

NASA Contractor Report 191051

1N-29
145785
p-93

Time-Dependent Computational Studies of Premixed Flames in Microgravity

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January 1993

Prepared for
Lewis Research Center
Under Contract C-32001-M

NASA
National Aeronautics and
Space Administration

(NASA-CR-191051) TIME-DEPENDENT
COMPUTATIONAL STUDIES OF PREMIXED
FLAMES IN MICROGRAVITY Final Report
(Naval Research Lab.) 93 p

N93-18387

Unclass

G3/29 0145785



**TIME-DEPENDENT COMPUTATIONAL STUDIES OF
PREMIXED FLAMES IN MICROGRAVITY**

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Final Report

to

**Space Experiments Division
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ABSTRACT

This report describes the research performed at the Center for Reactive Flow and Dynamical Systems in the Laboratory for Computational Physics and Fluid Dynamics, at the Naval Research Laboratory, in support of the NASA Microgravity Science and Applications Program. The primary focus of this research is on investigating fundamental questions concerning the propagation and extinction of premixed flames in earth gravity and in microgravity environments.

Our approach is to use detailed time-dependent, multispecies, numerical models as tools to simulate flames in different gravity environments. The models include a detailed chemical kinetics mechanism consisting of elementary reactions among the eight reactive species involved in hydrogen combustion, coupled to algorithms for convection, thermal conduction, viscosity, molecular and thermal diffusion, and external forces. The external force, gravity, can be put in any direction relative to flame propagation and can have a range of values. Recently more advanced wall boundary conditions such as isothermal and no-slip have been added to the model. This enables the simulation of flames propagating in more practical systems than before.

We have used the numerical simulations to investigate the effects of heat losses and buoyancy forces on the structure and stability of flames, to help resolve fundamental questions on the existence of flammability limits when there are no external losses or buoyancy forces in the system, to understand the interaction between the various processes leading to flame instabilities and extinguishment, and to study the dynamics of cell formation and splitting.

Our studies have been able to bring out the differences between upward- and downward-propagating flames and predict the zero-gravity behavior of these flames. The simulations have also highlighted the dominant role of wall heat losses in the case of downward-propagating flames. Details of the flame-extinguishment process during downward propagation at the flammability limit have been elucidated. The simulations have been able to qualitatively predict the formation of multiple cells and the cessation of cell-splitting. Our studies have also shown that some flames in a microgravity environment can be extinguished due to a chemical instability and without any external losses. However, further simulations are needed to more completely understand upward-propagating and zero-gravity flames as well as to understand the potential effect of radiative heat losses.

INTRODUCTION

In this report we describe the research performed at the Naval Research Laboratory in support of the NASA Microgravity Science and Applications Program over the period February 1989—September 1991. This work has been performed at the Center for Reactive Flow and Dynamical Systems in the Naval Research Laboratory by Drs. Gopal Patnaik, K. Kailasanath and Elaine Oran. The emphasis of our research has been on investigating fundamental combustion questions concerning the propagation and extinction of gas-phase flames in microgravity and earth-gravity environments. Our approach to resolving these fundamental questions has been to use detailed time-dependent, multispecies numerical models to perform carefully designed computational experiments. The basic questions we have addressed, a general description of the numerical approach, and a summary of the results are described in this introduction. More detailed discussions are presented in the next two sections and in the appendices to this report.

It is well known from flammability studies in Earth gravity that a flame propagating upward in a tube propagates in a wider range of mixture stoichiometries and dilutions than a flame propagating downward. This means that the apparent flammability limits of upward-propagating flames are broader than those of downward-propagating flames. Various explanations for this phenomena are based on factors such as buoyancy forces, preferential diffusion, flame chemistry, conductive and radiative heat losses, the aerodynamics of burnt gases, and flame stretch [1-8]. Furthermore, instabilities are often observed in the propagation of flames, especially near the flammability limits. One certainty is that gravitational acceleration (buoyancy) is very important and may also be interacting with and influencing other physical processes.

A systematic study which isolates the various processes that could lead to flame instabilities and extinction is needed to gain a better understanding of flammability limits in general and more specifically, the role of gravity on flame propagation and extinction. Numerical simulations, in which the various physical and chemical processes can be independently controlled, can significantly advance our understanding of flame instabilities in a microgravity environment and flammability limits on earth and in a microgravity environment. In the past three years, we have addressed a number of basic issues aimed at resolving some of the important aspects of flame structure and propagation. There are still many questions left unanswered and, as such, this final

report is also a progress report on a continuing investigation.

The basic issues we have addressed are: the effects of heat losses and buoyancy forces on the structure and stability of flames, the existence of flammability limits in the absence of external losses or buoyancy forces in the system, and the extinguishment process at the downward-propagation limit.

Our current numerical simulations of hydrogen-air flames in a two-dimensional channel have been able to bring out some of the differences observed between upward- and downward-propagating flames and predict the zero-gravity behavior of these flames. The simulations have also highlighted the dominant role of wall heat losses in the case of downward-propagating flames. Details of the flame-extinguishment process during downward propagation at the flammability limit have been elucidated. The simulations have been able to qualitatively predict the formation of multiple cells and the cessation of cell-splitting. Our studies have also shown that some flames in a microgravity environment can be extinguished without external losses. However, further simulations are needed to more completely understand upward-propagating and zero-gravity flames as well as to understand the potential effect of radiative heat losses.

