

## Mixing Rules for State Relationships of Methane and Acetylene/Air Diffusion Flames

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Measurements of concentrations of major gas species in laminar diffusion flames burning mixtures of CH<sub>4</sub> and C<sub>2</sub>H<sub>2</sub> in air are reported. The results are interpreted in terms of the laminar flamelet concept. Two methods of estimating the state relationships of fuel mixtures were evaluated. Overall, a mixing rule that utilizes the information concerning the state relationships of the individual fuels provides better estimates of species concentrations in flames burning fuel mixtures than the generalized state relationships.

### INTRODUCTION

Many practical applications such as oil well blowout fires and fuel additives for flame emissivity improvements involve combustion of hydrocarbon mixtures. However, past studies of flame structure have generally been restricted to single component fuels. The objective of the present work was to study the relationship between the structure properties of flames burning fuel mixtures and single component fuels.

Bilger [1] observed that concentrations of major gaseous species in laminar diffusion flames burning C<sub>7</sub>H<sub>16</sub> [2], CH<sub>4</sub> [3], and C<sub>3</sub>H<sub>8</sub> [4] in air could be correlated as a function of the local mixture fraction (the fraction of elemental mass that originated in the fuel stream) over a relatively wide range of stretch rates. Subsequently, such correlations were observed for laminar diffusion flames burning C<sub>2</sub>H<sub>4</sub> and C<sub>2</sub>H<sub>2</sub> in air [5]. These correlations have been called state relationships [6] and are properties of a particular fuel burning in atmospheric air. It must be noted that the correlations are not applicable to the concentrations of minor pollutant species and for conditions near flame extinction and attachment. The main application of the state relationship has been in the calculation of radiation heat transfer [5].

The state relationships for fuels with relatively

simple chemistry such as H<sub>2</sub> and CO can be obtained from chemical equilibrium calculations [7, 8]. State relationships for CH<sub>4</sub>/air [9] and CO-H<sub>2</sub>-N<sub>2</sub> air [10] flames have also been obtained from detailed chemical kinetics. However, for fuels with more complicated chemistry (particularly involving soot), measurements of species concentrations in laminar diffusion flames have been the source of state relationships [1-6].

For flames burning mixtures of fuels with relatively complex chemistry (including soot), measurements in laminar flames burning many possible blends are generally required. These measurements are tedious and probably impossible to make for all possible fuel blends. In fact, measurements for many single-component hydrocarbon fuels have not been completed.

Two distinct approaches are currently available for the treatment of flames burning hydrocarbon mixtures. The first involves the generalized state relationship concept of Sivathanu and Faeth [11]. Within this approach, the concentrations of major gaseous species (N<sub>2</sub>, O<sub>2</sub>, fuel, CO<sub>2</sub>, CO, H<sub>2</sub>O, and H<sub>2</sub>) can be estimated for any hydrocarbon fuel C<sub>n</sub>H<sub>m</sub> based on universal state relationship functions developed from experimental data for CH<sub>4</sub>, C<sub>3</sub>H<sub>8</sub>, C<sub>7</sub>H<sub>16</sub>, C<sub>2</sub>H<sub>4</sub>, and C<sub>2</sub>H<sub>2</sub>/air flames. For fuel mixtures, *n* and *m* take fractional values based on the chemical formulae and molar concentrations of the individual components.

Sivathanu and Faeth [11] found that the generalized functions for N<sub>2</sub>, O<sub>2</sub>, and fuel collapsed very well for the different hydrocarbons. The

\*Experiments conducted at the Center for Fire Research, National Institute of Standards and Technology, Gaithersburg, MD 20899 under the management of Dr. David D. Evans.

generalized functions for  $\text{CO}_2$  and  $\text{H}_2\text{O}$  showed somewhat increased scatter based on the type of fuel. The generalized plots for  $\text{CO}$  and  $\text{H}_2$  showed that flames burning heavily sooty fuels such as ( $\text{C}_2\text{H}_2$  and  $\text{C}_2\text{H}_4$ ) contain much larger quantities of  $\text{CO}$  and  $\text{H}_2$  than those burning paraffins— $\text{CH}_4$ ,  $\text{C}_3\text{H}_8$ , and  $\text{C}_7\text{H}_{16}$ . This difficulty would probably exist in the application of the generalized state relationships to fuel mixtures containing one or more heavily sooty components.

