# Combustion Processes in Two-Layered Configurations

E. E. ZUKOSKI, S. J. TONER, J. H. MOREHART, and T. KUBOTA California Institute of Technology Pasadena, California, 91125, USA

## ABSTRACT

The development of accurate descriptions of flame geometry, and the rates of entrainment, chemical species production and heat release in the plumes of large buoyant diffusion flames is still an important current problem for fire modelers. A description is given here of recent experimental work concerning these problem areas when a small part at the base of a large diffusion flame is immersed in air and the larger part, in vitiated gas.

### INTRODUCTION

During the development of a large fire in a room with an open door, the flame will first be exposed to an atmosphere of air, but later may be exposed to a stratified atmosphere in which the lower part is in air and the upper in a vitiated layer composed of a mixture of air and products of combustion. Finally, if ventilation is restricted, the flame may be completely immersed in a vitiated atmosphere. Each of these environments will have an effect on the flame geometry, rates of entrainment of gas into the fire plume, rates of production of chemical species, and heat release in the fire plume. Each of these features must be described accurately to produce a satisfactory model for the development of the fire.

We have a good understanding of the these features of the combustion process for many fuels burning in buoyant diffusion flames immersed in pure air and some understanding of these processes when the flame is completely immersed in a vitiated atmosphere which still contains sufficient oxygen to support combustion. For example see papers by Tewarson (1986), DeRis (1985), Cetegen et al (1984) and Lockwood (1986).

However, we have a much less well developed picture of the combustion process for flames burning in the two-layer configuration when the larger part of the flame is immersed in products of combustion in the upper layer which models the stratified room conditions often formed in the early stages of a room fire. The purpose of this paper is to give a brief summary of recent experimental work with buoyant diffusion flames burning in this two-layer configuration and several important results of these experiments will be described here.

295

## COMBUSTION IN THE TWO-LAYERED CONFIGURATION

When flames burn in the two-layered configuration, the concentration of oxygen in the upper layer may be reduced to a small value and this almost inert gas will be entrained into that part of the fire-plume which extends into the upper layer. Under these circumstances, the chemical species formed by the fire may be quite different from those found in the products formed by diffusion flames burning in unvitiated air or by flames completely immersed in vitiated gas.

The effects of the composition of the upper layer gases on the combustion process in the fire plume has been reported recently for the steady state case by Toner, see Toner (1986) for more details, and, in addition, Morehart is now examining the unsteady case, in which the stoichiometery of the gas supplied to the upper layer by the fire plume is different from that of the upper layer. Results of both of these studies will be reported here.

The flames considered here all had flame heights which were between 4 and 6 burner diameters high and hence were Regime III flames in notation of Zukoski (1985).

## EQUIVALENCE RATIOS:

Before beginning this discussion, it is convenient to define two equivalence ratios which are used to characterize the chemical species present in a gas mixture which is reacting chemically or which is formed by a chemical reaction. For example, if we are given the species mass fractions in a mixture formed by combustion or partial combustion of a fuel-air mixture, the fuel-air ratio of the gas which was burned to form the mixture can be determined in a straightforward manner.

We want to use this idea to characterize the composition of the gas which is carried through the interface by the fire plume and the gas which is present in the upper layer. Neither of these parameters depends on the degree of completion of the combustion process. The fuel-air ratio of the gas entering the upper layer as a result of the flow in the fire plume is defined as the ratio of the rate at which fuel is supplied to the fire, to the rate of entrainment of air into the flame beneath the interface and is called  $f_p$ . The corresponding equivalence ratio,  $\varphi_p$  is defined as the ratio of  $f_p$  and the stoichiometric fuel-air ratio  $f_s$ .

Similarly, the fuel-air ratio for the upper layer defined as  $f_l$ , is the ratio of the sum of the mass fractions of fuel-atoms contained in all species present in the gas to the corresponding sum for air-atoms contained in the same species. As usual, the equivalence ratio for the layer,  $\varphi_l$ , is the fuel-air ratio normalized by the stoichiometric fuel-air ratio.

Because these flows are turbulent, we do not expect molecular diffusion to lead to any substantial change in the atomic mixture ratio of the gas. Thus  $\varphi_l$  will always be uniquely related to the mass fractions of the atomic species in the gas mixture.

For steady flows, i.e., when the rate at which fuel is supplied to the fire and the thickness of the upper layer are constant, the mass fractions of the elements supplied to the upper layer by the fire plume will be the same as that of the gas within the layer, and then  $\varphi_l$  will be equal to  $\varphi_p$ . In contrast, rapid growth of the

fuel supplied to the flame can produce a situation in which the fuel-air ratio of the layer is much less than that in the plume, and  $\varphi_l$  will be less than  $\varphi_p$ .

