

## JOINT PDF CALCULATIONS OF A NON-EQUILIBRIUM TURBULENT DIFFUSION FLAME

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We report calculations of a syngas turbulent diffusion flame. The calculations are based on the Monte Carlo solution of a modelled transport equation for the joint probability density function (pdf) of the velocity and two scalar variables  $\xi$  and  $\eta$ . In the thermochemical model,  $\xi$  is the mixture fraction and  $\eta$  is a reaction progress variable. The usual two-body reactions (including  $\text{CO} + \text{OH} \rightleftharpoons \text{CO}_2 + \text{H}$ ) are assumed to be in partial equilibrium, while the three-body recombination reactions proceed at a finite rate.

The calculations are compared to the available experimental data and also to previous calculations based on an assumed joint pdf of  $\xi$  and  $\eta$ . In general there is good agreement between the calculations and the measurements of major species concentrations and of mean and rms OH concentrations. However, in the rich parts of the flame there are discrepancies in the calculated CO and OH concentrations which are most likely due to the assumption that CO is in partial equilibrium. The present calculations show some improvement over those based on an assumed pdf. It is found that  $\xi$  and  $\eta$  are highly correlated, contrary to one postulate of the assumed pdf model.

### Introduction

In many turbulent diffusion flames the instantaneous temperature, density and major species concentrations are close to their equilibrium values. Then these thermochemical properties are uniquely related to the mixture fraction  $\xi(x,t)$  which is a conserved scalar<sup>1</sup>. Several calculations of turbulent diffusion flames have been made based on this equilibrium-chemistry assumption<sup>2-4</sup>. But there are several important phenomena that are inherently dependent on non-equilibrium effects. Examples are: NO formation; CO burnout; and extinction.

The syngas (40%CO, 30%H<sub>2</sub>, 30%N<sub>2</sub>) turbulent diffusion flame studied by Drake et al.<sup>5</sup> and Correa et al.<sup>6</sup> reveals non-equilibrium effects. In particular, the mean OH concentration is found to be, typically, five times the value it would have if equilibrium prevailed.

We report here calculations of the syngas turbulent diffusion flame, including non-equilibrium OH. The thermochemistry is described by the two-variable ( $\xi, \eta$ ) model used by Correa et al.<sup>6</sup> In this model  $\xi$  is the mixture fraction (a conserved scalar) and  $\eta$  is the reaction prog-

ress variable. The two-body reactions, including  $\text{CO} + \text{OH} \rightleftharpoons \text{CO}_2 + \text{H}$ , are assumed to be in partial equilibrium<sup>7,8</sup>, but full equilibrium ( $\eta=1$ ) is approached at a finite-rate determined by the rates of the three-body recombination reactions.

The calculations are based on the Monte Carlo solution of a modelled transport equation<sup>9</sup> for the joint pdf of the velocities,  $\xi$  and  $\eta$ . The use of the joint pdf equation avoids several modelling problems: reaction and the convective transport of momentum and the scalars are treated exactly; and no assumption is needed about the shape of the joint pdf of  $\xi$  and  $\eta$ , since the joint pdf is calculated. The effects of pressure fluctuations and viscous and diffusive processes have to be modelled. The same modelling is used here as proved successful for the self-similar plane jet<sup>10</sup>.

The joint pdf calculations reported here are compared to the available experimental data<sup>5,6</sup> and also to previous calculations. Correa et al.<sup>6</sup> reported calculations of the mean and rms OH concentration, while Correa<sup>11</sup> reported calculated CO concentration at one axial location. These previous calculations pertain to the same

flame, and the same thermochemical model was used. However, the flow and turbulence were treated by the  $k$ - $\epsilon$  turbulence model, and a shape was assumed for the joint pdf of  $\xi$  and  $\eta$ . The two variables  $\xi$  and  $\eta$  were assumed to be statistically independent and their marginal pdf's were assumed to be a beta-function distribution and a triple-delta-function distribution respectively. The present method is superior in that convective transport is treated exactly, and the joint pdf of  $\xi$  and  $\eta$  is calculated rather than a parametric expression for its shape being assumed.

