

What is a Flame? A Review of 50 Years of Research

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"The great issues of science are more often qualitative than quantitative"

Introduction. Research on the structure and characteristics of flames has been in progress now for nearly two centuries, but in spite of being able to list well-known flame properties (Table 1), a unique definition of a flame would seem to be still elusive. The question this raises is whether flame properties are so diverse that such definition is impossible, or whether there may be aspects of flame behavior that are yet to be discovered. What evidently is missing is an axiomatic framework for organizing flame properties; and to create such a framework, first, requires answer to the Title question: "What is a flame?"

The answer to this question, as will be shown, is ambiguous; but it provides a direction to pursue. This leads to a conceptual division of flames into three broad categories, as set out in Table 2. The two major Categories, I and II, in this Table broadly represent Combustion Science and Combustion Engineering which, itself, identifies something of a previous disconnect in combustion analysis where engineering systems – notably industrial furnaces – are commonly sidelined in combustion texts. This Table 2 formulation places them more centrally in the general scheme of combustion analysis; it also has significant impact on concepts of flame properties, as will be seen.

There is also a starting thread of axiomatization in this Table 2 Classification, and this is substantially expanded by amplification of Category II, as set out in Table 3. The matter of definitions, nevertheless, is central; and to examine this, we first require some historical context. This also raises the issue implied by the paper sub-title: What period of time was the most active and productive in combustion research? and the pass-out question this also raises is: What might we expect in the next half century?

Historical. Origins. Combustion research can reasonably be regarded as having an ancestry and prologue in the oxidation studies of the last half of the 18th century which *inter alia* demolished the Phlogiston theory and set the basis for modern chemistry. The defining moment for the start of combustion science is generally accepted as the studies by Sir Humphrey Davy in 1813-1815 – in connection with very-practical problems of gas explosions in coal mines – with his two-property demonstration (simultaneously with his invention of the safety lamp) of: low combustion limits in fuel/air mixtures; and variation of flame speed with fuel concentration. In the most general terms, these two properties are central to the two principal characteristics of most flames as set by the questions: "Is ignition maintained, and is the flame stationary or moving?" In that sense, the field of combustion science was largely defined by Davy's discoveries.

Development. Historically, subsequent research was substantially guided by one or both of two needs: first, the need for information -- a data base -- essentially the measurements of flame speeds and combustion limits, and spin-offs from those; and, second, the need for data interpretations, or theoretical development of flame propagation and combustion mechanisms. Significant also were the drivers for the research that shifted with time but were very much dominated through much of the 19th century by considerations of fire and safety, notably in coal mines. On that account, much of the resulting emphasis was on ignition, low limits, and flame suppression; and propagation was investigated more as a consequence of behavior in large scale experiments, with large-scale explosion-gallery tests on gas and coal mixtures dating from the 1870's. In the current (20th) century, the drivers for fundamental investigations moved more to practical problems, notably, after WWII related to gas turbines and rockets, but then later to air pollution and other environmental problems.

It was also implicitly recognized even in the early studies that a data base by itself can have empirical engineering value but, scientifically, it is essentially worthless without (correct) interpretation. The emphasis in the safety-dominated investigations, consequently, was very much on *fundamental* studies, to be able to *understand and control* the practical problems of explosion; at the same time, surprisingly, there was evidently a notable disconnect from the practical problems of industrial combustion, for example, in furnaces (Table 2, Category II flames) which in consequence were developed on a substantially *ad-hoc* or empirical basis. The significant injection of science into combustion engineering appears to have been largely a consequence of World War II.

In the two centuries following Davy, pursuit of the data base development expanded the determination of combustion limits and flame speeds into measurements on fuels other than gases: vapors, solid particles, and liquid drops; and expanded the range of measurements into supportive and supplementary determinations of ignition properties, ignition temperatures, temperature profiles, flame temperatures, reaction times, and ultimately of reaction rates. The results, in general, showed variations in numerical properties from fuel to fuel but substantially, otherwise, showed the same qualitative patterns of behavior: there were upper and lower combustion limits; minimum ignition energy requirements; and flame speeds and temperatures that varied with

concentration in substantially the same pattern of: lower values at the limits; and peaking near the stoichiometric.