The alternate approach involves combining the different state relationships for individual fuels to obtain the state relationships for the fuel mixture [12, 13] using a mixing rule. This approach has been used for flames burning a  $\text{CH}_4/\text{C}_7\text{H}_{16}$  mixture [12] and those burning  $\text{CH}_4/\text{C}_2\text{H}_2$  mixtures [13]. In this approach, universal behavior for the individual fuel components is not assumed. However, interactions between the chemistry of individual fuels are neglected.

Neither the generalized state relationships of Ref. 11 nor the mixing rule of Refs. 12 and 13 have been evaluated by comparison with experimental data for flames burning fuel mixtures except for the 99%  $\text{CH}_4$  + 1%  $\text{C}_7\text{H}_8$  flame of Ref. 14. However, the concentrations of major gas species within this flame were identical to those in a single component  $\text{CH}_4$  flame within experimental uncertainty. The objective of the present work was to remove this deficiency by considering two representative mixtures of a relatively clean burning ( $\text{CH}_4$ ) and a highly sooty ( $\text{C}_2\text{H}_2$ ) fuel. Flames burning  $\text{CH}_4$  and  $\text{C}_2\text{H}_2$  individually were also studied as baseline.

The article begins with a description of the experimental methods. The generalized state relationships of Ref. 11 are then discussed in light of the present  $\text{CH}_4$  and  $\text{C}_2\text{H}_2$  single fuel data. The mixing rule of Refs. 12 and 13 is then described. The article concludes with a comparison of the experimental data for the mixture flames with the generalized state relationships and the mixing rule.

## EXPERIMENTAL METHODS

Following description of the experimental methods is brief. Details of similar experiments have been discussed elsewhere [5, 15-17].

## Apparatus

The state relationship data available in the literature have been inferred from four different flame configurations (see Ref. 11 for details). A co-flow laminar jet diffusion flame burner (similar to Refs. 5, 15 and 16) was used in the present work. The individual fuels ( $\text{C}_2\text{H}_2$  and  $\text{CH}_4$ ) were metered using calibrated rotameters and mixed in an opposed jet chamber with baffles and a long fuel pipe. All the flames were attached uniformly at the rim of the fuel tube. The burner was mounted on a vertical traverse with a measurement accuracy of 0.5 mm to probe the flames at different heights above the exit.

## Operating Conditions

Four flames were considered. Two of the flames burn the single component  $\text{C}_2\text{H}_2$  and  $\text{CH}_4$  and the remaining burn mixtures with 27% and 57%  $\text{CH}_4$  by mass in  $\text{C}_2\text{H}_2$ . The flames burning single component fuels were studied for comparison with existing measurements. The three flames containing  $\text{C}_2\text{H}_2$  in the fuel stream emitted smoke while the  $\text{CH}_4$  flame showed no evidence of soot at the flame tip.

## Gas Sampling and Analysis

The soot particles produced in the flames tend to clog the quartz microprobes used for sampling gases for chromatographic analysis [5]. In a continuous sampling mode, this problem prevented Gore [5] from obtaining measurements in the heavily sooty regions of the  $\text{C}_2\text{H}_2$ /air flames. In the present study, an evacuated bulb technique was used to obtain measurements in sooty regions of the flames. The technique is based on the simple principle that the flow that clogs the probe also carries the gas sample.