The principal objectives of this work are twofold: to assess the error resulting from the assumption that CO is in partial equilibrium; and, with the improved modelling afforded by the joint pdf approach, to determine whether super-equilibrium OH can be calculated without the significant discrepancies evident in the assumed-pdf calculations<sup>5</sup>.

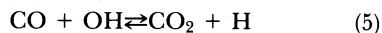
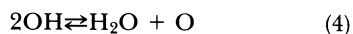
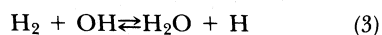
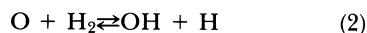
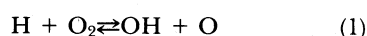
Previous combustion studies using the joint pdf equation include the premixed flame calculations of Pope & Anand<sup>12</sup>, Anand & Pope<sup>13</sup> and of Pope & Cheng<sup>14</sup>. Several calculations involving finite-rate reactions have been made using the scalar pdf equation<sup>15,16</sup>. Pope<sup>15</sup> used the scalar pdf equation with a similar thermochemical model to that used here to calculate the burnout of CO in a premixed propane/air flame.

### Syngas Flame

The turbulent diffusion flame studied<sup>5,6</sup> consists of a syngas fuel jet emerging from a nozzle of diameter  $d = 3.18$  mm at an average velocity of 54.6 m/s, and a co-flowing air stream with a velocity of 2.4 m/s. Simultaneous measurements of temperature, density and major species concentrations ( $H_2$ ,  $O_2$ ,  $N_2$ ,  $CO$ ,  $H_2O$ ) have been made<sup>5</sup> using pulsed Raman scattering. From these measurements (and the assumption that the hydrogen to carbon atom ratio is uniform) the concentration of  $CO_2$  and the mixture fraction  $\xi$  can be deduced. In addition, OH concentration has been measured<sup>5</sup> using laser saturated OH fluorescence.

### Thermochemical Model

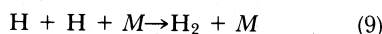
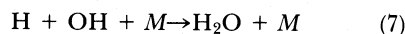
The thermochemical model is identical to that used by Correa et al.<sup>6</sup> Eight reactive species are considered— $O_2$ ,  $H_2$ ,  $CO$ ,  $CO_2$ ,  $H_2O$ ,  $O$ ,  $H$ ,  $OH$ —and  $N_2$  is taken to be inert. The five two-body reactions



are assumed to be in partial equilibrium. A composite mass fraction  $Y_{H_2}^*$  is defined<sup>17</sup> by

$$Y_{H_2}^* = Y_{H_2} + \frac{1}{2} \frac{M_{H_2}}{M_{OH}} Y_{OH} + \frac{M_{H_2}}{M_O} Y_O + \frac{3}{2} \frac{M_{H_2}}{M_H} Y_H + \frac{M_{H_2}}{M_{CO}} Y_{CO}, \quad (6)$$

where  $Y_i$  is the mass fraction and  $M_i$  is the molecular weight of species  $i$ . The rate of change of  $Y_{H_2}^*$  is determined by the rates of the three-body reactions



For a given mixture fraction  $\xi$ ,  $Y_{H_2}^*$  has a maximum value  $Y_{H_2}^{*u}$  corresponding to no reaction, and a value  $Y_{H_2}^{*e}$  corresponding to full chemical equilibrium. Accordingly the reaction progress variable  $\eta$  is defined by

$$\eta \equiv \frac{Y_{H_2}^* - Y_{H_2}^{*u}}{Y_{H_2}^{*e} - Y_{H_2}^{*u}} \quad (11)$$

All the thermochemical variables calculated from this scheme are tabulated as functions of  $\xi$  and  $\eta$  for use in the Monte Carlo solution procedure. The variables tabulated are: the species mass fractions; the density; the temperature; and,  $w(\xi, \eta)$ —the rate of change of  $\eta$  due to reaction.

A possible weakness of this thermochemical model is the assumption that the reaction  $CO + OH \rightleftharpoons CO_2 + H$  is in equilibrium. Fenimore and Moore<sup>8</sup> suggest that the assumption is reasonable at temperatures above 1800°K and Bilger and Stärner<sup>18</sup> reached a similar conclusion. In the syngas flame the peak instantaneous temperatures are in excess of 2,000°K, but the measured mean temperatures are typically less than 1700°K. As stated above, one objective of

this work is to assess the errors resulting from the assumed equilibrium of CO. This assumption can of course be avoided<sup>19,11</sup>; but then a more costly three-variable thermochemical model is needed.

