specified.

Initial Conditions

The choice of the initial reactor state for the *C/D* simulations is nontrivial and is the subject of continuing study. Its importance is derived from the dependence of flame stability not only on the presence of sufficient thermal energy, but also on sufficient concentration of chemical intermediates to sustain local chemical reaction. This is particularly crucial when the reactor conditions are close to ignition or incipient extinction. For the simulations reported here, the inlet flow velocity was assumed sonic at 3.2×10^4 cm/s and the nominal growth angle of each primary jet was taken as 30 deg. The particle concentrations were initialized by assuming a chemically equilibrated distribution of mole numbers and corresponding temperatures. This choice is computationally expedient since the physical properties of each particle need not be determined until it mixes with a particle having nonequilibrium properties. However, since the concentrations of intermediates (eg., OH, O, H) at equilibrium are significantly lower than those present during steady-state combustion, and since the incoming premixed particles have no chemical intermediates, the simulated ignition process was *entirely thermal* in character. This presented difficulty only at $\phi = 0.6$, where the simulation failed to achieve a stable condition. In previous work, actual blowout was measured for CH_4/air combustion at $\phi = 0.55$ and a loading of 40 kg/m$^3 \cdot$s for a similar reactor[21] and was estimated using a PSR model to occur at $\phi = 0.5$ for the conditions of this study.

Estimates of β

From a strictly empirical point of view, β may be thought of as that frequency which produces reasonable agreement between the numerical *C/D* predictions and measurement. With regard to the *C/D* simulation, two physical limits of β are clear: 1) when β is zero, the particles are segregated and remain independent of all others; and 2) when β approaches infinity, the simulation replicates the PSR model. The appropriate values must lie somewhere between these two limits, since the initially segregated particles (with dimensions characterized by the inlet macroscale) are reduced via fluid/mechanical processes to the molecular scale at which micromixing and reaction occur.

Turbulent dissipation length scales[22,23] and inviscid geometric scales[24] based on the experimental data have been used to estimate the mixing time scales and a concomitant value of β. Five elementary time scales have been identified:

1) t_0, a microtime scale associated with the Kolmogorov length scale.

2) t_e, a characteristic time for an eddy to make one complete rotation.

3) t_m, a macrotime scale based on the round trip time of a fluid element in the reactor.

4) t_d, a time scale based on the dissipation length scale.

5) t_s, the average residence or stay time of a fluid element in the reactor.

The time scale parameters along with numerical values based on the experimental conditions are summarized in Table 1.

Table 1 Reactor time scales

Time scale	Parameters	Value, ms
t_0	$2LR_e^{-3/4}/U$	1.3×10^{-4}
t_e	$2L\tan(\alpha/2)/U$	6.5×10^{-2}
t_m	$8L/U$	1.0
t_d	$(2t_sL^2/U^2)^{1/3}$	6.5×10^{-1}
t_s	$2\pi\rho L^3(1-\cos\theta)/3\dot{M}$	9.0

For the conditions of interest, t_0 is two orders of magnitude less than t_e, which, in turn, is one order of magnitude less than t_m or t_d. Finally, based on estimates of t_s, a fluid element would make, on average, nine round trips before exiting the reactor.

C/D simulations were carried out to empirically determine a mixing frequency appropriate for the CH_4/air combustion experiments. Reasonable agreement with measured *T*, OH, and O atom and data were established by numerically varying β with $\phi = 1.0$. As might be anticipated, a unique value of β for all of the measured parameters was not established. However, considering the reported measurement uncertainty, the appropriate choice of time scale was found to be on the same order as either t_m or $t_d \sim 1$ ms. This is consistent with the assumption of homogeneous combustion, e.g., the reactor response to changes in the inlet stream composition must be related to the time required for the incoming fluid to occupy the entire volume. Much as the slowest step of a chemical reaction mechanism determines the overall rate of a conversion, the slowest step of the mixing-scale reduction process strongly influences the overall mixing rate. With this in mind, and with due consideration given to the uncertainty of the measurements, $\beta \sim 1/t_m \sim 1000$ Hz was chosen for the *C/D* simulations.