Quantification. Quantitatively, the pattern of results for different fuels was both different and the same. In the "base-line" laminar flames (Category I of Table 2) for different hydrocarbon fuels – gases, liquids, and dispersions of particulate solids – reaction times or flame thicknesses are very fuel-type dependent: thicknesses of laminar flames range from 1 mm for gases to 1 m for pulverized coal, a range of three orders of magnitude. Likewise, propagation mechanisms range from conduction to radiation. Peak temperatures, however, are very similar, with energy-densities for stoichiometric fuel-air mixtures averaging close to 100 Btu/ft³ for hydrocarbon gases, liquids, or dispersions of particulate solids, and generating T_{ad} values of about 2000°C (3600°F). More surprising, laminar flame speeds and low limits, also, are essentially fuel-type independent, being roughly the same or of the same order, with most laminar speeds (S_u values) in the range half to one meter per second, and low limits at about 100% excess air, corresponding to mixture energy-density values averaging 50 Btu/ft³ or half that of the stoichiometric.

In fixed beds of particulate solids (e.g., lump coal), values (of flame speeds and reaction intensities) were different again, but historically such systems were generally treated or implicitly regarded as outside the "main" stream of combustion theory and development, although latest developments in "Filtration" combustion (Table 3, Class 11.2), as set out below, may change this perception. Flame spread, ignition and extinction behavior, and batch combustion (Category III of Table 2), likewise, were typically treated substantially on a stand-alone basis, and this is seen in the ordering and emphasis in today's standard combustion texts. Much of this reflected the disconnect between combustion science and combustion engineering noted earlier.

Associated with all this was the complementary development of steadily improving instrumentation, with increasing ability to make measurements of local temperatures, concentrations, and velocities, and fluctuations of those parameters, together with fast data recording and analysis. The development of optical methods, particularly by lasers, allowing non-intrusive measurement was particularly valuable. Such measurements are critically important, but less as part of fundamental combustion studies than as supporting techniques for accurate development of phenomenological descriptions, and for testing analytical predictions. Analysis, nevertheless, remains hostage to correct interpretation, preferably mechanistic, of the observed phenomena, and this remains the central issue even at this time.

Combustion Engineering. In parallel with this history, but with long pre-cedents, there were major but largely-independent developments in combustion engineering. Combustion science and engineering have now almost merged, but the historical account shows their effective independence until relatively recently. Even apart from use of flames in heating caves (with a "million" year history), the use of furnaces – and concomitant control of fire – for pottery, brick, and metals production has origins 10,000 and more years back. Additionally, in the last half of the 18th century there were three significant advances: first, James Watt's development of the steam engine, of which the invention of the condenser – a (recuperative) heat exchanger – was a crucial part; second, in the 1790's, the invention of the hot blast – another (regenerative) heat exchanger application – for blast furnaces (a form of Filtration Combustion) which transformed iron manufacture; and third, in the same decade, the start of coal gasification in coke ovens that subsequently, with pipelined distribution (already significant by the 1820's), ultimately transformed street lighting, home heating, cooking appliances, and industrial furnace operations. For the coal gas from coking ovens, with a typical content of 50% H₂ and 35% CH₄, the world was half way to a hydrogen economy two centuries ago. These industrial operations are presented in Table 2 as Category II and in Table 3 as Class I applications.

These developments all represented major commercial use of combustion in engineering systems, albeit developed largely empirically, but with considerable sophistication even in the early 1800's when combustion science studies had barely started. Through the following 19th century, these applications were extended by development of more advanced, high-temperature regenerative furnace systems, notably, in addition to bricks and refractories, for glass melting, and for steel making using the Bessemer and the Open Hearth (the BOF is a 1940's development, a century later). The significance of the (regenerative) preheat was the, generally unrecognized potential, jointly, for super-adiabatic flame temperatures, and for stabilization of high velocity flames. This meant that some major technological problems, notably the fast flame stabilization, had already been solved, by empirical development, long before they were even recognized and defined in scientific terms.