It can also be argued that the three-body reaction  $H + O_2 + M \rightleftharpoons HO_2 + M$  should be included. If this were done, the reaction rate  $w(\xi, \eta)$  might increase for lean mixtures<sup>19</sup>. However, it was decided to use the same thermochemical model as Correa et al. so that differences in the calculations are due solely to the different turbulence and pdf modelling.

### Joint PDF Equation

In the turbulent diffusion flame, the dependent variables  $U$ ,  $\xi$  and  $\eta$  are functions of the axial, radial and circumferential coordinates ( $x$ ,  $r$  and  $\theta$ ) and of time  $t$ . But because the flow is statistically stationary and axisymmetric, all one-point statistics depend on  $x$  and  $r$  only.

The joint pdf  $f(V, \xi, \hat{\eta}; x, r)$  is the probability density of the joint events  $U(x, r, \theta, t) = V$ ,  $\xi(x, r, \theta, t) = \xi$  and  $\eta(x, r, \theta, t) = \eta$ , where  $V$ ,  $\xi$  and  $\eta$  are independent velocity and composition variables. Since all thermochemical quantities are known functions of  $\xi$  and  $\eta$ , all one-point thermochemical statistics can be determined from  $f$ . For example, the mean density is

$$\langle \rho(x, r) \rangle = \iiint \rho(\xi, \hat{\eta}) f(V, \xi, \hat{\eta}; x, r) dV d\xi d\hat{\eta} \quad (12)$$

where integration is over all velocities and compositions.

In the transport equation for  $f$  the effects of convective transport, reaction, and the mean pressure gradient appear in closed form, even in variable-density flow<sup>9</sup>. The effects of pressure fluctuations and molecular transport have to be modelled.

The modelling used here is essentially the same as that used previously by Pope<sup>10</sup> to calculate the self-similar plane jet: the minor differences are noted below. One aspect of the modelling is to discriminate between turbulent and non-turbulent fluid according to whether  $\xi$  is positive or zero. Non-turbulent fluid ( $\xi=0$ ) is entrained (i.e. becomes turbulent,  $\xi>0$ ) at a specified rate and it exchanges momentum with the turbulent fluid at another rate. Pope<sup>10</sup> also prescribed an exchange of kinetic energy from the turbulent fluid to the non-turbulent fluid. Following Haworth and Pope<sup>20</sup> we retain the energy loss from the turbulent fluid, but do not transfer it to the non-turbulent fluid.

Within the turbulent fluid, improved sto-

chastic mixing models<sup>21</sup> account for the effects of molecular mixing, and the stochastic reorientation model<sup>9</sup> accounts for the effect of the pressure fluctuations. Since molecular mixing conserves (globally) the mass of each species, the mixing models are applied to  $Y_{H_2}^*$  (which is a linear combination of mass fractions) rather than to  $\eta$  (which is non-linear, Eq. 11). For a reactive scalar such as  $Y_{H_2}^*$ , the use of stochastic mixing models is justifiable provided that the steepest scalar gradients are predominantly caused by turbulent straining rather than being associated with flamelets<sup>9,12</sup>. This provision is met when the reaction time scale is large compared to the Kolmogorov time scale, as is the case here.

The values of all the model constants used are those given by Pope<sup>10</sup>.

Except for entrainment, all the modelled processes occur at a rate proportional to

$$\omega \equiv \epsilon/k, \quad (13)$$

where  $k$  is the turbulent kinetic energy and  $\epsilon$  is its rate of dissipation. As previously<sup>10</sup>,  $\omega$  is assumed to be constant across the flow, but the method of specifying  $\omega(x)$  for the self-similar plane jet is inapplicable to the diffusion flame. Instead we specify

$$\omega(x) = \omega^*(\bar{U}_1(x, 0) - U_\infty)/(r_{0.9} - r_{0.1}), \quad (14)$$

where  $\bar{U}_1(x, r)$  is the mass-averaged axial velocity,  $U_\infty$  is the co-flow velocity,  $r_{0.1}$  is defined such that

$$\bar{U}_1(x, r_{0.1}) - U_\infty = 0.1(\bar{U}_1(x, 0) - U_\infty); \quad (15)$$

and similarly  $r_{0.9}$ . The dimensionless frequency  $\omega^*$  is chosen to be 0.16 in order to obtain the experimentally-observed spreading rate  $r_{0.5}(x)$ .