## APPARATUS:

In all of the experiments described here the source of the fire was a 19 cm bed of glass beads and natural gas was the fuel. See Cetegen et al (1984) for more details. Heat release rates based on the heating value of the fuel ranged from 20 to 200 KW.

Two hoods were used in this work; the first, used by Cetegen, Lim, and Toner to study the steady flows, was a 1.2 meter cube, insulated on the inside with ceramic fiber insulation board and with the bottom side open. One side of the box contained two pyrex glass windows which were 0.3 meters square and which allow partial observation of the interior of the hood up to heights 0.9 meters above the lower edge of the hood.

Recently a new hood, 1.8 meter square by 1.2 m, has been constructed to study the unsteady flows. An injection system placed in the top of the hood allows gas to be injected into the top 20 cm of the hood from 5 cm lines and though an array of 324 injection ports which are 1.6 mm in diameter. These small holes insure rapid mixing between the injected gas and the flow produced by the fire plume. Gas is injected in a radial direction and roughly parallel to the mean stream lines of the flow as the gas in the fire plume impinges on the ceiling and is turned to recirculate within the hood. The flame lengths are kept below the injection plane to prevent air from being injected directly into the reacting flow in the flame.

With this apparatus, we can control the mixture ratio within the hood independent of that in the fire plume and hence study nonsteady flow conditions such as that described above in a steady experiment.

In these experiments, the hot products were allowed to spill out under the edges of the sides of the hood with a very small velocity. The interface between the vitiated gas in the hood and the ambient air is a laminar layer about 2 cm thick. Several flow visualization techniques showed that the entrainment though this interface was negligible. Hence, when no air is injected into the hood, the stoichiometry of the gas present in the hood is almost identical with the stoichiometry of the gas entering the hood in the fire plume.

Asperated thermocouples are used to measure the gas temperature distribution in the hood and a gas chromatograph system, see Toner (1986), is used to determine the mole fractions of the species present within the hood. Gas temperatures increased about 10% between the bottom and top of the hood but the mole fractions of chemical species are uniform throughout. Minimum residence times for gas within the hoods, based on the mass of gas in the hood and flow rate in the fire plume, were greater than 30 seconds for the either hood.

Beyler (1984) and (1986) has also carried out extensive experiments of this type using a wide variety of fuels. His hood was a cylinder one meter in diameter and about 0.5 m deep. Gas was withdrawn from a point 0.15 m below the top of the hood and as a consequence the interface between the hot and cold gas was 10 to 20 cm thick. Differences in the measurements of entrainment rates made by

the Caltech investigators and Beyler may be due to this poorly defined interface and mixing at the interface.

#### STEADY FLOW EXPERIMENTS:

Studies of the steady flow situation were first made by Cetegen et al (1984) for natural gas diffusion flames. By varying the spacing between the lower edge of the hood and the top of the burner, the amount of air entrained into the fire plume could be varied over a wide range and the equivalence ratio of the plume could easily be changed from 0.05 to 2.5. Work with this apparatus was extended by Lim (1984) and more recently by Toner (1986) who determined the species concentrations with a gas chromatograph system.

CHEMICAL SPECIES: As long as the mole fraction of oxygen in the hood was more than 5% mole, Toner found that the products of combustion were close to their equilibrium values and were quite similar to those suggested by Tewarson. However, when  $\varphi_p$  was greater than one the oxygen mole fraction was reduced to less than 1% in the upper layer and concentrations of the chemical species observed in the hood were far from their equilibrium values for the mixture of elements involved and for the gas temperatures observed.

Mole fractions of four of the species, oxygen, methane and carbon monoxide, hydrogen, carbon dioxide, and water, were measured in the small hood by Toner (1986), and values for the first four of these species, shown as crosses, are plotted as a function of the equivalence ratio for the layer in Figures 1 to 4. The data shown here were taken from all of the experiments carried out by Toner and hence cover a wide range of parameters: fuel flow rates, 2 to 40 g/s; heights of the hood above the 19 cm burner, 2 to 25 cm; mean residence times of gas within the hood, 50 to 400 seconds; and temperature of the gas within the burner, 450 to 850 K. Similar results were obtained with natural gas as the fuel by Lim (1984) and Cetegen(1984), and by Beyler (1984) who used a different type of apparatus and examined a wide range of fuels.

The mole fractions of the species present in the products were well correlated by the equivalence ratio of the layer alone and were independent of the residence time of the gas or the temperature of the gas within the hood even though the temperatures in the hood varied between 500 and 900 K for a given equivalence ratio. This result, which greatly simplifies the description of the problem, suggests that chemical reactions in the flame are quenched at temperatures above 900 K by the entrainment of inert gas into the flame.

The observed mole fractions were shown by Beyler to be a sensitive function of the fuel chemistry. The concentration of carbon monoxide in the gas within the upper layer were as high as 2 to 6% when the mixture ratio entering the hood was greater than 1.5 times the stoichiometric value. These concentrations are high enough to be almost instantly lethal and hence fires of the type discussed here can be potent sources of toxic gases.