Scientific Elements. The scientific aspects starting in the early 1800's could not, in fact, be addressed until key theoretical concepts had been developed. The two key theoretical elements required in analysis of even the simplest 1-D (Table 2: Category I) laminar flames are heat transfer and kinetics; and first applications of these elements to flames, in the Mallard and le Chatelier (MLC) model, were in the last half of the 19th century with prediction of flame speed and of the temperature profile through a flame. This became the prototype for subsequent flame models involving other modes of heat transfer (radiation and convective exchange), with extension then to analysis of other flame types, notably turbulent and 3-D flames. In that first laminar flame model, (conductive) heat transfer dominates the initial temperature rise up to "ignition", but some form of kinetics assumption is required from ignition through the reaction zone.

Separately, this model also re-raised the question of definition of ignition, in this case, in mid-flame; this problem was the practical driver both in Davy's original studies of gas combustion

and in Faraday's studies in 1844 of coal ignition, also in connection with coal mine explosions. It was not until the mid-1930's that the Semenov Thermal Explosion Theory (TET) provided a (thermal-based) theoretical model for batch ignition. The definition of ignition in a continuous (Category I) flame is still open, however, although the gap was potentially closed by Vulis' (late 1940's) treatment of the continuous flame as a PSR sequence with a definable TET ignition cell in the middle of the flame. This was updated in 1974 but is still incomplete.

The theoretical background needed for these flame model developments in the preceding century required two fundamental insights. The first was elimination of the Caloric theory, in the first half of last century, which played the same blocking role in thermal sciences that the Phlogiston theory had played in the chemical sciences half a century earlier. The second was formulation of kinetics, in the second half of the century, initially specified as a phenomenological statement in the Guldberg and Waage *Law of Mass Action* (LMA), and then importantly extended by the introduction of the Arrhenius statement for the velocity constants. This phenomenological model itself then opened up enquiry into the mechanism behind the LMA statement, and the answer that emerged, in the first half of the current century was the Bodenstein dissociation model leading critically into understanding of chain reactions and consequences of that behavior that, for gases particularly, could result in escalation to explosion and detonation.

In its turn, the "mechanism" of dissociation was identified as being, simultaneously, a phenomenological behavior in its own right, implying thereby a more detailed underlying mechanism, and this led to the postulates of the activated complexes and then, in more detail, the study of molecular orbital trajectories as the basis for *ab initio* calculations of reaction rates. Both these further approaches are major stand-alone developments in chemical physics; for purposes of combustion studies in flames, however, phenomenological kinetics mostly suffices.

Even so, only by the middle of this (20th.) century was the dissociative kinetic basis sufficiently established that attention could be turned to extensive measurement, and development of a kinetics data base. This first required identification of governing elemental reactions with debates continuing, for example, as late as the 1950's on whether H or OH was the dominant radical for hydrogen combustion. Even in the 1960's, for lack of adequate information on elementary reactions, and/or inability (for lack of computing power) to handle the equations sets involved, there was still continuing use of rates measured in global or quasi-global terms for fuel reacting, one-step, to end products (CO₂ and H₂O), or slightly more elaborately, reacting first to CO and H₂ with a selected suite of the hydrogen reactions to complete the model.

The developments since then, largely in the last 30 years, with the almost explosive growth of data on elementary reaction velocity constants, requires no further comment. This has been further supplemented by development of model codes, first for calculating equilibrium properties and then, in the last decade or so, kinetics codes, combined in many cases with flow codes to be able to calculate complete flame behavior in a flowing stream. This, however, may in some instances be a double-edged problem, if the codes - particularly the flow elements - are extrapolated outside the window of verification.

These developments represent major understanding of and potential for applications of combustion theory to engineering problems and (with some reservations) this procedure is now largely standard practice. Nevertheless, a potential for unification existed half a century ago, and it still deserves examination. This combines the Vulis PSR-sequence model for a 1-D flame already mentioned (in connection with in-flame ignition) with the Bragg Criterion for (gas-turbine) combustion chamber design.