### Solution Procedure

The modelled joint pdf equation is solved by the Monte Carlo method described by Pope<sup>9</sup>. Since in the flow considered there is negligible probability of negative axial velocities, a significant simplification—analogueous to the boundary-layer approximation—can be made. This enables the solution to be obtained by marching in the  $x$ -direction.

In the Monte Carlo method, at each step in the  $x$ -direction, the joint pdf is represented by a large number  $N$  of notional particles ( $N \approx 35,000$ ). At location  $x$ , the  $n$ -th particle has the radial position  $r^{(n)}(x)$ , velocity  $U^{(n)}(x)$  and compo-

sition  $\xi^{(n)}(x)$ ,  $\eta^{(n)}(x)$ . From these particle properties, the discrete pdf is defined by

$$f^*(V, \hat{\xi}, \hat{\eta}; x, r) \equiv m \sum_{n=1}^N \delta(V - \underline{V}^{(n)}[x]) \delta(\hat{\xi} - \xi^{(n)}[x]) \delta(\hat{\eta} - \eta^{(n)}[x]) \delta(r - r^{(n)}[x]) / \rho(\hat{\xi}, \hat{\eta}), \quad (16)$$

where  $m$  is the mass attributed to each particle. The Monte Carlo method is constructed so that the expectation of  $f^*$  is equal to  $f^g$ . It may be noted that this is a grid-free representation: the total mass of fluid is discretized into  $N$  representative particles, but the 6-dimensional  $V$ - $\hat{\xi}$ - $\hat{\eta}$ - $r$  space is not discretized.

Mean quantities—either mass-averaged or volume-averaged—are extracted from the particle properties by cross-validated cubic smoothing splines<sup>9</sup>. The mean quantities so obtained contain a statistical error of order  $N^{-1/2}$ . With  $N \approx 35,000$  this error is estimated to be a few per cent. More precise error estimates are provided by Haworth & Pope<sup>22</sup>.

Initial conditions are specified at the jet exit ( $x=0$ ). The measured initial conditions are not matched in detail, but care is taken to match the axial momentum and mixture fraction flow rates. The solution is obtained for  $0 \leq x/d \leq 100$  in a succession of 640 steps in  $x$ . In the general step from  $x$  to  $x + \Delta x$  the particle properties ( $r^{(n)}$ ,  $\underline{V}^{(n)}$ ,  $\xi^{(n)}$  and  $\eta^{(n)}$ ) evolve as described by Pope<sup>9</sup>. As a consequence of reaction,  $\eta^{(n)}$  evolves for a time  $\Delta x/U_1^{(n)}(x)$  according to the ordinary differential equation

$$\frac{d\eta^{(n)}}{dt} = w(\xi^{(n)}, \eta^{(n)}), \quad (17)$$

with the initial condition  $\eta^{(n)}(x)$ . A direct algorithm would require Eq. (17) to be integrated for each particle on each step, i.e. about 20 million times. Instead a computationally-efficient table-look-up algorithm is used.

With approximately 35,000 particles and 640 steps, the computation required 65 minutes CPU time on an IBM 3084QX. Only 6% of this time is used to perform reaction, compared to 33% to perform transport and 17% to extract means.

## Results and Discussion

Figure 1 shows profiles of the mass-averaged mixture fraction  $\bar{\xi}$  and the mean density  $\langle \rho \rangle$  and temperature  $\langle T \rangle$  at  $x/d = 25$ . The radial coordinate  $r$  is normalized by the nozzle radius  $a = \frac{1}{2}d$ .