A quartz microprobe with an orifice of 250-300  $\mu\text{m}$  diameter is connected to an evacuated sampling bulb (approximately 0.3 atm of carrier gas—99.995% helium) with an isolation valve. A purge line carrying a small flow of helium is also connected to the probe via a purge flow valve. The sampling bulb is equipped with an evacuation port, a carrier gas pressurizing valve, an absolute pressure gauge, and a sampling septum.

During operation, the probe is moved quickly to a desired location in the flame while carrying a small (just to maintain overpressure) purge flow of helium. The sampling bulb is isolated during this process. Once the probe is at the appropriate location, the purge flow valve is closed and the bulb isolation valve is opened. The pressure inside the bulb is monitored. If the probe does not clog, then the sampling ends when the pressure in the bulb reaches 1 atm. At this time the bulb isolation valve is closed and gas samples are removed with a syringe for routine chromatography. If the probe clogs, the bulb reaches a constant subatmospheric pressure. At this time, the isolation valve is closed and the pressure in the bulb noted. A flow of helium is started by opening the sample pressurization valve until the pressure in the bulb reaches 1 atm. The fraction of flame sample in the bulb is known from the various pressure readings. At this time gas samples are removed for routine chromatography. The measurements are corrected for the purge and pressurizing helium flows. Data were collected from three different axial positions and several radial positions within all flames.

The sample injected into the chromatograph first goes through a desiccant column to remove water vapor. Condensation of water was not observed in the sampling bulb or in the syringe. The sample leaving the desiccant section is analyzed on a dry basis. The measured concentrations are converted to a wet basis by assuming the C-H ratio of the products is identical to that of the fuel. The mass originating in the fuel stream is calculated by multiplying the number of carbon atoms in the sample by the molecular weight of the fuel and then dividing the result by the number of carbon atoms per fuel molecule. The mass originating in the air stream is calculated by multiplying the number of  $N_2$  moles in the sample by the number of moles of air per mole of  $N_2$  (1.266) and by the molecular weight of air (28). The mixture fraction is calculated using the above two estimates of the mass in the sample from each stream.

The mass of soot particles is not considered in the above procedure of calculating the mixture fraction. It has been found that the concentrations of major gas species correlate well with the gas-phase mixture fraction [5] calculated in this man-

ner. However, the effects of heterogeneous processes and differential diffusion on sooty flames have not been fully understood.

Finite probe and sampling size result in gradient broadening errors in concentration measurements. When the resulting data are interpreted in the mixture fraction space, the gradient broadening effects in the physical space are of no consequence in regions where concentrations vary linearly with mixture fraction. Near the flame sheet, the sign of the slope of the concentration profiles of product species in the mixture fraction space changes, leading to possible gradient broadening effects.

Based on the approximate rule given by Fristrom and Westenberg [18] for a cylindrical probe volume and estimates of scalar dissipation rate and mass diffusivity, the resolution of the present measurements in the mixture fraction space is estimated to be  $\pm 0.01$  in the vicinity of the flame sheet (in the range of mixture fractions 0.05–0.1 depending on the fuel). Within this range the concentrations of major species in the flame sheet region ( $CO_2$ , CO,  $N_2$ , and  $H_2O$ ) change by less than  $\pm 5\%$  of their flame sheet values. This variation is well within the uncertainty of the present data. The concentrations of fuel and oxygen are very small in this region and are not important for flame radiation calculations.

A second well known consideration in the interpretation of data obtained from sampling probes is quenching of the chemical reactions at the point of sampling. Colket et al. [19] have discussed the aerodynamics of sampling probes and conclude that the quenching mechanism for quartz microprobes does not result from a critical flow at the orifice. Schoenung and Hanson [20] found up to 100% variation in measured CO concentrations with back pressure using a choked probe. Increasing backpressure resulted in a factor of 2 increase in CO concentrations in their experiments.