In the research described here, we have used both one-dimensional and two-dimensional numerical simulations to systematically isolate and evaluate the importance of various processes that might be controlling the dynamics of flames, particularly near the flammability limits, and to evaluate the relative importance of these processes in normal gravity and microgravity. Both the numerical models are time-dependent and solve the multispecies coupled partial differential reactive-flow equations. These models include a detailed chemical kinetics mechanism coupled to algorithms for convection, thermal conduction, viscosity, molecular diffusion, thermal diffusion, and external forces. The external force, gravity, can be in any direction relative to flame propagation and can have a range of values. Energy sources and sinks may also be prescribed as a function of time. The models are described in greater detail in the next section.

COMPUTATIONAL TOOLS

Two computational models developed at NRL, FLIC2D and FLAME1D, have been used in the work reported here. FLIC2D has been used in the past to study the tendency of hydrogen-air mixtures to exhibit cellular structure [9] and to model diffusion flames [10]. Here, we are using it to study the effects of gravity and heat losses on structure and stability of multidimensional flames. Many submodels in FLIC2D are based on FLAME1D, a one-dimensional, time-dependent Lagrangian model which has been tested extensively and applied to various studies of flame phenomena including calculations of burning velocities [11,12], minimum ignition energies and quenching distances [12,13], effects of curvature and dilution [14], and flammability limits [15]. Here, this one-dimensional model is used to further explore the rich-flammability limit of hydrogen-air mixtures. Both models are briefly described below.

FLAME1D

This is a time-dependent model [12] which solves the compressible, conservation equations for mass, momentum and energy [16,17] in one spatial dimension. Since it was developed specifically to study the initiation, propagation, and quenching of laminar flames, it consists of algorithms for modelling convection, detailed chemical kinetics and energy release, thermal conduction, molecular and thermal diffusion of the individual species, and various energy deposition mechanisms. The model has a modular form and the algorithms representing the various chemical and physical processes are combined using an asymptotic timestep-split approach in which the individual processes are integrated separately and then coupled together [17,18]. The model also permits a variety of initial and boundary conditions.

The convective transport terms in the equations are solved by the algorithm ADINC, a Lagrangian convection algorithm which solves implicitly for the pressures [19]. Since ADINC communicates compression and expansion across the system implicitly, it overcomes the Courant time-step limit. Since it is Lagrangian, it can maintain steep gradients computationally without numerical diffusion for a long period of time. This is important in flame calculations where the diffusive transport of material and energy can govern the system evolution and therefore must be calculated accurately. An adaptive regridding algorithm has been developed to add and delete computational cells. For the flame calculation, cells are added in the region ahead of the flame so that the

front always propagates into a finely zoned region. Cells are removed behind the flame where the gradients are very shallow.

The chemical interactions are described by a set of nonlinear coupled ordinary differential equations. This set of equations may be stiff when there are large differences in the time constants associated with different chemical reactions, and are invariably stiff for combustion problems. These equations are solved using VSAIM, a fully vectorized version of the selected asymptotic integration method CHEMEQ [20,21]. In VSAIM, the stiff equations are identified and solved using a very stable asymptotic method while the remaining equations are solved using a standard classical method.

The diffusive transport processes considered in this model are molecular diffusion, thermal conduction and thermal diffusion. These processes are crucial to the description of flame phenomena since they are the mechanisms by which heat and reactive species are transported ahead to the unburned gas. An iterative algorithm, DFLUX, is used to obtain the diffusion velocities without the cost of performing matrix inversions [17,22]. This method has been vectorized and is substantially faster than matrix inversions when four or more species are involved.

Problems can be set up in either planar, cylindrical, or spherical coordinates as well as a variable power series coordinates which can model one-dimensional nozzle-like geometries. The model can also be configured with either an open or closed boundary at one end. The open boundary simulates an unconfined system.

FLIC2D

FLIC2D is a time-dependent, Eulerian, implicit, compressible, two-dimensional flame model. In FLAME1D, a Lagrangian method was chosen for convective transport because eliminating the advection term in effect means eliminating numerical diffusion from the calculation. Extending this Lagrangian approach to multidimensions would be extremely difficult and expensive, requiring many years of algorithm development and testing. Therefore, we felt that we would have more success in multidimensions with an Eulerian method. However, most Eulerian methods are either more numerically diffusive than what is acceptable in a flame model, or they are explicit and hence extremely inefficient at the very low velocities associated with laminar flames. To circumvent these numerical problems, we developed BIC-FCT, the Barely Implicit Correction to Flux-Corrected Transport [23,24]. BIC-FCT combines an explicit high-order, nonlinear FCT method [25,26] with an implicit correction process. This combination maintains

high-order accuracy and yet removes the timestep limit imposed by the speed of sound. By using FCT for the explicit step, BIC-FCT is accurate enough to compute sharp gradients without overshoots and undershoots. Thus spurious numerical oscillations that would lead to unphysical chemical reactions do not occur. The development of this new algorithm has made it possible to model multidimensional flames in detail. However, because there is always some residual numerical diffusion, even in high-order Eulerian algorithms, we need to monitor calculations to ensure that numerical diffusion never becomes larger than the physical diffusion processes we need to resolve.

In generalizing the flame model to two-dimensions, simplifications were made in the diffusive transport calculations in order to reduce the cost of computations. In FLIC2D, thermal conductivity of the individual species is modeled by a polynomial fit in temperature to existing experimental data. Individual conductivities are then averaged using a mixture rule [12,27] to get the thermal conductivity coefficient of the gas mixture. A similar process is used to obtain the mixture viscosity from individual viscosities. Heat and momentum diffusion are then calculated explicitly using these coefficients.