A Unifying Factor: The Vulis Model and the Bragg Criterion. The Bragg Criterion (1953) for combustion chamber design resulted from generalization of a detailed analysis of the Rolls Royce Trent jet engine, with the conclusion that: the ideal (design target) mixing configuration should be: a Perfectly-Stirred Reactor (PSR) inlet segment for fastest ignition, followed by a Plug-Flow Reactor (PFR) segment for optimum burn-out. The *engineering* problem of creating the PSR/PFR fluids mixing configuration is a major one, but separate. Importantly, however, it provides a clear target for the design intent; it also provides a link to the Vulis model.

The relation this has to the Vulis model then derives from the question: In the Vulis PSR-sequence model, what is the effect of changing the size of the PSR cells? This was explored in a separate publication (1974) obtaining the general result that: in the flame region *before* the point of inflection (ignition point) in the T-t curve, increasing the PSR size results in faster combustion or reduced time to ignition; and, after the POI, for fastest burn-out, requires the reverse, namely, reduced cell size. In the limit of a single cell for ignition and multiple cells for burn-out, this corresponds exactly to the Bragg Criterion.

Review of actual engineering (industrial) furnaces and burners, very largely developed empirically, typically show an approximation to this PSR/PFR mixing configuration. This will be more evident in what follows.

This provides the summary background to consider the definition of a flame.

What is a Flame? A flame is something that can always be identified as such after the fact, but an *a priori* definition that is *necessary and sufficient* does not seem to exist. Common flame properties often incorporated in "definitions" are listed in Table 1. None, however, is unique.

Property 1 is denied by pyrophoric and hypergolic materials; an example is pyrophoric ignition of very fine iron particles blown into air. *Property 2* is common to many reactions that are not combustion. *Properties 3 and 4* are denied in the limiting case of the Perfectly Stirred

Reactor (PSR) when a flame front and thus a flame speed can not be defined, as discussed in more detail below. *Property 5* is denied when there is thermal backmix, the Category II of Table 2. *Property 6* is reversed when there is thermal backmix, and the statement inverts to become: "When the flame is stable, the (local) flame speed and mixture speeds are in balance" (essentially, then, a trivial consequence). *Property 7* is denied with preheat: combustion limits are extended -- the requirement is for maintaining the sensible plus chemical energy above a minimum energy density of about 50 Btu/ft³. *Property 8* is denied under a range of thermal feedback conditions (see Category II of Table 2; also Table 3). *Property 9* is not unique to flames: hot materials, in general, radiate in the visible; moreover, radiation from hydrogen flames is not in the visible.

Flame Categories. This failure in finding a unique property for definition of a flame suggests that the factors governing in flames may be incompletely identified. An alternative approach suggested by examination of the Table 1 listings, and the comments above, led to the first two (primary) alternative Categories of flame types identified in the classification of Table 2. The key distinction between these two Categories is whether the flame speed is an *independent* or a *dependent* property of the fuel mixture and of the combustion system. However, study of Filtration Combustion (Class II.2 in Table 3) suggests that even this may turn out to be either simplistic or limited. To put these in perspective requires separate evaluation. Flame stabilization is examined first.

Flame Stabilization. Classical concepts of flame stabilization are formulated in terms of velocity balancing. The concept originates with 1-D laminar flames and has been translated, in turbulent flames, to local behavior. For the standard one-dimensional (1-D, MLC) flow system, the flame speed depends only on the mixture composition. The argument is that, if flame is propagating down a tube, against no flow or low velocity flow, then the flame will be stabilized if the mixture-approach flow-velocity is increased until the velocities balance. This is a valid representation in plug-flow systems, and in similar systems where there is no back-mix such as the Bunsen burner; but -- possibly excepting the Filtration Combustion (FC) systems, as already identified -- the velocity-balancing argument can fail if there is any degree of thermal preheat by backmix or other method.