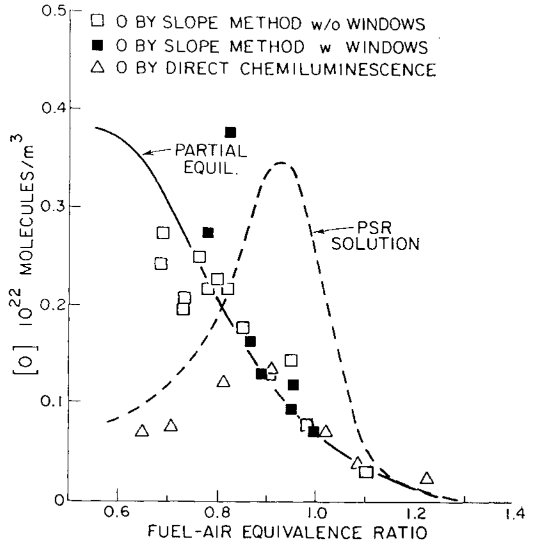

Fig. 3 Measured and predicted [O] (adapted from Ref. 12).

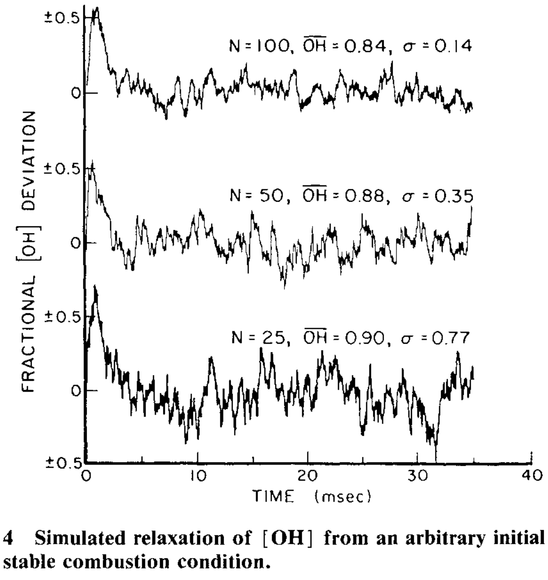

Fig. 4 Simulated relaxation of [OH] from an arbitrary initial state to a stable combustion condition.

C/D Model Limitations

The application of Curl's mixing model (binary mixing and uniform selection probability) provides significant computational advantages, but questions have been raised concerning the validity of the predicted species pdf's. By considering the case of decaying fluctuations of a passive scalar in a fixed volume, Pope[25] has clearly shown that simple binary mixing produces discontinuous pdf's and that the selection of *C/D* pairs without age bias does not ultimately lead to stationary Gaussian distributions. Several researchers[9,10,25] have demonstrated that discontinuities in the concentration pdf's can be avoided if a continuous pdf is used to control the extent of the mixing. It should be noted, however, that the requirement of smoothness is derived from the nature of physical processes and may not be required to provide accurate numerical resolution. In order to better approximate a stationary Gaussian distribution, Pope further suggested that the particles be selected to mix according to an age-biased pdf.[25] Improvements to Curl's approach may thus be achieved (in principle) by choosing two additional pdf's: one to select the mixing pair and one to determine the extent of mixing.

The difficulties resulting from potentially nonphysical concentration pdf's are addressed in the present work using an approach different from that suggested by Pope.[25] Starting from prescribed initial conditions, a sequence of *C/D* events is generated to simulate the combustion process. The reactor particle population is numerically sampled at equal time intervals to determine the time-dependent distribution of reactants. Each simulation is allowed to run several residence times beyond the point at which the mean and variance stabilize. This stable condition is generally achieved in one or two fluid residence times (on the order of 20 ms). The number of samples is determined by the product of the *C/D* interaction rate, the number of particles, and the number of mean residence times over which the sample is taken ($N_s \sim \beta N t_s$). If N_s is sufficiently large, the time series statistics of the ensemble will be Gaussian, even though the instantaneous species distributions are not.