Mass diffusion also plays a major role in determining the properties of laminar flames. Binary mass-diffusion coefficients are represented by an exponential fit to experimental data, and the individual species-diffusion coefficients are obtained by applying mixture rules [12]. The individual species-diffusion velocities are determined explicitly by applying Fick's law followed by a correction procedure to ensure zero net flux [27]. This procedure is equivalent to using the iterative algorithm DFLUX [22] (used in FLAME1D) to second order.

The chemical reaction-rate equations are modelled and solved as before using a vectorized version of CHEMEQ, an integrator for stiff ordinary differential equations [21]. Because of the complexity of the reaction scheme and the large number of computational cells in a two-dimensional calculation, the solution of the chemical rate equations takes a large fraction of the total computational time. A special version of CHEMEQ called TBA was developed to exploit the special hardware features of the CRAY X-MP and Y-MP.

As in the one-dimensional code, all of the chemical and physical processes are solved sequentially and then coupled asymptotically by timestep splitting [17,18]. This modular approach greatly simplifies the model and makes it easier to test and change

the model. Individual modules were tested against known analytic and other previously verified numerical solutions. One-dimensional predictions of the complete model were compared to those from the Lagrangian model FLAME1D which has been benchmarked extensively against theory and experiment.

In summary, all of the chemical and diffusive-transport algorithms are essentially the same in FLIC2D as in FLAME1D. Thus given the input data concerning the species, their transport coefficients, and the reactions among the species, either the one- or two-dimensional model can be run. The major differences between the two models are in the costs of running them and the algorithm chosen for convective transport. In general, we use the results of one-dimensional calculations as initial conditions for the two-dimensional calculations in order to reduce the cost of computations. A report on the code discusses more details than what is presented here [28].

SUMMARY OF RESEARCH

In this section we briefly describe the research performed for the Microgravity Science and Applications Program. The research can be broadly divided into five categories: (a) dynamics of cellular flames, (b) effects of gravity on flame instabilities and structure, (c) role of heat losses and viscosity on flame structure (d) extinguishment process at the lean-flammability limit and (e) instabilities near the rich-flammability limit. Each of these categories are discussed below.

Dynamics of Cellular Flames

Our initial simulations [29] of cellular flames were in qualitative agreement with experimental observations on the stability of flames in lean and rich hydrogen-oxygen-nitrogen mixtures [30]. However, the vigorous cell-splitting observed in the experiments in the NASA drop tower [31] were not observed in these simulations. This difference could have been due to the small system size of 2 cm used in the simulations. Therefore, we repeated some of the earlier simulations but with a larger system size of 5.1 cm. This particular size was chosen because it corresponds to the diameter of the standard flammability limit tube and one of the eventual aims of these simulations is to calculate flammability limits.

The early time history of the flame in a 1.5:1:10 mixture in a 5.1 cm channel is the same as that observed earlier for a flame in a 2 cm channel. However, for longer times, the cells present in the center of the wider channel grow larger and split into smaller cells. Then, these smaller cells grow larger and continue the splitting process.

After this demonstration of the ability of the code to simulate multiple cell-splits, we investigated the "cell-split limit" postulated from experimental observations [31]. The cell-split limit refers to Ronney's experimental observation that for mixtures leaner than 5.5 % hydrogen (in air), the cells that form do not split. He has also attributed this limit to radiative or other heat losses from the system. If heat losses are essential for this limit, then these calculations should not observe such a limit.

We simulated the structure and dynamics of flames in a series of mixtures with 15% or less hydrogen in air. We observe brisk formation of cells and their subsequent splitting in lean mixtures with more than 11% hydrogen. In leaner mixtures with 9.5-11% hydrogen, the formation and splitting of cells is less vigorous. For mixtures with 9% or less hydrogen, a single cell is formed which does not split at all. For these

mixtures, even calculations for very long times (0.4 seconds) in larger systems (10.2 cm channel width) did not show any indications of cell-splitting.

In summary, a complete cessation of cell splitting is observed in mixtures between 9 and 9.5 % hydrogen in air, indicating that a cell-split limit also exists in our numerical simulations. However, the mixture at which this limit occurs is higher than the 5.5 % mixture found in the experimental observations. The reasons for this discrepancy are not yet known and may be due to many differences that exist between the experimental set-up and the numerical simulations. For example, the experiments were carried out in a spherical vessel, in which the cellular phenomena is three-dimensional. The numerical calculations simulated cellular flames in a two-dimensional channel. Also, the effect of the halon additive on the cellular structures in the experiments is not fully understood.

Heat losses have been postulated as a mechanism that can cause a cell-split limit. However, the simulations in this study do not include any loss mechanism; yet such a limit is still observed. This suggests that external loss mechanisms are not required and that the cell-split limit is actually an intrinsic feature of the lean hydrogen-air mixture itself. However, the actual mixture at which cell splitting ceases may be affected by losses.

Details of this work have been presented at the 1989 Eastern States Combustion Institute Meeting [32] and the 1990 AIAA Aerospace Sciences Meeting and is published as AIAA Paper No. AIAA-90-0041 [33]. This paper is included in this report as Appendix A.

Effects of Gravity on Flame Instabilities and Structure

We have investigated the effects of gravity on flames in both a narrow 2 cm channel as well as a wider 5.1 cm channel. Only the phenomena observed in the wider channel is discussed here. Our earlier calculations can be found in Ref. [34,35]. In this systematic series of simulations, the effects of gravity on a sequence of hydrogen-air mixtures from 15 % hydrogen to 7.5 % hydrogen was studied.