**HEAT RELEASE:** The heat release per mass of fuel,  $\Delta H$ , was calculated from the measured species concentrations in the gas within the hood and experimental values (Toner et al 1986) are shown in Figure 5. These values are normalized by the standard heat release for natural gas burning in air  $\Delta H_s$ .

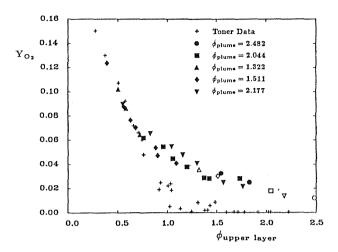


Figure 1: Mole fraction of oxygen as a function of equivalence ratio for the upper layer

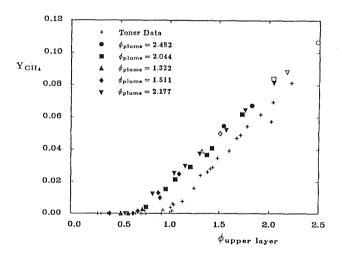


Figure 2: Mole fraction methane as a function of equivalence ratio for the upper layer

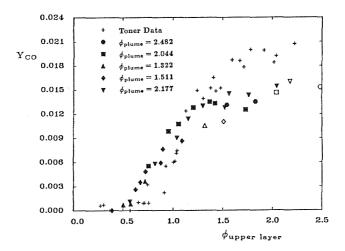


Figure 3: Mole fraction of carbon monoxide as a function of equivalence ratio for the upper layer

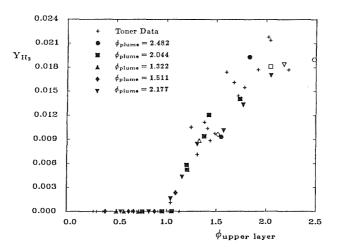


Figure 4: Mole fraction of hydrogen as a function of equivalence ratio

Values of the ratio  $\Delta H/\Delta H_s$  were almost constant and close to one when the equivalence ratio for the plume was less than one. This is plausible because there was more than enough oxygen in the fire plume to oxidize the fuel regardless of the chemical composition of the upper layer. However, when the equivalence ratio in the plume, and hence the layer too, exceeded one, insufficient oxygen was present to oxidize all of the fuel and, in the steady case, there was no oxygen present in the upper layer. Thus, some of the fuel will not burn, and the heat release in the fire-plume will decrease. For these fuel-rich flames, it is more useful to think of the combustion of the oxygen than of the fuel. Consequently, the analysis and observations of Huggett (1980), concerning the use of oxygen consumption rates for the estimation of heat release rates, are pertinent here.

Measured values of the ratio  $\Delta H/\Delta H_s$  are more than 90% of the heat release which would have occurred if the fuel and oxygen present in the plume had reacted at the stoichiometric ratio to form water and carbon dioxide and with the excess species remaining in an unreacted state. This ideal value is shown as the solid line in Figure 5 and this result is in keeping with the observation of Huggett (1980) that the heat released by the consumption of a mass of oxygen during a fire is roughly independent of the species formed during the reaction for most reasonable combustion reactions.

## UNSTEADY FLOW EXPERIMENTS:

When the flow is unsteady, the fuel-air ratio of the gas within the vitiated layer will not necessarily be equal to that in the fire plume. For example in a recent computer model study of a forced ventilation fire with an exponential growth rate for the fire size, the equivalence ratio for the plume gas reached values over 2 when that for the upper layer was still below the stoichiometric value.

In this situation, the possibility exists that oxygen entrained into the fire plume from the upper layer may react with the excess fuel present in a fuel-rich fire plume. Similarly, when the plume is lean nd fuel is present in the upper layer, some of the fuel may react with the excess oxygen in the plume. These reactions could substantially affect the species produced in the flame, the heat release rate and the flame geometry.

Preliminary measurements of species mole fractions measured in unsteady experiments have been made by Morehart in the large hood and the results are shown with the steady state results in Figure 1 to 4. The layer temperature ranged from 500-600 K for these tests. The open symbol in each series shows data obtained in the steady state mode, i.e., without injection of air into the upper layer; the corresponding closed symbols represent data obtained with the same fuel flow rate and interface height but with varying rates of addition of air to the upper layer. The plume equivalence ratio for each symbol, closed or open, is also shown in the figures. As the air addition rate is increased for a fixed plume equivalence ratio, the layer equivalence ratio is reduced.