The effects of backpressure in the sampling bulb on the measured concentrations were evaluated by varying the backpressure at the start of sampling from 0.3 to 0.1 atm and the backpressure at the end of sampling from 1 to 0.5 atm. The changes observed in the concentrations of CO and  $CO_2$  were less than 10% and those for  $N_2$ ,  $O_2$ , and the fuel were less than 5%. Furthermore, the changes were not systematic. The pres-

ent results can also be evaluated by comparison of the data with those obtained by Smyth et al. [21] using 100–150- $\mu\text{m}$  probes under much higher-pressure ratios. The maximum differences between concentrations of  $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{CH}_4$ ,  $\text{N}_2$ , and  $\text{O}_2$  measured in methane flames in the present study and those reported in Ref. 21 were less than 10% (concentrations of  $\text{CO}$  were estimated from equilibration of the water gas shift reaction in Ref. 21).

The experimental uncertainties in the species concentration measurements are estimated to be less than 15% for concentrations above 5% of the measured peak values governed primarily by limited data. The lower bound (5% of the peak measurements) for reliable (within 15%) concentration measurements is set by the dynamic range of the calibrations and the integrator.

## RESULTS AND DISCUSSION

### Single Component Fuels

As discussed above, the present measurements of state relationships for the  $\text{CH}_4$  flames are in excellent agreement with the data of Ref. 21. These data are also in good agreement with measurements reported in Refs. 3–5.

Estimates of species ( $\text{N}_2$ ,  $\text{O}_2$ ,  $\text{CO}_2$ ,  $\text{CH}_4$ , and  $\text{CO}$ ) concentrations based on the generalized state relationships of Ref. 11 showed very good agreement with the present  $\text{CH}_4$  data. These results are shown in Ref. 17 and are not repeated here in view of the existing data for  $\text{CH}_4$  in the literature.

The measurements of  $\text{N}_2$ ,  $\text{O}_2$ , and  $\text{C}_2\text{H}_2$  plotted as a function of local mixture fraction for the present  $\text{C}_2\text{H}_2$  flame are shown in Fig. 1. The estimates of these quantities based on the generalized state relationships of Ref. 11 shown in Fig. 1 agree very well with the present data. The present data are also in good agreement with the measurements reported in Ref. 5.

The comparison between the generalized state relationship estimates of  $\text{CO}$  and  $\text{CO}_2$  concentrations and the present measurements is shown in Fig. 2. The estimates are in good agreement with the data on the fuel lean side. However, similar to the findings of Ref. 11, the concentrations are underestimated by more than 50% and the  $\text{CO}_2$

concentrations are overestimated by an equivalent amount on the fuel-rich side. The present data are in general agreement with those of Ref. 5. However, the present measurements of  $\text{CO}_2$  concentrations are lower than those of Ref. 5 in the heavily sooty regions. The difficulties with clogging and buildup of soot around the probe encountered in Ref. 5 are probably the cause of this discrepancy.

As discussed in Ref. 11, the difficulties associated with estimation of  $\text{CO}$  (and the corresponding difficulty with the estimation of  $\text{CO}_2$  found above) are associated with the complications of soot chemistry. When a mixture of heavily sooty fuel and a relatively clean fuel is considered, these difficulties may persist. Motivated by this, a mixture rule that utilizes the information from the state relationships of the individual fuels was proposed in Refs. 12 and 13. This rule is described briefly in the following.

### Mixture Rule for State Relationships

In constructing the mixture rule for state relationships, it is assumed that the chemistry of the individual fuels in the mixture cannot be generalized. However, the interactions between the reactions involving the two fuels are neglected under the present hypothesis. Since, differential diffusion effects are neglected under the conserved scalar hypothesis, the ratio of the reacted and unreacted mass of fuel 1 to the reacted and unreacted mass of fuel 2 remains unchanged. Thus the state relationships for identical mixture fractions are selected for constructing the mixing rule.