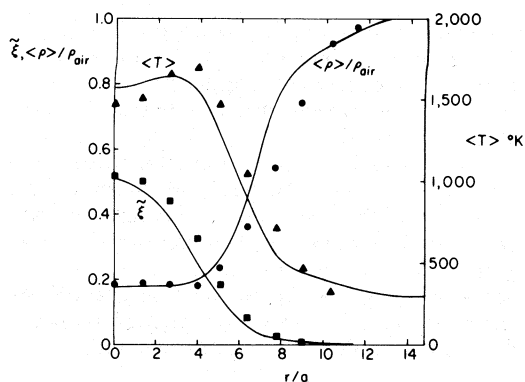


FIG. 1. Mass-averaged mixture fraction  $\bar{\xi}$ , mean density  $\langle \rho \rangle / \rho_{air}$  and mean temperature  $\langle T \rangle$  against  $r/a$  at  $x/d = 25$ . Joint pdf calculations, lines; experimental data<sup>5</sup>, symbols.

The joint pdf calculations (solid lines) are compared with the experimental data of Drake et al.<sup>5</sup>. The calculated profile of  $\bar{\xi}$  is in fairly good agreement with the data, but is slightly too narrow. The density and temperature profiles also show good agreement but, consistent with the  $\bar{\xi}$  profile, they too are slightly too narrow. The calculated mean temperature on the centerline is 1560°K, which is 70°K greater than that measured.

At the other two measurement stations  $x/d = 10$  and  $x/d = 50$ , these profiles (not shown) exhibit a comparable level of agreement. The only significant difference being that at  $x/d = 10$  the calculated centerline temperature is 200°K more than the measured value of 550°K.

Figure 2 shows profiles of the mean mole fractions of  $H_2$ ,  $O_2$ ,  $CO$  and  $CO_2$  at  $x/d = 25$ . The profiles of  $H_2$  and  $O_2$  are in excellent agreement with the data. But there are significant discrepancies in the  $CO$  and  $CO_2$  profiles. In the central part of the flame ( $r/a < 4$ ), where the mean mixture fraction  $\bar{\xi}$  is on the rich side of stoichiometric ( $\xi_{stoich} \approx 0.3$ ), the calculated  $CO$  mole fraction is 30% less than the measurement; and the calculated  $CO_2$  mole fraction is correspondingly greater than that measured. At  $x/d = 10$  (not shown) the calculated mole fraction of  $CO$  is 10% too low on the axis, while at  $x/d = 50$  (not shown) it agrees with the data (to within experimental error).

It is reasonable to suppose that the cause of these discrepancies lies in the assumption that the reaction  $CO + OH \rightleftharpoons CO_2 + H$  is in partial equilibrium. The inapplicability of this assumption explains not only the calculated excess of  $CO_2$  but also the too high calculated temperatures in the center of the flame at  $x/d = 10$  and, to a lesser extent, at  $x/d = 25$ . By  $x/d = 50$

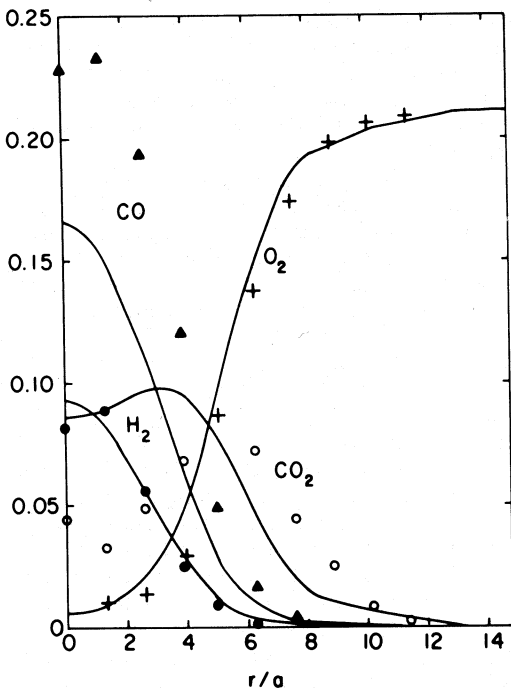


FIG. 2. Mean mole fraction of  $H_2$ ,  $O_2$ ,  $CO$ , and  $CO_2$  against  $r/a$  at  $x/d = 25$ . Joint pdf calculations, lines; experimental data<sup>5</sup>, symbols.

sufficient time may have elapsed for  $CO$  to be in equilibrium, hence explaining the observed agreement there.