The calculations show that the effects of gravity become more important as the lean flammability limit is approached. In mixtures with 12% or more of hydrogen in air, gravity plays only a secondary role in determining the multidimensional structure of the flame, with the stability and structure of the flame controlled primarily by the thermo-diffusive instability mechanism. However, in leaner hydrogen-air mixtures

gravity becomes more important. Upward-propagating flames are highly curved and evolve into a bubble rising upwards in the tube. Downward-propagating flames are flat or even oscillate between structures with concave and convex curvatures. The zero-gravity flame shows only cellular structures. Cellular structures which are present in zero gravity can be suppressed by the effect of buoyancy for mixtures leaner than 11% hydrogen. These observations have been explained on the basis of an interaction between the processes leading to buoyancy-induced Rayleigh-Taylor instability and the thermo-diffusive instability.

The thermo-diffusive instability was found to be present in all the mixtures studied and at both orientations of gravity and is responsible for the small, cell-like structures which are observed in the early stages of all these simulations. However, the presence of the buoyancy-induced instability alters the large scale structure of the flame. The effect of buoyancy on the cellular structures formed by the thermo-diffusive instability is small in mixtures which have greater than 11% hydrogen. In a 11% mixture, a downward-propagating flame is flat with small cells that repeatedly split. In leaner mixtures, the effect of gravity is more dramatic. In these mixtures, the upward-propagating flame has the characteristic bubble shape observed experimentally and the downward propagating flame has oscillations characteristic of the Rayleigh-Taylor instability and does not have any cellular structures. These results agree with the theory [36] that indicates that the influence of gravity is greater for lower flame speeds and that such oscillatory behavior is possible. These results indicate that the instability mechanisms can interact in a quite complex manner, and even though one mechanism can mask the other, in certain regimes, both can be equally important.

Calculations for still leaner mixtures are needed to address the actual extinction behavior of upward and downward-propagating flames. Loss mechanisms such as heat and radical losses to the walls as well as radiation might also play a role in determining the detailed extinction behavior of these flames. These effects are being systematically considered in further calculations some of which are discussed next.

These results have been presented and published in the Proceedings of the Twenty-third Symposium (International) on Combustion [37] and is enclosed as Appendix B.

Role of Heat Losses and Viscosity on Flame Structure

In many practical applications as well as in the determination of flammability limits, flames are confined. Heat and radical losses will occur to the confining walls and the flow will also have to come to rest at the surface of the walls. All these processes may significantly affect the structure and dynamics of flames.

As a first step, detailed two-dimensional numerical simulations have been carried out to investigate the effects of heat and momentum losses (due to viscosity) to the walls on the structure and dynamics of flames in a 10% hydrogen-air mixture. The simulations of flames in zero-gravity show that momentum losses to adiabatic walls result in flames with very complex shapes in which the portion of the flame near the walls moves significantly faster than the central portion of the flame. This phenomena occurs because of the slower flow velocities near the walls. However, viscous effects play only a secondary role when the walls are assumed to be isothermal. In this case, heat losses to the walls play a dominant role resulting in the formation of additional cellular structures near but slightly displaced from the walls. This displacement from the walls is a key factor in the secondary role played by viscous effects because it moves the flame to the outer edges of the viscous boundary layer.

We also investigated how gravity modifies the structure and dynamics of flames in an isothermal, two-dimensional channel. Heat losses are found to significantly modify upward- and downward-propagating flames. In the upward-propagating case, instead of a single bubble rising in the tube, a two-fingered flame is observed. This flame has some features similar to experimentally observed flames in lean hydrogen-air mixtures. The downward-propagating flame is also quite different when the effect of heat losses are considered. In this case, a flame is "visible" only in the central portion of the channel. Again, this flame is qualitatively similar to some near-limit flames in experiments. Further calculations described below provide valuable insights into the dynamical behavior observed experimentally in flames near extinguishment. Details of this work have been presented and published as AIAA paper No. 91-0784 [38] and is enclosed as Appendix C.

Extinguishment Process at the Lean-Flammability Limit

The experimentally observed flammability limit is between 9 and 10% for downward flame propagation in lean Hydrogen-air mixtures. Therefore further simulations

including the effect of wall heat losses were carried out for flames in 10%, 9.75%, 9.5% and 9% hydrogen-air mixtures. The computational results show that a downward propagating flame in an isothermal channel has a flammability limit of around 9.75%. The details of the extinguishment process in this mixture has been analysed and found to be in excellent agreement with experimental observations.

In experiments, the flame is first observed to halt its downward propagation and then actually move back up into the burnt products. From the simulations we can understand the reason for this observed behavior. The simulations indicate that the flame is quenched at the walls and tongues of colder gases, comprised mainly of burnt products flow down the sides. At the same time, there is an upward motion of the gases at the center of the channel, causing the flame to rise up into the burnt products. Further simulations also indicate that the dilution of the unburnt mixture with the products of combustion is an essential step in the extinguishment

Our earlier simulations have shown that a flame can propagate downward in a 9% mixture in a channel with adiabatic walls and cellular flames occur in this mixture in a zero-gravity environment. These observations taken in conjunction with our current simulations indicate that both heat losses and gravity are simultaneously required to cause the observed limit. A general conclusion from this work is that detailed numerical simulations that include wall heat losses can adequately simulate the dynamics of the extinguishment process in downward-propagating flames. Further details of this work are given in Appendix D where a paper presented at the 30th Aerospace Sciences Meeting is enclosed [39].

