Flame structure studies: past, present and future*

Robert M. Fristrom

The Johns Hopkins University Applied Physics Laboratory
Laurel, Maryland 20707, USA

Abstract – Scientific observations on flame structure date back to Faraday in the nineteenth century and perhaps even as early as Roger Bacon in the thirteenth century. However, quantitative studies of the structure of premixed laminar flames are more recent. The rigorous formulation of flame theory and experimental methods for quantitative measurements were first developed in the 1950's. This has been an active area of research since that time. The relative strengths of various experimental techniques and data acquisition and reduction methods will be discussed and a brief summary of the information available in the literature will be given.

I INTRODUCTION**

Our branch of science is an ancient and honorable one. The discovery and control of fire was one of the first great inventions of mankind. Its possession and use are one of the criterion used to differentiate "true" man from his ancestors. Archeological evidence places the first use of fire between 500,000 and 100,000 years BP (before present). The artificial production of fire occurred much later in the Paleolithic era sometime between 30,000 and 100,000 years BP. Speculation on fire and flames probably began in the Stone Age and by historic times was integrated into myths and religious beliefs such as the Greek legend of Prometheus, fire's deification in the Zorastrian religion, and the burning bush and hell fires of the Bible. Scientific thinking probably began with the Greek philosopher Heraclitus (ca 500 BC) [1] who proposed fire as the fundamental substance. This was broadened by Empedocles (500-430 BC) [1] who chose fire as one of the four elements of the universe. It is interesting to note the parallelism between his elements and our present classification of states of matter. His air is our gas state, his water is our liquid state, his earth is our solid state, and his fire is our plasma. The four element idea was accepted by Aristotle and his prestige froze these ideas in Western thought until the renaissance. Elsewhere, combustion studies continued. The Chinese invented gunpowder and used rockets. The Byzantines made extensive studies of "Greek fire" flame throwers as a naval weapon and kept it as a closely guarded military secret. By the 16th century, authority began to be supplemented by experiment in Natural Philosophy, as Physics and Chemistry were then called. This initiated the scientific revolution in which we are still immersed. Combustion studies played a prominent role in the beginnings of Chemistry as a separate science and many great names in science have made contributions to combustion studies.

The first recorded probing of flame structure was by Francis Bacon [1], who thrust an arrow in a candle flame and found that the center was cool and hollow and that soot was formed on the outer edge. Somewhat later, Faraday in his Christmas lecture to British children and the Royal family gave a masterly presentation called "The Chemical History of a Candle" [3]. As an illustrative experiment, he inserted a clay pipe stem in the center of a candle and showed that the dark cool center contained combustible fuel and that the combustion products were water and carbon dioxide. We modernists began probing almost a century later, and one early study was made of a candle flame using thermocouples [4] and a quartz microprobe [5]. These approaches are compared in Figure 1.

What distinguishes recent work is that results can be interpreted quantitatively in terms of elementary processes of fluid dynamics, chemical kinetics and molecular transport. Flame theory [6] links combustion with the main body of science and allows us, in turn, to make contributions to the high temperature chemical kinetics and molecular transport.
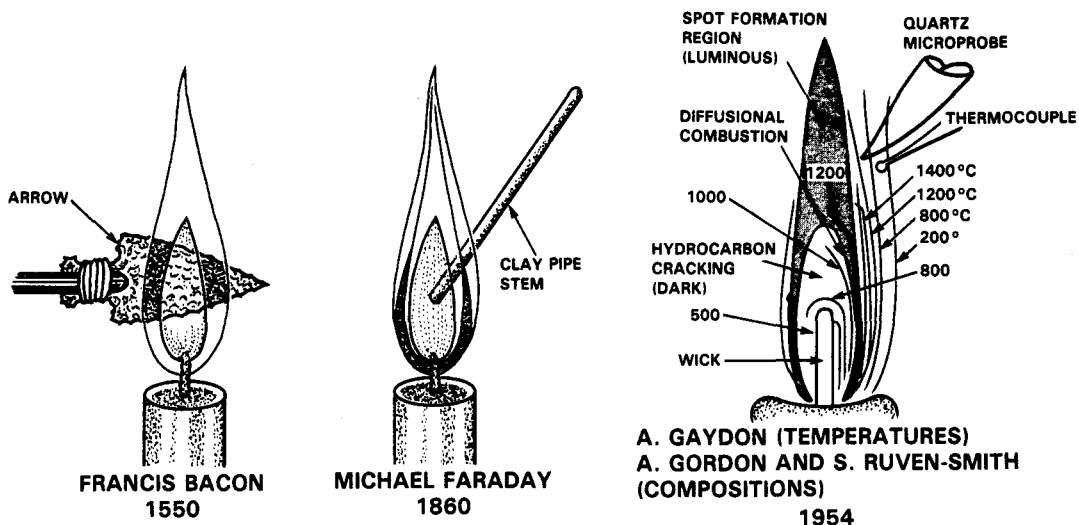
Fig. 1. Flame probing through the years, showing the increase in complexity
         of probing techniques.