Correa<sup>11</sup> made calculations of this flame with the same two-variable thermochemical model but with an assumed shape for the joint pdf of  $\xi$  and  $\eta$ . The  $CO$  mole fractions obtained at  $x/d = 25$  are, typically, 15% less than those reported here, in slightly worse agreement with the data. Correa<sup>11</sup> also employed a three-variable thermochemical model that allows for the disequilibrium of  $CO$ . With this model the calculated mole fractions of  $CO$  at  $x/d = 25$  are, typically, 20% greater than those reported here, in significantly better agreement with the data. This lends support to the argument that the cause of the discrepancies in the present calculations lies in the assumed partial equilibrium of the reaction  $CO + OH \rightleftharpoons CO_2 + H$ .

It may be observed that at  $r/a = 3$  the calculated mean mole fraction of  $CO$  is significantly less than the measured value, and yet the calculated mean temperature agrees with that measured. There are two possible explanations of this superficial inconsistency. First, since the  $CO$  mole fraction and the temperature are non-linear functions of the fluctuating quantities,  $\xi$  and  $\eta$ , there is no direct correspondence

between the mean  $CO$  concentration and the mean temperature: thus there may be no inconsistency. Second, it is found that the temperature deduced from measured major species concentrations is in error compared to the directly measured temperature (Drake, private communication). A possible source of this discrepancy may be a systematic error in the measured  $CO$  concentrations.

We now look at profiles of mean  $OH$  concentration. Figures 3 and 4 show (at  $x/d = 25$  and 50 respectively) the mean  $OH$  concentration: from the joint pdf calculation (solid line); from

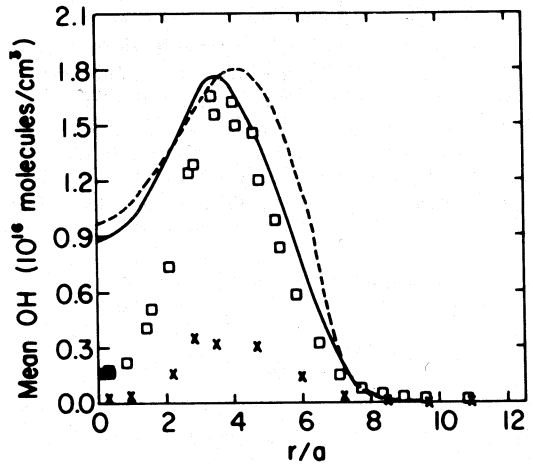


FIG. 3. Mean  $OH$  concentrations against  $r/a$  at  $x/d = 25$ . Joint pdf calculations, solid line; assumed pdf calculations<sup>6</sup>, dashed line; experimental data<sup>6</sup>, squares; equilibrium value deduced from major species measurements<sup>6</sup>, crosses.

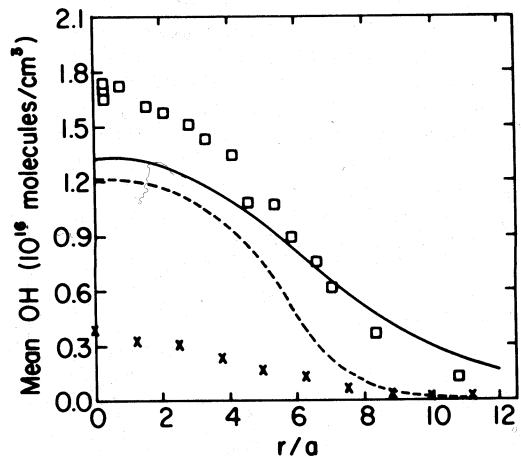


FIG. 4. Mean  $OH$  concentration against  $r/a$  at  $x/d = 50$ . Symbols same as Fig. 3.

the two-variable assumed-pdf calculations of Correa et al.<sup>6</sup> (dashed line); from the measurements<sup>6</sup> (squares); and, that deduced<sup>6</sup> from the major species measurements and the assumption of full equilibrium for OH. It is immediately evident that OH, and hence the radical pool, is not in equilibrium: even at  $x/d = 50$  the mean OH concentration is four times the equilibrium value.