Instabilities Near the Rich-Flammability Limit

As discussed in the previous section, we have a detailed description of the behavior of flames at the downward-propagation limit. At the upward-propagation limit, the flames are highly three-dimensional and therefore must await the development of a three-dimensional model for a complete description. Meanwhile, we have also been studying the issue of fundamental flammability limits in a zero-gravity environment.

Even in the absence of buoyancy forces, there are many competing factors such as preferential diffusion, flame chemistry, conductive and radiative heat losses, the aerodynamics of burnt gases, and flame stretch that can cause flammability limits. Numerical simulations provide an ideal way to systematically isolate and evaluate the role of var-

ious factors. In an earlier study (supported by the MSAP) on the effects of curvature and dilution on flame propagation [14], it was shown that a flame can be extinguished with less dilution in one geometry than in another, even in the absence of external heat losses and buoyancy effects. Specifically, we showed that a planar flame can propagate steadily in a dilute mixture which does not support a spherically expanding flame. Because this behavior is related to the effects of flame stretch and Lewis number, it raises the fundamental question of whether there is an extinguishment limit in the absence of stretch effects. This question can be addressed by carefully studying the behavior of planar flames in the absence of external heat losses and gravitational effects. Our preliminary results [15] showed that there is such a limit. However, a more recent numerical study of methane flames suggests that there may not be any limit for methane-air mixtures in the absence of heat losses [40].

Therefore, we conducted a more systematic study of flames in rich hydrogen-air mixtures. Because these flames are nearly one-dimensional, we conducted these investigations with the FLAME1D code. Results from the numerical simulations indicate that a steady burning velocity is not obtained for very rich hydrogen-air mixtures. As the amount of hydrogen is increased, at first a damped oscillation is observed in the flame and burning velocities, and then with further increase in the amount of hydrogen, an undamped oscillation with a complex set of frequencies is observed. Simulations with a simplified one-step irreversible chemical reaction do not show these oscillations, suggesting that chemical kinetics plays a strong role in inducing these oscillations. Further analysis shows that the oscillations are due to a competition for H atoms between chain branching and chain-terminating reactions. However, the limiting mixture predicted by these simulations is beyond the experimentally observed limit. These differences may be because of the neglect of phenomena such as stretch and radiative heat losses.

Simulations of spherically expanding flames suggest that stretch effects (due to curvature) will cause the oscillations to occur in less rich mixtures than that observed for planar flames. Further calculations including multidimensional effects and radiative and conductive heat losses are needed to quantitatively determine the flammability limits in zero-gravity as well as to more fully understand the significance of the instability induced by chemical kinetics.

This work was presented at the 13th ICDERS meeting [41] and is described in greater detail in Appendix E.

A list of recent papers and presentations related to all the work described above are given below.

Journal Articles and Meeting Proceedings

1. Detailed Numerical Simulations of Cellular Flames, G. Patnaik, K. Kailasanath, K.J. Laskey and E.S. Oran, *Proceedings of the 22nd Symposium (International) on Combustion*, pp. 1517-1526, The Combustion Institute, Pittsburgh, PA., 1989.
2. Effect of Gravity on Flame Instabilities in Premixed Gases, G. Patnaik, K. Kailasanath, and E.S. Oran, AIAA Paper No. 89-0502, AIAA 27th Aerospace Sciences Meeting, Reno, NV, AIAA, Washington, DC, January 1989.
3. Flame Instabilities in Premixed Gases, K. Kailasanath, *Proceedings of the International Microgravity Combustion Workshop*, Cleveland, OH, Jan. 25-26, 1989.
4. Numerical Simulations of Flames and Detonations, K. Kailasanath, E.S. Oran and J.P. Boris, p. 82, in *Numerical Combustion*, A. Dervieux and B. Larrouturou, eds., *Lecture Notes in Physics*, 351, Springer-Verlag, 1989.
5. Cellular Structure of Lean Hydrogen Flames in Microgravity, G. Patnaik and K. Kailasanath, AIAA Paper No. 90-0041, AIAA 28th Aerospace Sciences Meeting, Reno, NV, AIAA, Washington, DC, January 1990.
6. Effect of Gravity on the Stability and Structure of Lean Hydrogen-Air Flames, G. Patnaik and K. Kailasanath, *Proceedings of the 23rd Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, PA., pp. 1641-1647, 1990.
7. Effect of Heat Losses on the Cellular Structure of Lean Hydrogen-Air Flames, G. Patnaik and K. Kailasanath, AIAA Paper No. 91-0784, 29th AIAA Aerospace Sciences Meeting, Reno, Nevada, AIAA, Washington, DC, January 1991.
8. Laminar Flames in Premixed Gases, K. Kailasanath, in *Numerical Approaches to Combustion Modelling*, *Prog. Astro. Aero.*, Vol. 135, pp.225-256, AIAA, 1991.
9. Multidimensional Flame Instabilities, K. Kailasanath and Patnaik, G., 1991 NRL Review, Naval Research Laboratory Publication 181-4830, Washington, D.C., 1991.
10. Effect of Gravity on Flame Instabilities in Premixed Gases, G. Patnaik, K. Kailasanath, and E.S. Oran, *AIAA Journal*, Vol. 29, pp. 2141-2148, 1991.

11. Lean Flammability Limit of Downward Propagating Hydrogen-Air Flames, G. Patnaik and K. Kailasanath, AIAA Paper No. 92-0336, 30th AIAA Aerospace Sciences Meeting, Reno, Nevada, AIAA, Washington, DC, January 1992.