At a given mixture fraction, the state relationships for the individual fuels involve mole fractions of the major gaseous species ( $\text{CO}_2$ ,  $\text{CO}$ ,  $\text{H}_2$ ,  $\text{H}_2\text{O}$ ,  $\text{N}_2$ ,  $\text{O}_2$ ,  $\text{CH}_4$ , and  $\text{C}_2\text{H}_2$  in the present). The state relationships for a fuel mixture containing " $R$ " kg of  $\text{CH}_4$  mixed with 1 kg of  $\text{C}_2\text{H}_2$  in the fuel stream ( $R = 0.37$  and  $1.33$  for the present mixtures) are obtained as follows:

$$X_{i,m} = \frac{(26aR/16c)X_{i,\text{CH}_4} + X_{i,\text{C}_2\text{H}_2}}{1 + 26aR/16c} \quad (1)$$

In Eq. 1,  $X_{i,m}$  represents the mole fraction of species  $i$  ( $i = \text{CO}_2, \text{CO}, \text{N}_2, \text{O}_2, \text{H}_2\text{O}, \text{H}_2,$

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CH<sub>4</sub>, C<sub>2</sub>H<sub>2</sub>) in the state relationship for the mixture flame and  $X_{i, CH_4}$  and  $X_{i, C_2H_2}$  represent the mole fractions of species  $i$  in the state relationships for CH<sub>4</sub> and C<sub>2</sub>H<sub>2</sub>, respectively. The factor weighing the mole fractions from individual fuel state relationships is the ratio of number of moles of products generated by that fuel and the total number of product moles. This factor is calculated from the state relationships for individual fuels. In particular, "a" is the number of moles of C<sub>2</sub>H<sub>2</sub> burned to generate 1 mol of its products and "c" is the number of

moles of CH<sub>4</sub> required to generate 1 mol of its products. The mole fractions of the individual fuel components in the incoming mixture were used as the weighing factors for the reciprocal flammability limit by Hustad and Sonju [22]. This is different than the present mixing rule where the normalized sums of mole fractions of products generated by the individual fuel component are used as the weighing factors.  
It is noted that if the state relationships of individual fuels in the mixture are in close agreement with the generalized state relationships then

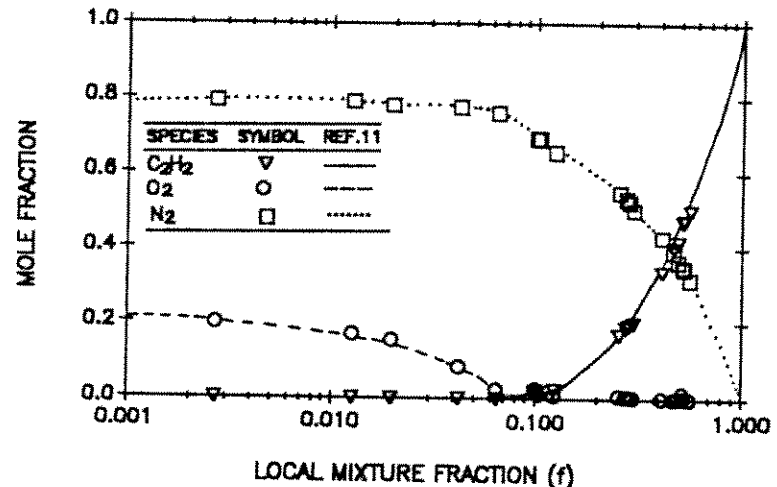


Fig. 1. Concentration measurements of N<sub>2</sub>, O<sub>2</sub>, and C<sub>2</sub>H<sub>2</sub> for C<sub>2</sub>H<sub>2</sub> air flames and comparison with generalized state relationships of Ref. 11.