Quantitative studies of flame microstructure are a recent innovation. It is the prod-
uct of analytical tools: mass spectrometers, gas chromotography, and lasers with the sup-
port of modern electronics and computers. Flame structure has an extensive literature.
This is the third International Symposium on the subject. Four books [7-10] have been
devoted to Flame Structure, and a fifth has been completed but is not yet published [2].

As you are aware, flames are strongly exothermic reactions with the ability to propa-
gate through space. This results from the strong interaction between chemical reaction and
the molecular transport processes of diffusion and thermal conduction. Flames are examples
of systems with strong positive feedback. Reaction raises the temperature and induces tem-
perature and concentration gradients. These gradients induce fluxes of heat and reactive
species into the unburned gasses through transport processes. This speeds reaction. The
faster the reaction, the steeper the gradients, and the steeper the gradients, the higher
the transport fluxes speeding the reaction. This feedback loop is opposed by flame propa-
gation or by gas flow if a burner is used to hold the flame stationary. Analysis shows
that the system is highly overdetermined and as a result there is a unique propagation
velocity.

II EXPERIMENTAL METHODS AND ANALYSIS

A variety of techniques have been applied to flame studies as can be seen in this
symposium. Specification of a flame requires temperature, gas velocity and composition
profiles as a function of distance through the flame front (Fig. 2). Experimental methods
have been the subject of several reviews [11-13]. Two general methods are employed: one
is physical probes; the second is optical probes. Physical probes disturb the flame. In
compensation they allow great flexibility in analytical methods and are cheap relative to
laser systems.   Optical probes when used rationally are non-perturbing; however, it should
be borne in mind that improperly used lasers are strong enough to cause local electrical
breaknown, and laser-induced reactions could affect results.

A Optical probes

Optical probes are available which can measure all of the properties of interest in
flames. Although in principle any light source might be used, the convenience and power of
lasers have made them the usual choice.   Complete analysis requires several complementary
techniques to accommodate the wide range of concentrations and species types found in
flames. This can be inconvenient since either several lasers are required or a single
laser must be utilized in several set ups. For many species the wave lengths required may
lie in the ultra violet region which is difficult for lasers. This problem can be avoided
by using multiphoton absorption techniques [13,15,17]. One common combination is to use
Raman or CARS spectroscopy [13,15] for the major species, fluorescence spectroscopy [18]
for trace radical species and infrared absorption spectroscopy [19] for hydrocarbon
intermediates. Temperature can be obtained from any of the spectroscopic methods by com-
paring line intensity as a function of energy level. This is a very sensitive method which
is capable of detecting non-Maxwellian distributions. In principle, a temperature could be
obtained for each species, indeed for each vibrational level, if this information is
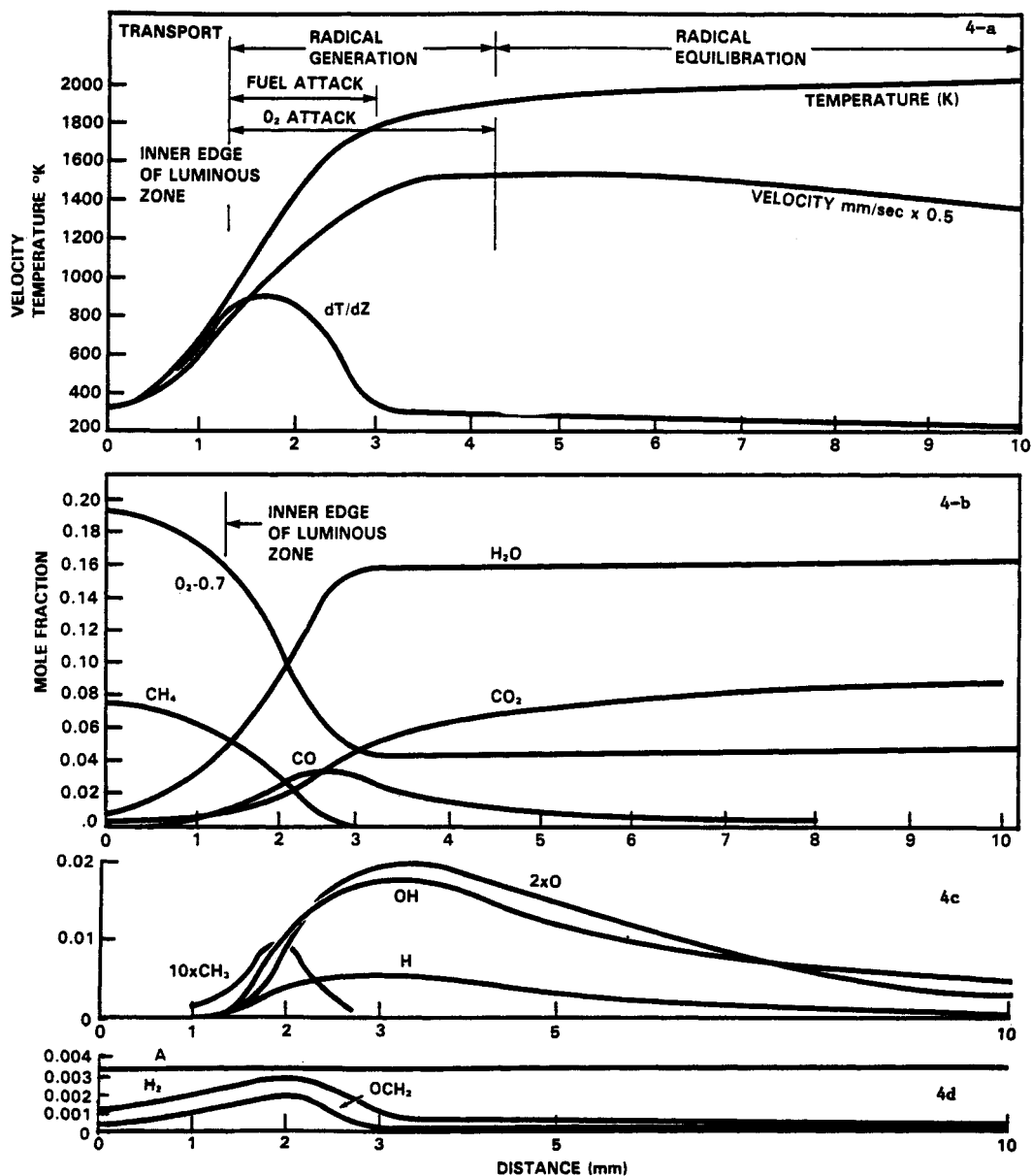
Fig. 2. Typical structure of a low pressure (0.1 atms), lean (0.078 CH_4-0.921 O_2) flame and the relative positions of the zones in the flame. Replotted Fristrom, Grunfelder and Favin [48] with the addition of unpublished work (radical profiles).
  a - Temperature velocity and dominant processes in the various zones
  b - Reactants, products and major intermediates
  c - Radical species
  d - Minor intermediate species