At  $x/d = 25$  (Fig. 3) in the outer, lean, part of the flame ( $r/a > 4$ ,  $\xi < \xi_{\text{stoich}}$ ) joint pdf calculations are in good agreement with the data, and show an improvement over the assumed-pdf calculations. In the inner part of the flame, the overprediction of OH by both models is most likely associated with the overprediction of CO<sub>2</sub> and T. At  $x/d = 50$  the joint pdf calculations agree with the data to within the estimated experimental error<sup>6</sup> of  $\pm 30\%$ .

Figures 5 and 6 show rms OH concentration profiles at  $x/d = 25$  and 50. The joint pdf calculations yield higher rms values than the

assumed pdf calculations—by a factor of two at  $x/d = 50$ —and are generally in good agreement with the data. The only significant discrepancy is in the inner part of the flame at  $x/d = 25$ . Again this is explained by the overprediction of CO<sub>2</sub> and T.

The results of the joint pdf calculations can be used to test the validity of assumed pdf's. A major assumption made by Correa et al.<sup>6</sup> is that  $\xi$  and  $\eta$  are statistically independent. If this were so, the covariance  $\widetilde{\xi''\eta''}$  and the correlation coefficient  $R$  between  $\xi$  and  $\eta$  would be zero:

$$R \equiv \widetilde{\xi''\eta''} [\widetilde{\xi''\xi''} \widetilde{\eta''\eta''}]^{-1/2}. \quad (18)$$

Figures 7 and 8 show the variances  $\widetilde{\xi''\xi''}$  and  $\widetilde{\eta''\eta''}$ , and covariance  $\widetilde{\xi''\eta''}$  at  $x/d = 25$  and 50. These profiles show that  $\xi$  and  $\eta$  are highly correlated. At  $x/d = 25$  the correlation coefficient on the centerline is 0.82 and at  $r/a = 5$  it is  $-0.44$ . At  $x/d = 50$ , at  $r/a = 0$  and 8, the correlation coefficient is  $-0.57$  and  $-0.75$  respectively.

Thus the joint pdf calculations show that  $\xi$  and  $\eta$  are far from being independent and hence the assumed pdf of Correa et al.<sup>6</sup> is not a good approximation. One effect of this poor approximation is the inferior calculations of the mean and rms of OH compared to the joint pdf calculations.

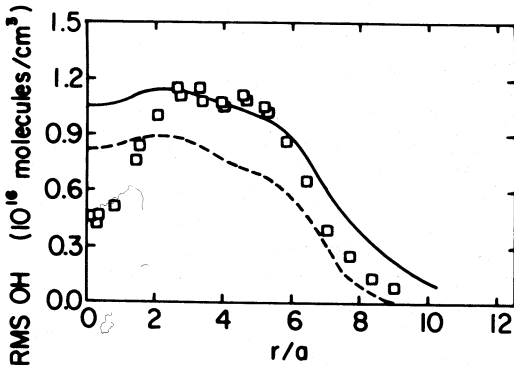


FIG. 5. Rms of OH concentration against  $r/a$  at  $x/d = 25$ . Symbols same as Fig. 3.

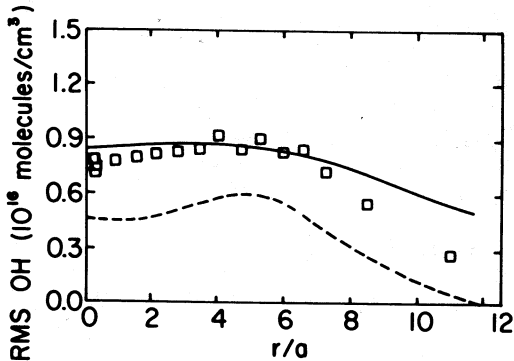


FIG. 6. Rms of OH concentration against  $r/a$  at  $x/d = 50$ . Symbols same as Fig. 3.