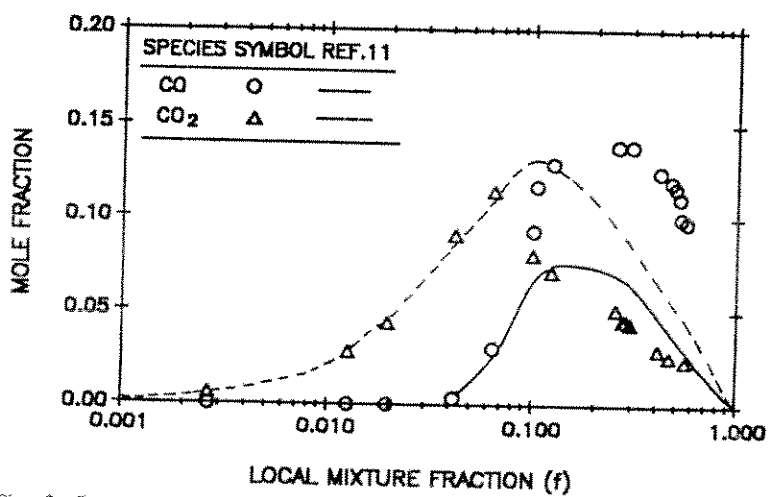


Fig. 2. Concentration measurements of CO<sub>2</sub> and CO for C<sub>2</sub>H<sub>2</sub>/air flames and comparison with generalized state relationships of Ref. 11.

the estimates of the state relationships for mixtures based on the mixing rule are identical to those based on the generalized state relationships.

### State Relationships for Fuel Mixtures

The concentrations of major gas species for a flame burning 27%  $\text{CH}_4$  by mass in  $\text{C}_2\text{H}_2$  ( $R = 0.37$ ) are plotted as a function of local mixture fraction in Fig. 3. For comparison, the estimates based on the generalized state relationships of Ref. 11 and the mixing rule discussed above are also plotted. For  $\text{N}_2$ ,  $\text{O}_2$ , and fuel, the estimates based on the generalized state relationships and the mixing rule are identical and are in reasonable agreement with the measurements. The estimates of concentrations of  $\text{CO}_2$  and  $\text{CO}$  based on the two methods are similar in the fuel-lean region and are also in good agreement with the measure-

ments. The generalized state relationships underestimate the  $\text{CO}$  and overestimate the  $\text{CO}_2$  for fuel-rich regions by up to 50%. This is similar to the difficulties encountered by this method for all sooty fuels [11]. The mixing rule accounts for the differences in the chemistry of the sooty fuels by including information from the individual relationships. Therefore, the estimates based on the mixing rule are in close agreement with the data.

Figure 4 shows measurements of species concentrations and estimates based on the generalized state relationships as well as the mixing rule for a flame burning 57%  $\text{CH}_4$  by mass in  $\text{C}_2\text{H}_2$  ( $R = 1.33$ ). Once again, reasonable estimates of  $\text{O}_2$ ,  $\text{N}_2$ , and fuel are obtained by both methods. Both estimates of  $\text{CO}_2$  and  $\text{CO}$  on the fuel-lean side are also reasonable. Similar to the other sooty fuels, the generalized state relationships underestimate the  $\text{CO}$  and overestimate the  $\text{CO}_2$