The choice of *C/D* particle number density thus directly affects the accuracy of the simulation. The probability approach and the stochastic particle model become equivalent only as the number of particles approaches infinity.[26] Since for practical *C/D* computations, N must remain finite, some degree of statistical uncertainty is unavoidable. Nguyen and Pope[27] addressed a similar problem when using Monte Carlo techniques to solve the pdf equations for the transport of a conserved scalar in a turbulent diffusion flame. In their work, equilibrium chemistry (which implies infinite chemical rates) was assumed and Curl's model was used for the molecular mixing term. This must be contrasted to the jet-stirred reactor flowfield in which mixing and chemical energy release rates are comparable. Their numerical experiments indicated that the mean value of the scalar was relatively insensitive to N, that the standard error varied approximately as $N^{-½}$, and that the CPU time varied directly with N. It is important to note, however, that "particle density" N is used differently in Nguyen and Pope's formal Monte Carlo solution and the simulation technique presented here. In the former, N is not physically tied to the mass of the system, whereas in this study, N and ρ are directly related. Although the effect of mass discretization is not directly assessable, qualitative evidence of the dependence on N can be demonstrated. Figure 4 presents the relaxation of the reactor OH concentration as it changes from an arbitrary initial state to a stable combustion condition. The general properties of this process are also reflected in the simulations of other major species. For each value of N, the fractional deviation of OH has been normalized by its final time-averaged value, i.e., $\Delta OH = 1 - [OH]/[\overline{OH}]$. If N were sufficiently large, the fractional deviations of all species should approach zero as a stationary distribution is established. However, since a slow time dependence is demonstrated in Fig. 4, a true steady-state distribution has not been achieved.

The final time-mean value of OH and its associated standard deviation are also in Fig. 4 for each value of N. While the time average shows a slight sensitivity to N, the standard deviation varies approximately as N^{-1}. As the simulations illustrate, the amplitude of the low-frequency oscillation diminishes as N increases, but the time dependence remains essentially the same. This long-term nonsteady behavior is thought to result from the choice of physical model and, to that accuracy, satisfactorily represents the combustion process. The high-frequency, low-amplitude ripple that tracks the slower variation can be shown to be a direct result of the discretized inlet particle flow rate. The ripple frequency thus scales as N^{-1} and is a measure of the minimum simulation time scale that can be resolved. The uncertainty introduced by the choice of N is therefore reduced as N increases. The simulation results presented below have been produced using a microcomputer with 256 Kbytes of internal memory. This limited the maximum reactor particle population to 40 and the minimum resolvable time scale to approximately 0.25 ms. Although the confidence level assigned to the results must therefore be qualified, the predicted magnitudes and trends are physically consistent and will be shown to be reasonable estimates of the system behavior.

Comparison of *C/D* Simulations and Combustor Measurements

Coalescence/dispersion simulations have been carried out over the range of fuel-air equivalence ratios investigated in the previous experimental work.[11,12] The numerical results are plotted in Figs. 5 and 6 with the corresponding experimental data for T, OH, and O, and PSR predictions. As explained above, each simulation was initialized with chemically equilibrated particles and the *C/D* mixing frequency β was fixed at 1000 Hz. The reactor statistics at each value of ϕ were obtained by sampling over a time interval of not less than $2t_s$ after a stable state was realized. Time averages of the time-dependent ensemble mean are shown as X's.

The comparisons of temperature and OH concentration for $0.7 < \phi < 1.4$ shown in Fig. 5 are satisfactory. The uncorrected thermocouple temperature is shown along with PSR predictions and estimates of the OH rotational temperature.[11] Differences between the OH rotational temperatures and the *C/D* results are within 5% for the full range of equivalence ratios. Moreover, the agreement is potentially better than indicated since the present simulations assume adiabatic reactor condi-

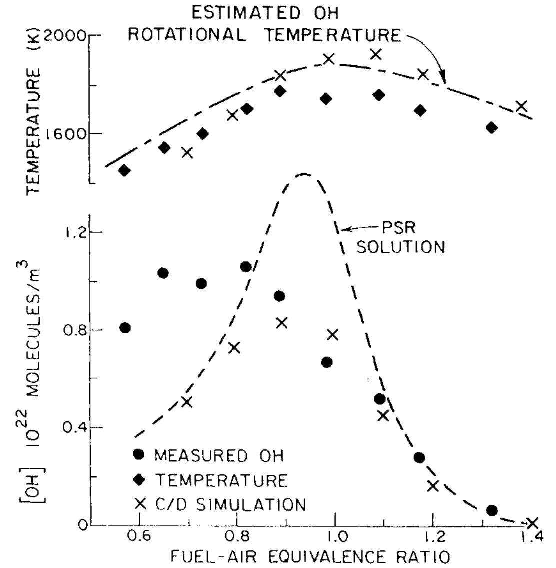

Fig. 5 Comparison of *C/D* simulation results with measured and predicted [OH] and uncorrected thermocouple temperature from Ref. 12.