desired. This wealth of information is an overkill at the present time, but in the not-too-distant future this may prove to be a great advantage in studying state-to-state reaction rates. Velocity is the least accessible measurement. It can be calculated from a knowledge of burning velocity if the flame geometry is known. LDV is, strictly speaking, a material probe since particles must be injected. Purely optical methods include homogeneous LDV [20], the phase contrast Doppler velocimetry [21], and the photothermal deflection [22]. Photodeflection can be used for velocity, temperature and composition. However, it is necessary to find a suitable absorption line for each species.

B Physical probes

Physical probes disturb flames, but they are generally less expensive than laser methods. These problems can often be overcome and most of the complete, quantitative stud-

ies of flames have been made using them. Velocity and temperature measurements are
straightforward, but composition requires sampling. A major problem is that of aligning
profiles from the various techniques [2,9]. This is complicated by distortion introduced
by the probes. The problem can be avoided by using a single probe for all measurements.
For example: a single quartz microprobe operated as a pneumatic probe for temperature, as a
Pitot probe for velocity, and as a sampling probe for composition. The most direct tech-
nique for velocity measurement is the particle trace method; for temperature it is the use
of small thermocouples, and for composition, it is microprobe sampling.

There are two general sampling techniques: microprobe and molecular beam sampling.
In the former, a sample is removed, and any suitable analytical technique can be employed;
in the latter, the usual detector is a mass spectrometer. In microprobe sampling, the rad-
icals recombine and quenching can become a problem. These problems can be resolved by
using the scavenger probe technique [23] in which the radicals are reacted with a scaven-
ger to produce analyzing stable molecules. This offers the added advantage of quenching
the sample since flame reactions are all radical driven. Molecular beam inlets offer the
advantage of allowing direct analysis of radicals with negligible quenching problems. The
principal difficulty lies with the disturbance which such a probe offers flames and the
complexity of analysis in cases where hundreds of compounds and radicals are involved.
Such difficulties can be surmounted as Bittner and Howard have shown [24] by identifying
and measuring over two hundred molecules and radicals in the study of a near-sooting flame
of benzene.

Optical methods are particularly well adapted to studying the behavior of one or two
species in a flame system. Direct sampling with gas chromatography/mass spectrometry
probably offers the best technique for mixtures such as those occurring in rich hydrocarbon
flames. Scavenger probing could allow detection and identification and analysis of complex
radicals. Molecular beam inlet is probably the best general technique for radical species.
For complex studies, a mix is best. The physical probes allow complete analysis, while the
optical methods can guard against sampling and flame disturbance errors, and detect non-
equilibrium species. The optimal mix depends on what methods are locally available and the
requirements of the problem.