Reports

1. FLIC- A Detailed Two-Dimensional Flame Model, G. Patnaik, K.J. Laskey, K. Kailasanath, E.S. Oran and T.A. Brun, NRL Memorandum Report 6555, September 1989.
2. The Stability and Structure of Lean Hydrogen-Air Flames: Effects of Gravity, G. Patanik and K. Kailasanath, NRL Memorandum Report 6649, June 1990.

Meeting Presentations

1. Effect of Gravity on Flame Instabilities in Premixed Gases, G. Patnaik, K. Kailasanath, and E.S. Oran, AIAA Paper No. 89-0502, AIAA 27th Aerospace Sciences Meeting, Reno, NV, Jan. 9-12, 1989.
2. Flame Instabilities in Premixed Gases, K. Kailasanath, International Microgravity Combustion Workshop, Cleveland, OH, Jan. 25-26, 1989.
3. Numerical Simulations of Flames and Detonations, K. Kailasanath, invited presentation, Third International Conference on Numerical Combustion, Antibes, France, May 23-26, 1989.
4. The Role of Intermediate Species in the Cellular Structure of Flames, G. Patnaik and K. Kailasanath, 12th International Colloquium on the Dynamics of Explosions and Reactive Systems, Ann Arbor, MI, July 23-28, 1989.
5. Cellular Structure of Very Lean Hydrogen Flames, G. Patnaik and K. Kailasanath, Paper No. 29, Fall Technical Meeting of the Eastern States Section of the Combustion Institute, Albany, NY, Oct 30-Nov 1, 1989.
6. Cellular Structure of Lean Hydrogen Flames in Microgravity, G. Patnaik and K. Kailasanath, AIAA Paper No. 90-0041, AIAA 28th Aerospace Sciences Meeting, Reno, Nevada, January 8-11, 1990.
7. Effect of Gravity on the Stability and Structure of Lean Hydrogen-Air Flames, G. Patnaik and K. Kailasanath, presented at the 23rd Symposium (International) on Combustion, Orleans, France, July 22-27, 1990.

8. On Flames Near the Rich-Flammability Limit of Hydrogen-Air Mixtures, K. Kailasanath, K. Ganguly and G. Patnaik, Paper No. 32, Fall Technical Meeting of the Eastern States Section of the Combustion Institute, Orlando, FL, Dec 3-5, 1990.
9. Use of Single-step Chemistry in Multi-Dimensional Flame Modeling, G. Patnaik and K. Kailasanath, Paper No. 41, Fall Technical Meeting of the Eastern States Section of the Combustion Institute, Orlando, FL, Dec 3-5, 1990.
10. Effect of Heat Losses on the Cellular Structure of Lean Hydrogen-Air Flames, G. Patnaik and K. Kailasanath, AIAA Paper No. 91-0784, 29th AIAA Aerospace Sciences Meeting, Reno, Nevada, Jan. 7-10, 1991.
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FUTURE DIRECTIONS

The calculations discussed above have led to a better understanding of the structure and propagation of flames. We now understand the reasons for some of the observed differences in the structure of upward and downward propagating flames. We also have a detailed description of the extinguishment process in downward-propagating flames and rich hydrogen-air flames. However, these simulations have also brought up a number of unresolved issues which need to be examined. Four major issues which need to be addressed are: (1) What is the role of radiation in near-limit flame phenomena, especially for hydrocarbon flames? (2) Will the extinguishment process and the relative importance of various instability mechanisms be the same for hydrocarbon flames as for hydrogen flames? (3) Are burner-stabilized flames significantly different from freely propagating flames? and (4) How does three-dimensionality modify the observations from the two-dimensional simulations?

The issues raised above are ideal for being studied cost-effectively using numerical simulations. However, they have not been studied systematically due to the lack of a three-dimensional, time-dependent flame model and adequate sub-models for hydrocarbon flames (especially radiation models). Detailed reaction mechanisms are available for some hydrocarbons but the cost of including such mechanisms in time-dependent, multidimensional simulations is very high. The solution of the chemical rate equations takes a significant portion (60 - 70 %) of the computer time because of the large number of species and the large number of rate equations involved in a mechanism consisting of elementary reactions. One approach to reducing the cost of multidimensional flame simulations is to use a greatly simplified reaction mechanism. However, a satisfactory simplified reaction mechanism does not exist currently, even for methane. Although, we have reduced a 108 reaction-rate mechanism for methane to about 50 rates [42], this is still too expensive to use in a multidimensional flame calculation. Currently there are many ongoing efforts to find ways of easing the burden of integrating full chemical reaction-rate mechanisms [43-46]. Using such simplified approaches in FLIC2D is straightforward and will enable us to simulate multidimensional flames in hydrocarbon mixtures.

Another approach to reducing the cost of chemistry calculations is to develop faster chemical integration algorithms. We are continually finding ways to make the algorithms we use faster. In addition, there are some new approaches to sorting stiff

equations [47] which are promising. However, it is unlikely that reducing the computer time required by using faster algorithms alone will be sufficient to allow us to perform detailed calculations of complex hydrocarbon flames cost-effectively.

The major advances being made in parallel computing will probably be the crucial factor enabling us to simulate the hydrocarbon flames to the same level of detail as we do with the hydrogen flames. Currently, we are working on an increasingly more parallel-processing approach for doing our multidimensional flame simulations. Meanwhile, additional efforts are also being made in developing simplified chemistry and radiation models.