In flow-backmix and heat recovery or Thermally Assisted Flame (TAF) systems (Table 3), which thus includes nearly all practical (engineering) flame systems in furnaces or engines, the incoming mixture is preheated in one way or another, and the flame speed is increased. The flame speed, however, is then a dependent property of the system. In the case, for example, of a standard unswirled jet burner firing into a furnace in an SE (Sudden Expansion) configuration (a Table 3 Class II.1 device), the jet velocity is commonly of the order of 10 m/s, but the fundamental flame speed for the incoming mixture, whether gas, or oil, or pulverized coal, is usually about 1 m/s. The flame, nevertheless, is stable on account of the backmix flow of hot combustion products generated by the momentum of the incoming jet. The aerodynamics of this mixing behavior is key, and is a pattern studied intensively through the late 1940's to early 1960's, notably by the IFRF but also by many others.

The standard interpretation of that behavior is that the flame is then stabilized on account of the *increase in flame speed* due to the backmix-governed preheat. The conclusion is arguable, however, even for this single unswirled jet configuration since the degree of backmix-preheat -- and thus the increase in flame speed -- depends on the primary jet momentum so that the flame speed is now a *dependent* not an independent property of the system. In other words, "Which is the cart and which is the horse?" The flame "speed" can be changed for the same mass flow of incoming mixture solely by increasing the jet velocity (for example, by reducing a pipe diameter). Indeed, with increased jet velocity, the flame generally moves *upstream* (up to a final blow-off or blow-out limit).

That argument becomes even less supportable if the degree of backmix is proportionately increased, for example, by introducing swirl or double swirl or additional down-stream jets, to the point that the primary flame region converges to an approximation of a Perfectly Stirred or zero-dimensional (0-D) Reactor. At that limit, ignition is then distributed throughout the combustion volume; thus, there is no formally definable "flame front" where ignition starts; a flame speed can not therefore be defined; flame stabilization no longer depends on velocity balancing, and interpretation in those terms thus becomes meaningless. Separately, this also has consequences for burn-out and combustion efficiency; this is considered later.

The clearest practical demonstration of this 0-D structure is the Putnam "Octopus" burner, consisting of 8 raw gas jets at the corners of a cube, directed at the cube center. The flame produced is substantially spherical, inside a flame envelope, but there is no flame "front" in the conventional sense of the word so, supporting the statements above, no flame speed can be defined, and there is therefore no potential for defining velocity balancing. Likewise, there is no "flame holder", nor flame "attachment" to a holder (behavior commonly identified in burner studies and assumed to be significant). Since velocity-balancing is evidently invalid as explanation for the flame stabilization, some other criterion is required.

Intuitively, there would appear to be two criteria for stabilization of such a flame, one thermal and one mechanical. The evident thermal stability condition is that the reacting mixture in the flame zone satisfies the standard PSR or WSR thermal extinction (TET) conditions (based on balance between thermal generation by reaction and thermal loss by convection/radiation). The mechanical stability condition is thought to be a zero momentum integral over the surface of

a Control Volume enclosing the flame. For other standard jet flame systems (straight jet, or swirled, or other) the same conditions could apply: it requires definition of a relevant CV inside the combustion chamber, to which the two criteria proposed may then apply. This needs to be examined further; first steps have been taken resulting in definition of an "information flow path" as part of the required CV definition, but the results at this time are essentially open-ended.

Thus, in support of the earlier assessment, this view of flame stabilization clearly incorporates all the thermally-assisted systems listed as Class I and Class II.1 in Table 3. The essential characteristic of the thermal assist in these two classes is that it is provided either externally, through heat exchangers, or internally but by a (fluid) backmix flow process. The focus of flame stabilization in practical terms is being able to design engineering devices in which flame stability is reasonably assured over the expected operating conditions, such as no *flame-out* in a jet engine at 35,000 ft, for example.

As also previously noted, however, this approach does not address the further class, Class II.2, of Filtration Combustion. This is also one that also operates with thermal assist; the thermal assist in this case, however, is in the form of a direct "counterflow" to the mixture flow. Consequently, it presents ambiguities, and may in fact be a stand-alone case.