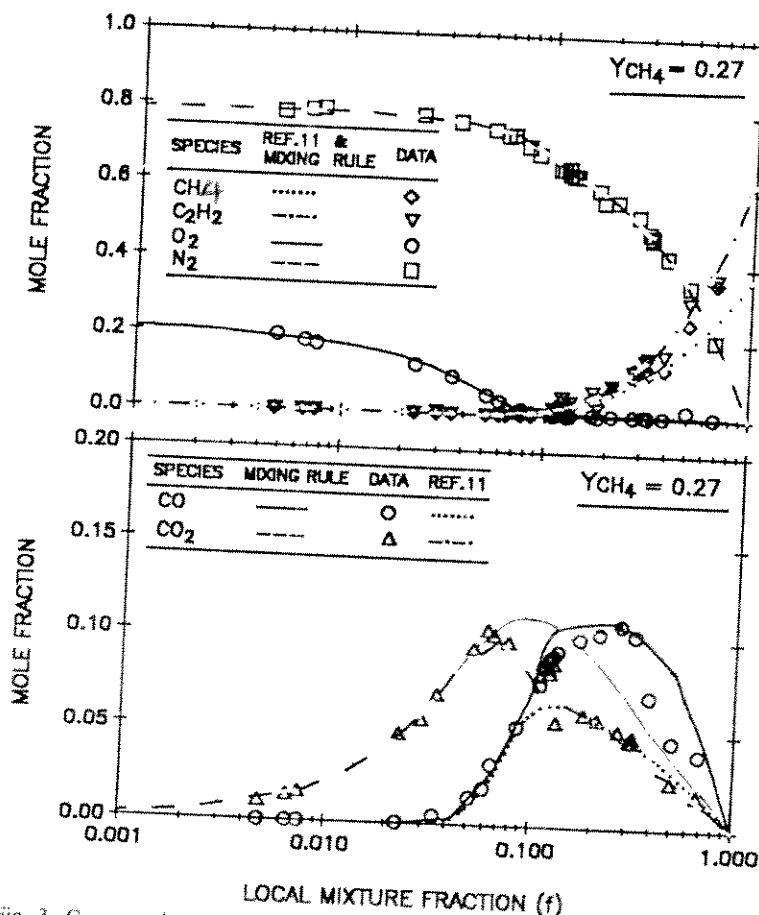


Fig. 3. Concentration measurements for 27%  $\text{CH}_4 + \text{C}_2\text{H}_2$ /air flames and comparison with generalized state relationships of Ref. 11 and mixing rule of Refs. 12 and 13.

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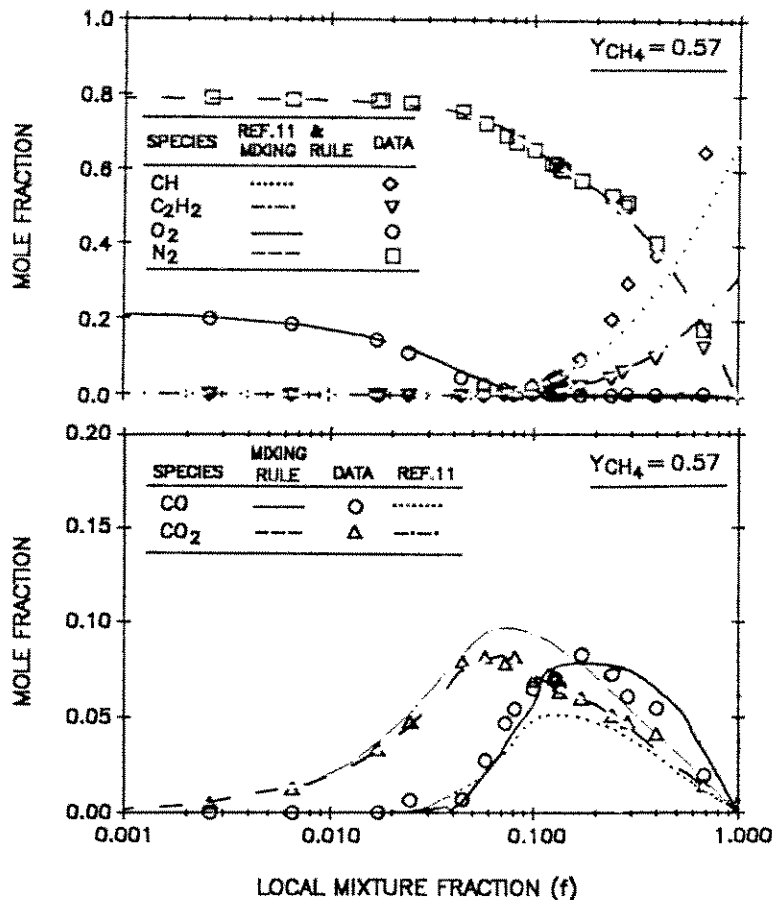