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Appendix A

Dynamics of Cellular Flames in Microgravity



AIAA-90-0041

**CELLULAR STRUCTURE
OF LEAN HYDROGEN FLAMES
IN MICROGRAVITY**

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28th Aerospace Sciences Meeting
January 8-11, 1990/Reno, Nevada

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Abstract

Detailed, time-dependent, two-dimensional numerical simulations of premixed laminar flames have been used to study the initiation and subsequent development of cellular structures in lean hydrogen-air flames. The model includes detailed hydrogen-oxygen combustion with 24 elementary reactions of eight reactive species and a nitrogen diluent, molecular diffusion of all species, thermal conduction, viscosity, and convection. This model has been used to study the non-linear evolution of cellular flame structure and shows that cell splitting, as observed in experiments, can be predicted numerically for sufficiently reactive mixtures. The structures that evolved also resembled the cellular structures observed in experiments. The present study shows that the "cell-split limit" postulated from experimental observations is an intrinsic property of the mixture and that external factors such as heat losses are not necessary to cause this limit.

1. Introduction

The structure of flames, especially near flammability limits, is usually multidimensional. To investigate such multidimensional effects, we have developed a time-dependent, two-dimensional model that can simulate either premixed or diffusion flames. In this paper, we present results of calculations using this model to study the structure of premixed laminar flames in lean hydrogen-air mixtures. These results are discussed in terms of the formation of cellular structure, and the existence of a "cell-split" limit as observed experimentally in a microgravity environment¹. In this study we restrict our attention to lean hydrogen flames in zero gravity, thus eliminating the effects of gravity.

Cellular structures in flames have been observed in the microgravity experiments in the NASA drop tower² and Lear-jets³. Ronney has recently reported some detailed experimental observations of cellular structures of lean hydrogen flames in microgravity¹. He describes the formation of numerous cells which split vigorously in hydrogen-air mixtures with greater than 5.5% hydrogen. For mixtures close to 5.5%, fewer cells are formed which then subsequently split. For leaner mixtures below 5.5% cells form, but do not undergo division. He has observed the suppression of cell division in a 7% mixture to which a halon flame retardant has been added.

He proposes a "cell-split limit" below which cells still form, but do not undergo any cell splitting. This limit has been attributed to radiative or other heat losses⁴.

Multidimensional structures are often formed as the result of the instability of a one-dimensional flame to perturbations. Linear stability analyses can provide information on the roles of various processes at the onset of instability. However, the prediction of the growth of this instability to the final form is beyond the scope of these analyses. Numerical calculations can be used to help understand both the onset of the instability and the evolutionary process that produces the multidimensional structure. The numerical simulations of flames presented here include as input a multireaction mechanism for hydrogen combustion, molecular diffusion between the reactants, intermediates, and products, thermal conduction, and convection. Such a detailed model allows us to investigate the multidimensional structure of flames and to evaluate the importance of various contributing physical processes. We have already used simulations to show that, in the absence of gravity, cellular structure is caused by the thermo-diffusive mechanism⁵.

The thermo-diffusive instability investigated by Barenblatt and Zeldovich^{6,7} is the major mechanism that gives rise to cellular structure that can occur in premixed flames in zero gravity. This instability mechanism involves a competition between mass diffusion of the deficient reactant and diffusion of heat in the mixture. For lean hydrogen-air mixtures, hydrogen is the limiting reactant and its mass diffusivity significantly exceeds the thermal diffusivity of the mixture, and the flame is predicted to be unstable. Because our numerical model of the premixed hydrogen flame includes all the physical mechanisms that give rise to this instability, it provides an ideal test-bed to study the evolution of cellular structures.

The goal of this paper is to investigate the cellular structure arising from flame instability and, in particular, to determine if the cell-split limit is an intrinsic feature of lean hydrogen mixtures which exists even in the absence of heat losses. Below we describe the numerical model, show its results in simulating very lean hydrogen-air flames with different stoichiometries, determine a cell-split limit, and show that it is an inherent property of the mixture.

II. Multidimensional Flame Model

A detailed model of a flame must contain accurate representations of the convective, diffusive, and chemical processes. The individual importance of these processes varies from rich to lean flames, and is especially notable near the flammability limits⁸ where the exact behavior of these flames depends on a delicate balance among the processes. The reactive-flow conservation equations are solved for density, ρ , momentum, $\rho\vec{V}$, total energy E , and the number densities of individual species, n_k , $k = 1, \dots, n_p$ according to:

$$\begin{aligned} \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{V}) &= 0, \\ \frac{\partial \rho \vec{V}}{\partial t} + \nabla \cdot (\rho \vec{V} \vec{V}) &= -\nabla P + \vec{F} - \nabla \times \mu \nabla \times \vec{V} + \nabla \cdot \left(\frac{4}{3} \mu \nabla \cdot \vec{V} \right), \\ \frac{\partial E}{\partial t} + \nabla \cdot (E \vec{V}) &= -\nabla \cdot (P \vec{V}) + \nabla \cdot (\kappa \nabla T) - \\ &\quad \sum_{k=1}^{n_p} \nabla \cdot (n_k h_k \vec{V}_k) + \sum_{r=1}^{n_r} Q_r, \\ \frac{\partial n_k}{\partial t} + \nabla \cdot (n_k \vec{V}) &= -\nabla \cdot (n_k \vec{V}_k) + w_k. \end{aligned}$$

Here \vec{V} is the fluid velocity, P is the pressure, μ is the coefficient of viscosity, \vec{F} is the body force due to gravity, κ is the thermal conductivity of the mixture of gases, h_k is the enthalpy of species k , \vec{V}_k is the diffusion velocity of species k , Q_r is the heat released from reaction r , and w_k is production of species k by chemical reaction. These equations are solved assuming that the individual species are ideal gases obeying the thermal equation of state,

$$P_k = n_k k T,$$

and that the differential relation between internal energy u and pressure P is given by

$$\delta u = \frac{\delta P}{\gamma - 1},$$

where γ , the ratio of specific heats of the mixture, is a function of its temperature and composition. In the rest of this section, we briefly describe the algorithms and input data used to model and couple the various physical processes. Full details of the numerical scheme can be found in Reference 9.