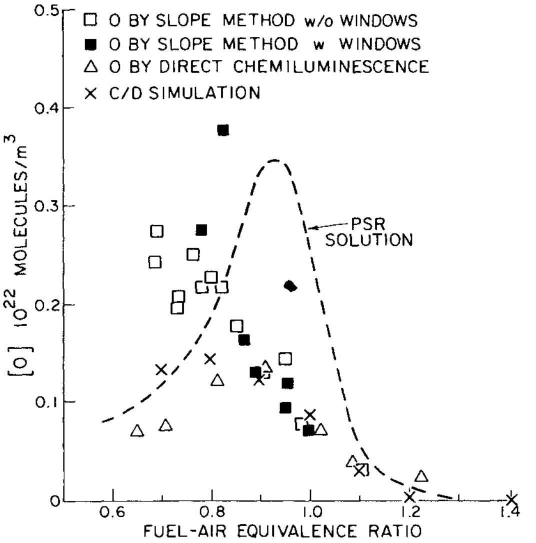

Fig. 6 Comparison of *C/D* simulation results with measured and predicted [O] from Ref. 12.

tions. The simulated and measured OH concentrations agree with the PSR solution for fuel-lean and fuel-rich conditions. In this regard, both the PSR and the *C/D* simulation underpredict the measured fuel-lean values. However, for ϕ near 1.0, the *C/D* simulation closely follows the data and does not replicate the PSR peak.

In Fig. 6, the comparison of simulated O atom concentrations with that measured by chemiluminescence techniques indicates satisfactory agreement over a wide range of ϕ. The O atom measurements inferred by the "slope method,"[11,12] however, are significantly higher than the simulated values for $\phi < 0.8$. Considering the acknowledged experimental difficulty and related uncertainty of these measurements at fuel-lean conditions,[12] the *C/D* simulations appear to adequately represent the combustion physics. This is once again evident for ϕ near 1.0, where both chemiluminescence measurement techniques infer similar values of O atom concentration. In this region, the PSR predictions once again show a well-defined but unsubstantiated maximum, while the *C/D* simulations closely follow the data.

The observed disparity between concentration data and predictions for $0.6 < \phi < 0.9$ underscores the importance of the physical coupling between chemical energy release rate and fluid mixing rates. The simulation results and the PSR predictions both represent fixed mixing rate approximations to the actual combustion fluid mechanics. It would be remarkable if either calculation could adequately match the measured values over the entire range of fuel-air equivalence ratios. Further consideration of the data and numerical results presented in Figs. 5 and 6 suggests that, for $0.6 < \phi < 0.9$, the characteristic mixing time scales are longer than estimated and that the *C/D* mixing frequency should be reduced to provide better agreement. Lowering the mixing rate would, in fact, increase the predicted OH concentrations at the expense of the available O atoms. In turn, this would force better agreement between simulation and O atom data taken by direct chemiluminescence (open triangles, Fig. 6). An appropriate model for the variation of mixing time scales with ϕ is presently under investigation.

Conclusions

Coalescence/dispersion simulations of previously reported CH_4/air combustion experiments in a high-intensity jet-stirred reactor have been used to predict temperature, OH and O concentrations for identical reactor conditions. A priori estimates of a reactor mixing frequency β have been compared with frequencies determined a posteriori by adjusting the *C/D* frequency to force agreement between measured and simulated values of temperature and species concentrations at $\phi = 1.0$. This comparison indicates that, to the same order of magnitude, an appropriate choice of β may be determined either by consideration of the energy dissipation time scales or by an appropriate description of the reactor length and velocity scales.

The *C/D* simulations follow both the magnitude and trends of the measured values of T, O, and OH for $0.7 < \phi < 1.4$ and thus appear to satisfactorily represent the physics of the high-intensity combustion process. For fuel-lean conditions, the PSR calculations and the *C/D* simulations each underpredict the measured OH concentrations, but support the O atom concentrations measured by the direct chemiluminescence method. Both techniques adequately represent the fuel-rich measurements. However, near stoichiometric conditions where mixing and reaction rates are comparable, the PSR predictions indicate distinct maximums in both O atom and OH concentrations that are not supported by experiment. The *C/D* simulations, on the other hand, closely follow the data in this region and thus indicate the ability of the *C/D* technique to represent the influence of finite-rate mixing in turbulent combustion.

Acknowledgment

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