Fig. 4. Concentration measurements for 57% CH<sub>4</sub> + C<sub>2</sub>H<sub>2</sub>/air flames and comparison with generalized state relationships of Ref. 11 and mixing rule of Refs. 12 and 13.

in the fuel-rich region. Since the sooting tendency of the  $R = 1.33$  mixture is lower than that of C<sub>2</sub>H<sub>2</sub> and the  $R = 0.37$  mixture, the discrepancies are somewhat reduced. The mixing rule estimates the CO and CO<sub>2</sub> variations reasonably well in the  $R = 1.33$  case as well.

For both fuel mixtures the estimates of O<sub>2</sub>, N<sub>2</sub>, and fuel based on the mixing rule and the generalized state relationships are identical. This is because the state relationships for these species for the two fuels are in close agreement with the generalized state relationships. For concentrations of CO and CO<sub>2</sub> in the fuel-rich regions, the state relationships of C<sub>2</sub>H<sub>2</sub> for CO and CO<sub>2</sub> depart significantly from the generalized state relationships leading to the differences between these estimates and the experimental data as well as the mixing rule. Thus mixing rule provides a way of accounting for the departures of individual fuels from generalized state relationships.

The present tests have shown the results for mixtures of CH<sub>4</sub> and C<sub>2</sub>H<sub>2</sub>. Sivanthanu and Faeth [11] have suggested that the generalized state relationships have difficulties in the estimation of CO and H<sub>2</sub> (and consequently CO<sub>2</sub> and H<sub>2</sub>O) in regions containing soot particles. In their study [11], C<sub>2</sub>H<sub>4</sub> flames showed these problems in addition to C<sub>2</sub>H<sub>2</sub> flames. Tests for mixtures involving other fuels have not been performed but similar difficulties can be expected with generalized state relationships for aromatic and other sooty fuels.

## CONCLUSIONS

The mixing rules proposed in Refs. 12 and 13 provide good estimates of state relationships for all major gaseous species for the two flames burning CH<sub>4</sub> and C<sub>2</sub>H<sub>2</sub> mixtures. Although a relatively clean burning fuel and a heavily sooty

fuel were selected to provide a stringent test for the concepts, further experimental evaluation using more than two fuels and extension to multi-phase fuel mixtures is warranted.

The generalized state relationships of Ref. 11 provide reasonable estimates of  $N_2$ ,  $O_2$ , and fuel for all the flames considered in the present study. The concentrations of  $CO_2$  and  $CO$  are predicted well for relatively clean fuels and for fuel-lean conditions in sooty fuels. However, concentrations of  $CO$  are substantially underestimated and those of  $CO_2$  are correspondingly overestimated by the generalized state relationship method in the very sooty regions of all flames.

It is therefore logical to use the mixing rules for obtaining state relationships of fuel mixtures when those of the individual fuels are available. When the state relationships for the individual fuels are not available, the generalized state relationships can be used with caution regarding the low  $CO$  and high  $CO_2$  estimates. It is noted that when the state relationships of one or more species of all fuels in a mixture follow the generalized state relationships closely, then the estimates based on the mixing rule and the generalized state relationships collapse.

*The work was completed while the authors were members of the Mechanical Engineering Department at the University of Maryland at College Park. This work was supported by the Center for Fire Research of the National Institute of Standards and Technology, Grant No. 60NANB8D0834, with Dr. D. D. Evans serving as Scientific Officer. The oil well blowout fires program at NIST is sponsored by the Mineral Management Service of the Department of Interior, with Mr. Ed Tennyson and Mr. Charles Smyth serving as program managers.*

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Received 17 January 1991; revised 7 August 1991

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