Filtration Combustion (FC). This is defined as combustion of a reactive gas in a porous bed and, as listed in Table 3 (Class II.2) has two sub-sets: (1) systems in which the gas reacts with the porous medium, such as oxygen in air reacting with coke or coal; and (2) a fuel mixture such as methane-air reacting in the bed pores. Filtration Combustion has been proposed as a new category of flame types but, in fact, it has a long though largely unrecognized history. The second sub-set [Class II.2(2)] has at least a two-decade history of study, but the history of the first sub-set – study of coal combustion and sinter beds – is over a century. Recognition of the commonality of the two sub-sets is also very recent, certainly within the last decade; likewise, essentially complete solution of the governing equations (for combustion of anthracite in a fixed bed) with experimental verification is also as recent (1984). As mentioned earlier, such combustion systems have commonly been regarded as stand-alone or non-mainstream. What is important in this new recognition is the degree to which this flame Class can evidently co-ordinate a range of apparently disparate reaction systems, unexpectedly including, as a limiting condition, the 1-D, Category I flames, as will now be shown.

The physical system consists of a porous bed of particles or porous sintered block with reactive gas or gas mixture flowing through. The reaction is in the gas phase and/or at the surface of the particles. Heat released by reaction raises the temperature of both the gas mixture and the porous solid, and generates different temperature profiles that will also cross. Heat transfer required for ignition of incoming material is by conduction through the gas (as in the Category I flames) but also by radiative-conduction through the porous solid which provides the thermal assist. At any local point in the bed a key element is the (conductive/convective) heat exchange between the gas and the solid, governed by a heat exchange coefficient, h . The complete system is then described by two DE's of substantially identical form, namely that of the MLC equation, but with an additional interchange term involving h , and with different parameter values for the gas and the solid phases.

The outcome of the process, predicted theoretically and supported experimentally, is that 1-D flames can behave in the classic manner of the MLC (Table 2, Category I) model, showing blow-off and flash back. Unlike the basic MLC model, however, the flames show "unusual" characteristics, notably: flame stabilization at very much higher fluid flow speeds than for the mixture without the porous body support – as in the other TAF systems; also, superadiabatic flame temperatures; very low limiting blow-off velocities, as low as 1 mm/s; and extended combustion limits.

As already noted above, however, with the exception of the very slow blow-off velocity, these "unusual" properties are shared by all the other Thermally Assisted Flame (TAF) systems although this is not commonly identified. In addition, however, a different property also exists that depends on the heat exchange coefficient, h , between the gas and solid. If this is progressively reduced, the reduction is effectively equivalent to progressive reduction of the solid density. In the limit, corresponding to no solid bed, the thermal interchange term in the equations vanishes, and the effect is to reduce the two governing equations to one, namely the MLC equation for the combustion of a free gas mixture; thus, the system converges to a Category I flame system. The outcome is a possible change of focus. We might reasonably regard the twin DE's of the Filtration Combustion (FC) set as the base DE's for propagating laminar 1-D flames; and the original classic MLC system then defines a limiting or special-case boundary behavior for zero porous body density. In this sense, it is the original MLC flame rather than the FC flame that might then be seen as "anomalous".

This alone suggests a need for a re-evaluation of the Category I flame properties. It sets the "peripheral" engineering systems of fixed beds (coal, coke, MSW, etc.) in a central role. Most particularly, however, it challenges even the classic 1-D (MLC) system as defining a "fundamental" flame speed. The same gas/air mixture composition can have different stationary flow velocities, i.e., flame speeds for different porous configurations and materials. What then is so special or "fundamental" in the case of propagation in gas phase combustion governed only by conductive heat transfer as in the MLC flame?