III DATA INTERPRETATION

To obtain scientifically useful information from flame structure data requires inter-
pretation accomplished through flame theory [2,6,9,10,25] which applies the laws of conser-
vation of energy and conservation of atomic species together with the differential
equations describing reaction and molecular transport. Flames are complicated by strong
gradients which induce large fluxes which vary from species to species so that energy is
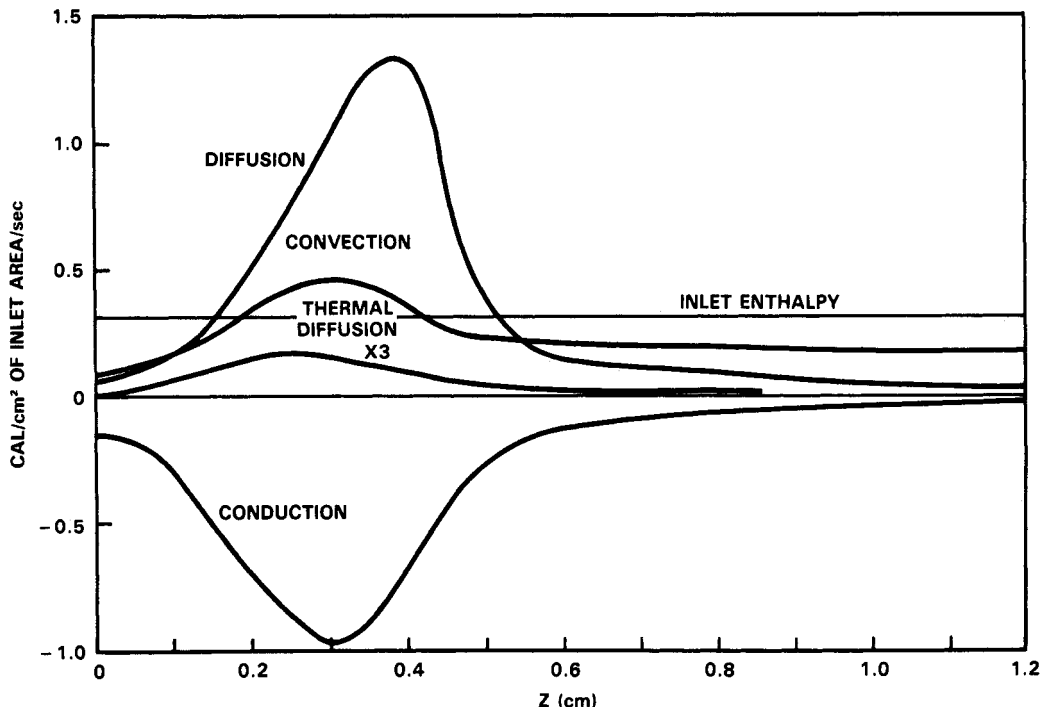transported by convection, thermal conduction and diffusion. As can be seen in Fig. 3,

Fig. 3. Energy transport in a low pressure flame (same as Fig. 2). From the data of
Westenberg and Fristrom [50] with the addition of thermal diffusion.

energy carried by conduction and diffusion is generally larger than that carried by convection; the same is the case for species concentration. Because of diffusion, the fractional flux of a species can differ widely from the concentration (Figs. 4,5) and may even be negative. The consequence of this is that quantitative interpretation of flame structure data requires not only knowledge of local temperature, concentration and gas velocity, but also a knowledge of the local gradients of temperature and concentration and the multicomponent transport coefficients of the species involved. Since rates are derived from the spatial derivative of the fluxes, it can be seen that deriving rates of species reaction and heat release requires a quantitative knowledge of concentration, temperature, velocity and their first and second derivative. Because of dependence on derivatives, the quality of data depends strongly on precision and the knowledge of the transport coefficients.
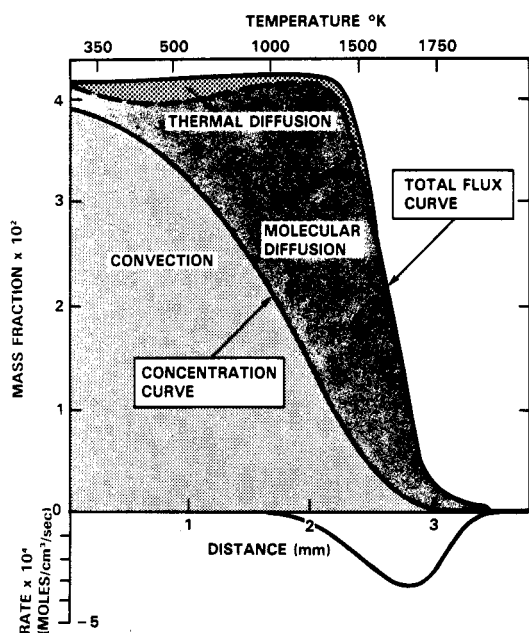

Fig. 4. Relation of concentration, flux and rate for methane in a low-pressure-methane flame.
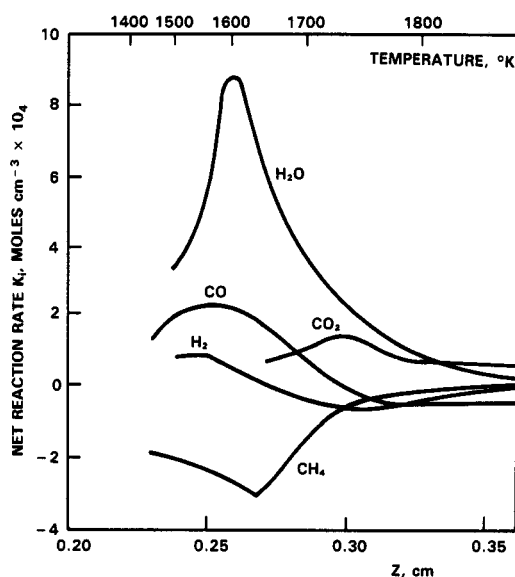
Fig. 5. Net reaction rates in a low-pressure methane-oxygen flame (see Fig. 2).