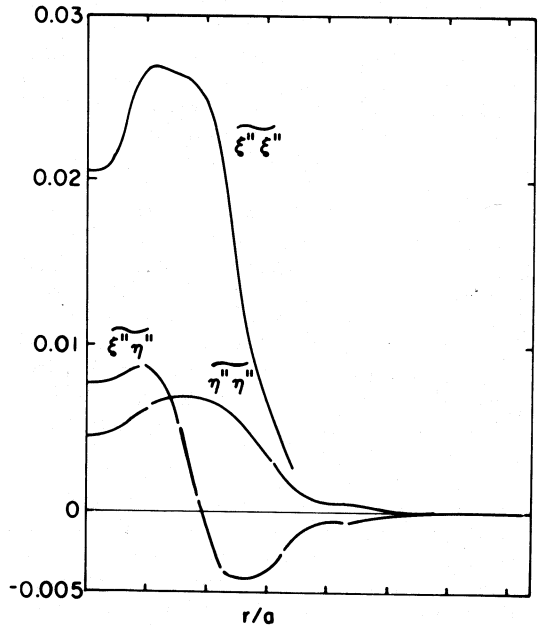


FIG. 7. Mass-averaged variances and covariance of  $\xi$  and  $\eta$  against  $r/a$  at  $x/d = 25$ . Joint pdf calculations.

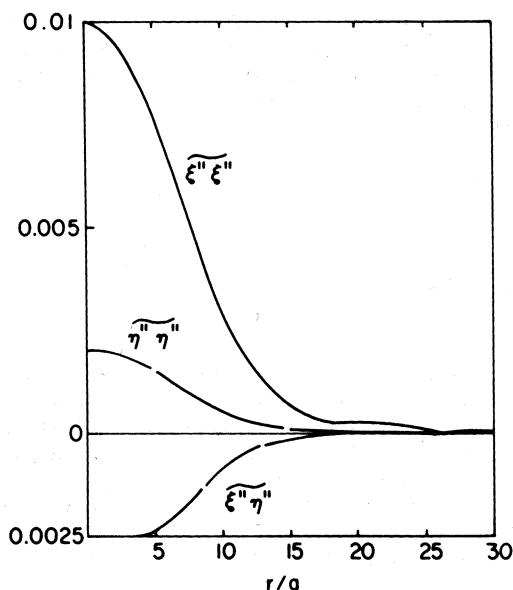


FIG. 8. Mass-averaged variance and covariance of  $\xi$  and  $\eta$  against  $r/a$  at  $x/d = 50$ . Joint pdf calculations.

### Conclusion

A modelled transport equation for the joint pdf of velocities and compositions has been solved by a Monte Carlo method to calculate the properties of a syngas turbulent diffusion flame. The calculated mean mixture fraction, temperature, density, and major species concentrations are, in general, in good agreement with the available experimental data. However, at  $x/d = 25$ , in the fuel rich part of the flame, CO is underpredicted, presumably as a consequence of its assumed partial equilibrium. Except at this location, the calculated mean and rms OH concentrations are in good agreement with the data and show an improvement over assumed-pdf calculations<sup>6</sup>.

The calculations of CO and CO<sub>2</sub> would be improved by use of a three-variable thermochemical model in which the reaction  $\text{CO} + \text{OH} \rightleftharpoons \text{CO}_2 + \text{H}$  is not assumed to be in the partial equilibrium. Such schemes have been developed and used by Morr & Heywood<sup>19</sup> and by Correa<sup>11</sup>. In the Monte Carlo method, with the two-variable scheme, only 6% of the CPU time is used to perform reaction. Consequently there is no doubt that a three-variable scheme is computationally feasible.

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## COMMENTS

*M.A. Delichatsios, Factory Material Research, USA.* A major and crucial uncertainty in your model is the lack of a convincing thermo-kinetic chemical model for the fuel that you have used in your model and experiments. Could you attribute, for example, the discrepancies of your predictions with experiments to the turbulent modeling assumptions or to incomplete chemical kinetic modeling?

*Author's Reply.* As stated in the paper, we used the

same thermochemical model as Correa et al.<sup>6</sup> (in spite of its known deficiencies) so that we could examine the effect of the different turbulence modelling. We have no reason to doubt that the observed discrepancies between calculations and measurements are due to the inadequate thermochemical model. We intend to perform calculations with a three-variable thermochemical model, which will decide the question.