Review and Evaluation. This representation of 1-D flame systems as members of a general Filtration Combustion set, with the MLC flame as a special case for an infinitely porous solid, can now be contrasted with the PSR or zero dimensional (0-D) limit of the Category II (thermal backmix) flames. What are commonly regarded as 2-D or 3-D flame systems are definable as incompletely 0-D with 1-D components (i.e., Well Stirred as contrasted with Perfectly Stirred). The initial emphasis for nearly all flames of *practical* interest is then on their 0-D or near 0-D characteristics at the burner: notably, the mechanical and thermal stability of a relevant CV, if such can be defined. This focus transfers attention away from flame speed as a unique governing property, and one that has been a major focus of many past flame studies. This transfer of attention gains particular significance when examining practical flame systems which by default are nearly all turbulent. It is in turbulent flames that the idea of flame front and flame speed need particular examination.

Adequate discussion of turbulent flames is not possible here for space limitations. Nevertheless, to identify key elements, studies of turbulent flames commonly show dispersed regions of reaction, and a common view of the development of the reaction through the flame zone is that it can be represented in many cases by contorted surfaces propagating into unignited fuel mixture. The detailed modeling of the behavior is then addressed by such means as strained wrinkled laminar flamelet analysis, assuming the flamelet is "thin", or by distributed reaction zone theory for thick flamelet or reacting regions. The pre-supposition here is that the unignited region requires transfer of heat and/or dissociated species for extension of the reaction (flame propagation) into the fresh mixture, as in the standard laminar flame model. For a turbulent jet flame in an open cold environment this has substantial relevance. For a jet in an enclosure such as a furnace, however, where measurements typically show substantial temperature fields adjacent to the jet, the question may be less to do with what it takes for ignition and more to do with why flames extinguish: in particular, with distributed reaction zones found in turbulent flames, there can be local extinction of identifiable volumes.

In the form of the existence of low limits, the problem of flame extinction is, in fact, one of the two critical characteristics first addressed by Davy in 1813, and still essentially unresolved in spite of many studies. A major common factor, identified by Burgess and Wheeler in 1912, is that the low combustion limit, calculated as an energy density, is approximately constant at about 50 Btu/ft³ for all hydrocarbon fuels, solid, liquid, or gas. This has never been elaborated except to the demonstration that the (50 Btu/ft³) critical energy density can also be satisfied by the sum of chemical (reaction) energy and sensible energy from preheat. It is this that allows reduction of the low limit by preheat. The reason for this is still unidentified but it could be key both to a solution to that problem and to a final interpretation of flame behavior.

Summary and Conclusions. The conclusion that emerges from this review is that, if a common organizing principle exists, that can be used to create combustion studies in an axiomatic framework, that organizing principle does not yet appear to have been identified or formulated. However, this evaluation also provides a framework to define the direction of continued research, starting with a review of combustion knowledge with the objective, if possible, of creating an appropriate axiomatic framework. A starting point and current example is the Filtration Combustion concept, as discussed above. More generally, there is the potential identified by the classification into Categories I, II, and III. The direction taken to develop such a framework could then start to set the general agenda for the next phase of work into next century. This can be addressed at two levels: the "tactical" level, and the "strategic" level.

At the tactical level, this concerns possible theoretical/analytical procedures (but appropriately including advanced experimental methods for more detailed and accurate determination of reaction processes). Of possible analytical procedures, there are four in particular that would seem to justify particular attention. The first is the use of integral formulations of the governing equations of different reacting systems. The oldest and best known of these procedures is the Rankine-Hugoniot analysis. These well-known formulations are generally limited to 1-D systems, however, and it is a question to consider whether similar analysis of more complex geometries might not be rewarding. It has been successful, for example, in application to diffusion flames, and also to flame spread, leading in this latter case to the inverse fuel-density relation governing flame spread rate. The potential for other configurations needs to be explored.

Less well known is the Furnace and Engine Analysis procedure that provides an equation (of common form) for the Firing Curve for any furnace or engine. This provides an immediate and common integral analytical framework for engineering devices that also defines many of the first-level combustion problems that are found inside the devices. This is substantially developed. Wider use essentially needs only appropriate attention.

A third procedure that is now being pursued aggressively, that is mentioned here for note, is analysis by Deterministic Chaos. Current application to fluid beds and to i.c. engines shows the versatility of the approach. This procedure, in particular, is showing value jointly in practical engineering application and, at the scientific level, in improving precision of knowledge. To a degree, this is also an integral approach. Significantly, this is removing the past constraint of linearization of non-linear behavior that has disguised much real behavior of both practical and intrinsic interest.