Manual data reduction is laborious and several programs have been written to reduce this task [25]. General experience is that even the best of data requires smoothing.

Computerized interpretation of data is driven by the exponential expansion of computer capacities. One word of caution: although computer capacities are enormous and easily available, the programming required for these systems is far from trivial, so that one is well advised to make use of as much available software as is feasible. Packages such as the "Chemkin" [16] family are very useful.

Once net rates of species production are available (Fig. 5), it is possible to interpret the information in terms of elementary reaction processes, but this requires selection of a chemical mechanism. Except for the simplest flame systems, this is a somewhat arbitrary process. Two approaches are feasible: the analytic one of testing mechanisms for each species, and the global one of modeling with sensitivity analysis [26].

Flame structure data provide the tools to investigate the mechanism of reaction of individual species. It should be remembered that a single profile does not provide sufficient information to unambiguously derive kinetic constants unless a functional form and mechanism is assumed [9]. For this reason it is usually desirable to study a variety of flame conditions. In many cases, so many reactions are involved that an unambiguous choice of mechanism may not be possible.

In modeling, kinetic and transport coefficients are taken from the literature, a mechanism is assumed, and the flame equations are solved to yield the burning velocity and the temperature and composition profiles. These are then compared with the experimental data. The modeling procedure is considered by others in this symposium. Modeling provides an excellent test of the validity of a chemical model but so many parameters are involved that it becomes a clumsy tool for evaluating the quality of flame structure data. Eventually

more sophisticated approaches will combine the versatility of the analytic approach with the power of the modeling approach.

IV RESULTS

Quantitative flame structure studies date back some thirty years. Hundreds of studies have been made with varying degrees of analysis. Some studies were primarily qualitative or aimed at a single aspect of the flame, while others were completely analyzed. The information is of two general categories: (1) flame processes, and (2) kinetics and mechanisms of flame systems.

A Flame processes

Early flame studies were aimed at establishing the processes involved in flame and the general properties of flames are now well understood.

One-Dimensionality - It is important to establish is the quantitative applicability of the one-dimensional model to premixed flames. This problem was attacked early with the conclusion that with burners of reasonable size, most premixed flames could be quantitatively analyzed with a one-dimensional model providing stream tube expansion was accounted for [2,9]. Consideration of this factor and the effects of using various surfaces of visualization has allowed rationalization of variations of burning velocity which had been a long-standing problem [2,9,27].

Radical Reactions - All of the flame mechanisms which have been established involve exclusively reactive radical reactions [2,9,26,28], that is, species with unpaired electrons. This greatly simplifies flame chemistry since it becomes the high temperature chemistry of a few radical species, principally H, O, OH, HO_2 and the halogen atoms. The key to flame chemistry then becomes the mode of generation of the reactive radicals. In oxygen (CHX) flames, they are produced dominantly by the reaction $H+O_2=OH+O$, while in halogen flames, the dominant process is dissociation of the halogens.

Zones in Flames and the Ignition Approximation - Flames show spatially separated regions: a transport zone, a primary reaction zone, and a radical recombination zone (Fig. 2). The existence of a transport zone implies that reaction does not occur below some critical temperature. This approximation was recognized early and is useful in flame theories [29,30]. It should be observed that the lack of reaction results from a paucity of radicals, not the temperature dependence of reaction, as originally thought, and indeed in hydrogen flames reaction due to H atom recombination can be detected at the hot boundary [28]. The separation of a narrow primary bimolecular reaction zone of radical generation and fuel attack, followed by a slow three-body recombination zone, is an artifact of the pressure regime in which we normally study. If the pressure is high enough, one may expect to find the two zones merged.

Burning Velocity and "Flame Thickness" - If flame thickness is taken to be that of the primary bimolecular zone which in turn controls flame gradients, one might expect a linear relation between burning velocity and flame thickness, and this has been observed and commented on [2,9]. Flame thickness and velocity control the number of collisions available for reaction. If this relation is extrapolated to the limit of sonic velocity, the intercept occurs near the flame thickness which allows only a single collision. This establishes a rough relation between reaction probability and burning velocity.

Diffusion Velocities in Flames - Because of the steep concentration and temperature gradients in flames, diffusion and thermal diffusion are significant. Diffusion velocities can be much larger than the convective velocity (Figs. 4,6,) and are generally larger than corresponding thermal diffusion velocities (Fig. 7).