The most interesting conclusion to be drawn from these data is that the mole fractions for the species are still well correlated by the equivalence ratio for the layer and are independent of the equivalence ratio for the plume. Consider, for

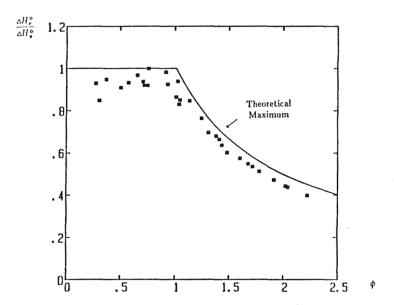


Figure 5: Heat release in fire plume

example, the data for a fire plume with an equivalence ratio of 2.15 which burns in a layer with equivalence ratios between 0.55 and 2.15. This data agrees well with the other experimental results regardless of the plume equivalence ratio and shows the dominant influence of the equivalence ratio for the layer on the products produced when the fire plume has a fixed equivalence ratio.

If this result is verified by our ongoing experimental program and if the correlation is found to be independent of burner diameter, heat release rates, and interface height above the burner, fire models for species production will be greatly simplified.

A second interesting feature of these results is that although the steady and unsteady data fall on the same curve for all of Morehart's data which were obtained in the large, bare metal hood, this curve is slightly different from the steady data of Toner. At present we believe that the most reasonable explanation for these small differences concerns the insulation in Toner's experiments, which was placed ion the inner surface of the ceiling of the hood and hence was directly over the flame and certainly reached temperatures much higher than the average temperature of the gas within the hood. This fibrous material had a large surface area which was covered with soot and may have influenced the chemical reaction rates within the hood by enhancing reactions between residual amounts of oxygen and soot which was trapped on the hot ceramic fibers of the insulation. Such reactions may explain the very low oxygen mass fractions in Toner's data and the differences in concentrations of carbon monoxide and dioxide we find in these experiments.

We do not have information yet for situations in which the equivalence ratio in the upper layer is greater than that in the plume.

## CONCLUSIONS

A description has been given of recent experimental work concerning species which are present in a layer of vitiated gas when the base of the natural gas flame which produces the gas is immersed in air and the larger part of the flame, in the vitiated gas. These experiments model the species production processes which occur in the ceiling layer of a room with a fire. Additional air is added to the upper layer in some of these experiments to model the unsteady development of the ceiling layer.

The experimental results show that the chemical species present in the layer of vitiated gas, called here the ceiling layer, depend primarily on the stoichiometry of the gas within the upper layer and not at all on the stoichiometry of the gas which enters the upper layer through the fire plume. This result is independent of the amount of air added to the upper layer and the temperature of the gas in the layer over the range 450-850 K.

However, these results also suggest that secondary combustion may occur within the layer if surface of the structure which contains it reach temperatures above a lower limit which is certainly above 900 K.

The heat released in the combustion process is within 10% of that which would be obtained if the fuel and entrained air reacted at the stoichiometric proportions and the species in surplus were unchanged by the reaction.

In a later paper we will discuss a method which can be used to predict the source strength which the fire represents for various species present in the ceiling layer when the composition of the layer is changing with time. This method is based on values of the concentrations measured in the unsteady experiments described here. However, given this method and appropriate data, we will be able to predict the variation of concentrations for various species in the ceiling layer as a function of time.

## ACKNOWLEDGEMENT

The authors are pleased to be able to acknowledge the financial and intelluctual support we received form The Fire Research Center of the National Bureau of Standards, the Department of Commerce in carrying out this work.

## REFERENCES

- Beyler, C. L. (1984) "Ignition and Burning of a Layer of Incomplete Combustion Products," Comb. Sci and Tech., 39:287-303.
- Beyler, C. L. (1986) "Major Species Production by Diffusion Flames In a Two-layer Compartment Environment," Fire Safety J., 10:47-56.
- Cetegen, B. M., Zukoski, E. E., and Kubota, T. (1984) "Entrainment in the Near and Far Field of Fire Plumes," Comb. Sci. and Tech., 39:305-331.
- deRis, John (1985) "A Scientific Approach to Flame Radiation and Material Flammability," presented at the Eighth U.S.-Japan Panel on Fire Research and Safety, Tsukuba, Japan.
- Huggett, Clayton (1980) "Estimation of Rate of Heat Release by Means of Oxygen Consumption Measurements," Fire and Materials, 4-2:61-65.
- Lim, C. (1984) "I. Mixing in Doorway Flows, II. Entrainment in Fire Plumes," Aeronautical Engineering Degree Thesis, California Institute of Technology.
- Tewarson, A. (1986) "Prediction of Fire Properties of Materials," Technical Report for the National Bureau of Standards, Center for Fire Research.
- Toner, S. J. (1986) "Entrainment, Chemistry and Structure of Fire Plumes," PhD Thesis, California Institute of Technology.
- Zukoski, E. E. (1985) "Fluid Dynamic Aspects of Room Fires," Proc. of the First International Symposium on Fire Safety, Hemisphere Pub. 1-30.