A fourth procedure that beyond statement about a decade ago is not known to have been used, is the Species Stream Function (SSF). What this is potentially capable of doing, as shown in the original SSF paper by re-analysis of the classic Burke-Schumann diffusion flame, is to

determine the trajectories of the reacting molecules (and thus their temperature and, potentially, their reaction histories), and simultaneously determine the fuel flux density arriving at any location on the flame or reacting surface. Intuitively, this would seem to be relevant to analysis of turbulent flames.

At the strategic level, this concerns the possible axiomatization of combustion concepts, particularly incorporating and merging both combustion science and combustion engineering. One example is provided as already discussed by the new developments in Filtration Combustion where this can be seen as an organizing principle that merges propagating 1-D gas flames, either free or in porous bodies, and combustion in solid fuel beds. A second example is the potential of the Vulis PSR-sequence flame model combined with the Bragg Criterion. Similar critical examination of the reality of the other entries in the Thermally Assisted Flames category is also needed. All these approaches would seem to have a useful degree of organizing principle even though, as already noted, this still excludes the major areas of, for example, fires and flame spread. Further study is clearly needed.

Finally, this overall evaluation also provides a framework for answering the implied question of the sub-title. The 19th century developments were critical in first formulating the combustion problem and developing the initial concepts, mostly in phenomenological terms, but also initiating mechanistic descriptions. The first half of the present century mostly saw transformation of those concepts into mathematical formulations with a degree of initial testing. The second half has been more focused on developments of data bases and applications with, also, more detailed and sophisticated (largely computerized) analytical treatments. In spite of all these results, however, there is still no clear answer to the question: "What is a flame?" It would appear that to answer this question, new concepts or insights are required. It would seem reasonable to expect, in response to the pass-out question, that this will be the contribution of the 21st century.

Table 1: Flame Phenomena and Characteristics

1. Minimum ignition energy requirement: potential for bifurcation characteristics
2. Reaction zone: region of exothermic reaction
3. Bounded reaction zone: flame front division between non-reacting and reacting region
4. Propagation: translation of flame front into unburned mixture
5. Flame speed: fundamental property of the mixture composition
6. Flame stabilization: obtained when flame speed is matched by mixture speed.
7. Combustion limits: flame propagation fails below and above lower and upper limits
8. Flame Temperatures: limited at adiabatic
9. Visible radiation characteristics of reaction zone

Table 2: A Classification of Flames

Category I: Fundamental Flame Systems

Primary Property: Flame speed is a fundamental *independent* property of the fuel mixture

Secondary Property: Flame stabilizes when the mixture and burning velocity are matched

Primary Characteristic: Non-recirculating flow, nor thermal assist

Defined by: Rankine-Hugoniot equations

Interpreted by: Mallard and le Chatelier (MLC) model

Category II: Thermally-Assisted Flame Systems (see also Table 3)

Primary Property: Flame stabilization is possible in high speed flows

Secondary Property: Flame speed is a variable, *dependent* property of the combustion system

Primary Characteristic: Thermally assisted (backmix flow and other)

Defined and Interpreted by: 3-D conservation and kinetics equations

Category III: Miscellaneous

Surface flames (flame spread); fires, intermittent/batch combustion, ignition . . .

Table 3: Thermally-Assisted Flame Systems

Class I: External Heat Recovery by downstream heat exchanger(s):

Exs: Blast furnace; Open Hearth; glass tank; brick kiln; boiler . . .

Class II: Internal Heat Recovery

- II.1 *Flow driven*: standard burners; non-swirling/swirling jets; FB's; etc;
applications in standard [3-D] industrial furnaces and engines
- II.2 *"Filtration" Porous Body Systems*
 - (1) Reacting porous body (solid fuel bed, sinter bed, blast furnace . . .)
 - (2) Reactive gas mixture in Porous Body