Lewis Number Approximation - Klein [31] showed that if a flame product or reactant had a Lewis number of unity and its appearance (or disappearance) is linearly connected to the heat release, its concentration profile will be linear in the T-X plane. This is a reasonable approximation for many species in common flames (Fig. 8).

Energy Transport in Flames - There are three modes of energy transport in flames: (1) thermal conduction; (2) convection; (3) diffusion; and (4) thermal diffusion. Simple kinetic theory considerations predict that if all species had unity Lewis numbers, there would be a cancellation of energy transport by thermal conduction and that by diffusion and thermal diffusion [6] leading to a system with constant enthalpy at every point. In real flames, unity Lewis number is poor for many important species. Despite this, there is a rough cancellation and, as Klein observed [31], the constant enthalpy Lewis number unity approximation is a convenient first approximation for most flames. This is the case because common flames which use air as the oxidizer have a major diluent, nitrogen, whose Lewis number is close to oxygen. This overrides the fact that many fuel and product species have Lewis numbers which differ significantly from unity.
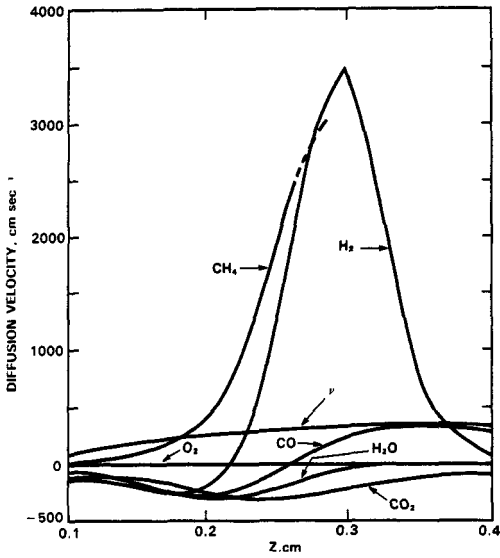
Fig. 6. Diffusion velocity profiles in a
low-pressure methane-oxygen flame
(see Fig. 2). Note comparison with
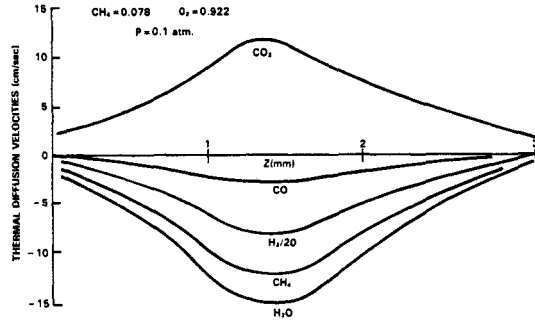mass velocity. After Fristrom and
Westenberg [1].

Fig. 7. Thermal diffusion velocity
profiles in a low-pressure
methane-oxygen flame (see Fig. 2).
Calculated from the data of
Fristrom, Grunfelder and Favin,
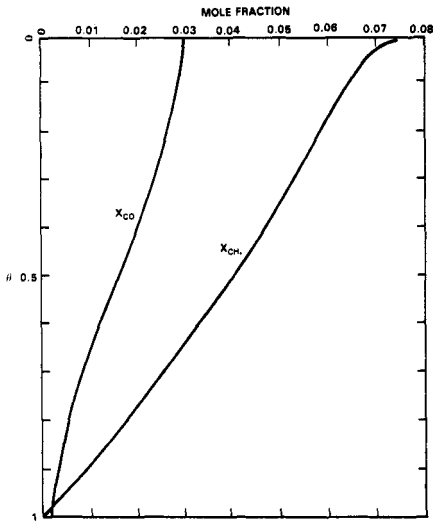using the method of Fristrom
and Monchick [53].

Fig. 8. Test of the unity Lewis number
approximation for a methane-oxygen
flame (Fig. 2). θ is defined as
$(T_f-T)/(T_f-T_o)$ for methane and carbon
monoxide. After Fristrom
and Westenberg [1].

Pressure Scaling in Flames - Because reaction and transport are both collision
dominated, they scale with mean free path and inversely with pressure. Thus this part of
flame structure is approximately invariant when plotted on a scale of distance divided by
pressure. By contrast, the slow recombination zone depends on three-body collisions which
vary cubically with pressure, and the thickness of this zone varies with the three halves
power of pressure. The coupling between the two zones is reduced as pressure is decreased
and the recombination zone becomes so large that laboratory flames become predominantly
bimolecular processes since there are not sufficient collisions in practical sized systems
to allow attainment of total equilibrium. At the other end of the scale, as pressure
increases, the scales of the two regions tend to approach one another and at pressures
around one hundred atmospheres the distinction between two- and three-body processes and
the general kinetic theory approximations must be modified using the Enskog extension [6]
or other formulations.

Partial Equilibrium - Because of the relative speeds of two- and three-body reactions
at normal and low pressures, one would expect to find a region where two-body reactions are
essentially in equilibrium while three-body processes slowly continue. In such regions in
flames the temperature and concentration gradients are also low and the system is essen-
tially at constant enthalpy. Under these conditions there is a unique calculable relation
between composition and temperature [2].

The Steady State Approximation - As a consequence of the strong effects of transport in flames the steady state approximation is not generally valid in flames. However, it should be observed that it is useful at any point where reaction rate of a species passes through zero. For intermediate species there are two such points.

B Flame reactions

Some of the most useful results of flame studies have been the elucidation of the mechanism of flame reactions and in providing high temperature kinetic constants. To summarize the situation one should consider what can burn and, more explicitly, what can form premixed flame systems. Looking at the periodic table of the elements (Fig. 9), one can see that most elements and many of their compounds, particularly hydrides, can be burned by oxygen under suitable conditions. The exceptions are: the halogen elements, the inert gasses, and a few noble and transition metals which have thermally unstable oxides. Even some of these, such as palladium, can be burned as powders, even though when heated to the
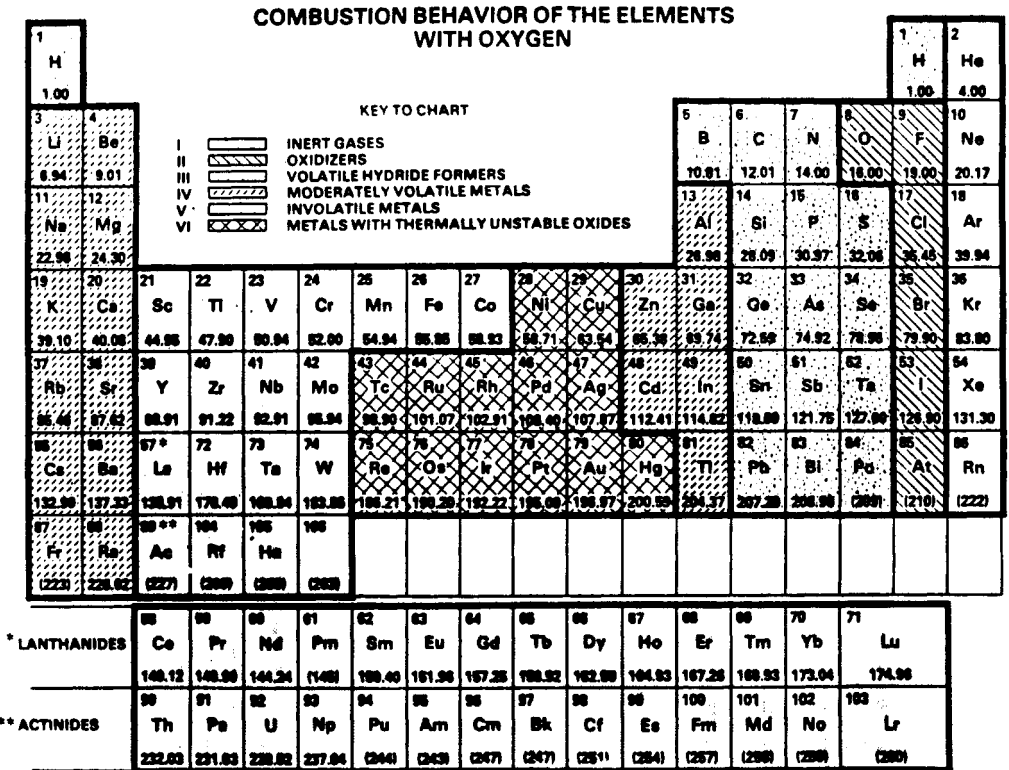

Fig. 9. Combustion behavior of the elements with oxygen, after Fristrom. [2].

higher temperatures of hydrocarbon-oxygen flames the oxide decomposes, regenerating the metals. Similar considerations would apply to the halogen elements (the exception being iodine). Indeed, fluorine burns a wider range of compounds, even forming flame systems with chlorine and many halocarbons. Beyond these reactive elements, there are a host of more complex oxidizers which support combustion. In addition, even nitrogen can burn certain metals, and alkali metals can be burned in low pressure flames with carbon dioxide and halocarbons as oxidizers. Thus, it can be seen that combustion is a widespread phenomena. Premixed flames provide a more restricted field because the reactants must be in the gas phase. This restricts consideration to moderately volatile systems. Nevertheless, the number of possible systems is large. Many systems have been investigated. Figure 10 provides bar graphs indicating the distribution of studies. Systems where modeling has confirmed qualitatively satisfactory detailed mechanisms are available are shaded.

V SUMMARY OF STATUS AND FUTURE PROSPECTS

The study of flame structure has become a well-established discipline with a well-stocked arsenal of experimental methods. It seems worthwhile to speculate on what future directions might be.
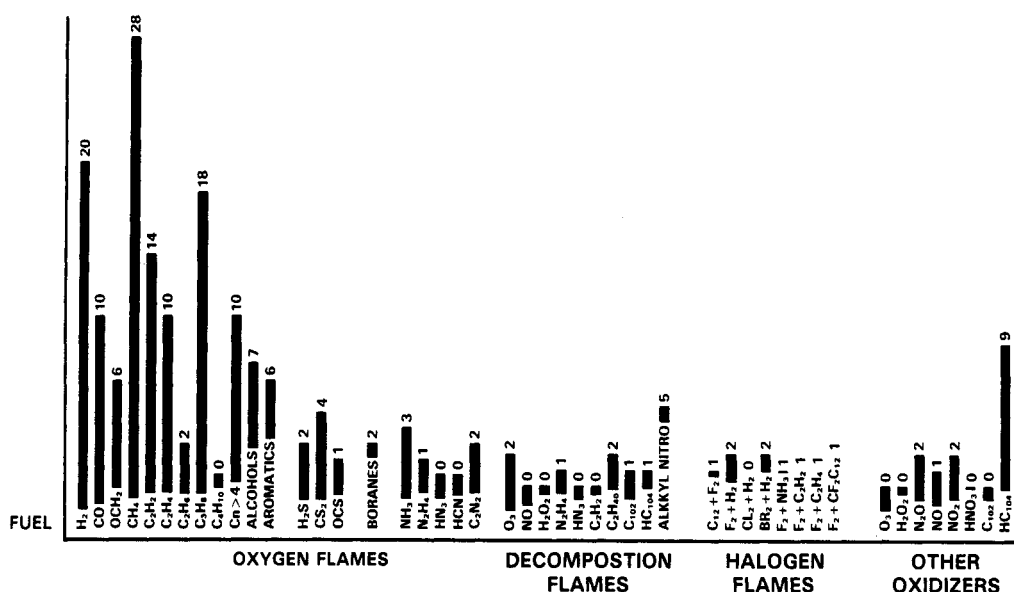
Fig. 10. Distribution of flame structure studies as of 1989.

(1) Systematic Flame Structure Studies - Although there are hundreds of flame structure studies in the literature (Fig. 5), the coverage of flame chemistries is spotty. Ideally, one would like to have available examples of each basic flame system at at least three stoichiometries: fuel-lean, stoichiometric, and fuel-rich, and several pressures. The present status has been documented by the author in a recently completed book [2]. One remedy is discussions at meetings such as this to identify areas where increased coverage would be desirable. This would do much to reduce this problem and will encourage laboratories to channel efforts into obvious holes in the system.

(2) Extension to Higher Pressure Flames - One deficiency in available studies is the structure of flames in the transition pressure regime where reaction processes change from dominantly bimolecular to dominantly termolecular. This transition would begin somewhat above atmospheric pressure and would be dominated by three-body processes between thirty and one hundred atmospheres. The reason for the paucity of studies is spatial resolution since flame thickness decreases with increasing pressure. The best resolution of present studies is around tenth of a millimeter. Conceivably this could be improved at least an order of magnitude, and this would allow the quantitative study of a reasonable range of high pressure systems.

(3) Generalization of Optical Techniques - One of the weaknesses of optical techniques is that complete quantitative analyses are difficult because no single optical method applies to all species with the generality found in mass spectrometry and gas chromatography. A standard protocol which would allow analysis of all species would be a desirable objective. This might well require combining several techniques and a variety of wave lengths. Temperature and velocity would presumably be accessible through auxiliary techniques.

(4) Development of Minimal Perturbing Probe-Sampling Techniques - In the absence of completely general non-perturbing methods, the development of probe sampling techniques which offer minimum flame disturbance would be desirable. Molecular beam sampling generally significantly disturbs flame systems and does not allow the application of separation techniques such as gas chromatography. Quartz microprobe sampling shows much less flame disturbance but normally leaves radical concentrations in doubt and is subject to quenching problems. One solution is the use of scavenger probe [9,23] where radicals are converted into identifiable stable species by mixing the sample rapidly with a suitable scavenger. Quantitative scavenging provides quenching and offers a method for absolute determination of radical concentrations. Deuterium is a suitable scavenger for H, O, and OH, while iodine is suitable for hydrocarbon radicals.

(5) Systematic Methods for Determining Reaction Schemes - The brute force method of measuring flame structure, together with other techniques and trying to fit this data with fifty or a hundred reaction schemes, is inefficient. Even when coupled with sensitivity analyses [26], it would appear desirable to develop systematic experimental methods for probing reaction schemes experimentally to complement the sensitivity analyses of modeling methods. This has the advantage over modeling of not requiring prior knowledge of many

parameters whose precision may not be well known. Isotopic labeling methods offer a model.
Other possibilities might include studies where excited populations were maintained in a
flame, perhaps by putting the flame in a suitable laser cavity.

Extending flame structure measurements and analyses to two- and three-dimensional sys-
tems and time-dependent flame should be feasible, offering more realistic models for prac-
tical systems. This would be facilitated by the automation of experiments and analyses
allowing systematic studies of flames by reducing the effort required to obtaiin data.
This would allow the routine accumulation of useful information.

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