The fluid convection algorithm must be able to maintain the sharp gradients present in flames. Numerically this means that the numerical diffusion in the calculation must be considerably less than any important physical diffusion effect. Many explicit algorithms now exist that treat sharp discontinuities in flow variables accurately, but these methods are extremely inefficient at the very low velocities associated

with laminar flames. The Barely Implicit Correction Flux-Corrected Transport (BIC-FCT) algorithm¹⁰ was developed specifically to solve low-speed flow problems with high accuracy. BIC-FCT combines an explicit high-order, nonlinear FCT method^{11,12} with an implicit correction process. This combination maintains high-order accuracy and yet removes the timestep limit imposed by the speed of sound. By using FCT for the explicit step, BIC-FCT is accurate enough to compute with sharp gradients without overshoots and undershoots. Thus spurious numerical oscillations that would lead to unphysical chemical reactions do not occur.

Thermal conductivity of the individual species is modelled by a polynomial fit in temperature to existing experimental data. Individual conductivities are then averaged using a mixture rule^{13,14} to get the thermal conductivity coefficient of the gas mixture. A similar process is used to obtain the mixture viscosity from individual viscosities. Heat and momentum diffusion are then calculated explicitly using these coefficients. All viscous terms in the compressible Navier-Stokes equation have been included. In the problem considered in this paper, the timestep imposed by the explicit method for the diffusion terms is comparable to that used in the fluid transport step. Thus, there is no loss in efficiency sometimes associated with explicit methods.

Mass diffusion also plays a major role in determining the properties of laminar flames. Binary mass diffusion coefficients are represented by an exponential fit to experimental data, and the individual species diffusion coefficients are obtained by applying mixture rules¹⁵. The individual species diffusion velocities are solved for explicitly by applying Fick's law followed by a correction procedure to ensure zero net flux¹⁴. This procedure is equivalent to using the iterative algorithm DFLUX¹⁶ to second order. This method is substantially faster than one that uses matrix inversions and is well suited for a vector computer.

Chemistry of the hydrogen-oxygen flame is modelled by a set of 24 reversible reaction rates describing the interaction of eight species, H_2 , O_2 , H , O , OH , HO_2 , H_2O_2 , H_2O , and N_2 is considered a nonreacting diluent¹⁶. This reaction set is solved at each timestep with a vectorized version of CHEMEQ, an integrator for stiff ordinary differential equations¹⁷. Because of the complexity of the reaction scheme and the large number of computational cells in a two-dimensional calculation, the solution of the chemical rate equations takes a large fraction of the total computational time. A special version of CHEMEQ called TBA was developed to exploit the special hardware features of the CRAY X-MP vector computer.

All of the chemical and physical processes are solved sequentially and then are coupled asymptotically by timestep splitting¹⁰. This modular approach greatly simplifies the model and makes it easier to test and change the model. Individual modules were tested against known analytic and other previously verified numerical solutions. One-dimensional predictions of the complete model were compared to those from the Lagrangian model FLAME1D which has been benchmarked extensively against theory and experiment¹².

III. Results and Discussion

Initial conditions for the two-dimensional calculations were obtained by performing a one-dimensional calculation to provide the conditions for steady, propagating flames. Figure 1 shows the configuration under study and gives the boundary conditions of the computational domain. Fresh unburned gas flows in from the left, and the products of chemical reaction at the flame front flow out to the right. If the inlet velocity is set to the burning velocity of the flame, the flame zone is fixed in space and there is a steady, propagating flame. Thus, the transient effects arising from the ignition process can be eliminated and the one-dimensional solution provides the initial condition for the two-dimensional calculation. The computational domain for the two-dimensional calculation was 5.1 cm x 12 cm, which was resolved by a 144 x 128 variably spaced grid. Fine zones were clustered around the flame front. Calculations have been performed for several hydrogen-air mixtures, ranging from 15% hydrogen down to 7.5% hydrogen. However, only calculations for some representative hydrogen-air mixtures will be discussed in detail.

15 - 12% Hydrogen-Air Mixtures

The results of our calculations for the various hydrogen air mixtures can be grouped roughly into three regimes, according to the cellular structures that are formed. The first regime consists of vigorously burning flames in mixtures which have greater than 12% hydrogen. The burning velocity for these flames exceeds 10 cm/s.

The first calculation which will be presented in detail as representative of flames in this regime is for a 12% hydrogen-air mixture, a flame that clearly exhibits cellular structure. The initial condition described by Fig. 1 is perturbed by displacing the center portion of the flame against the direction of the flow. The first set of frames in Fig. 2 show isotherms just after the perturbation, and their subsequent evolution in time. The second set of frames show the OH radical concentrations at these times. In each of these frames the fresh, unburnt mixture flows in from the left, and the combustion products flow out at the right. The entire width of the channel is shown in this and subsequent figures. By 30 ms, the large central cell which is formed

early in the calculation shows signs of splitting and several small cells flanking it have appeared. By 45 ms, numerous small cells can be seen. By 60 ms, the central cell has undergone multiple divisions and well defined cells are observed along the entire flame front. The mixtures in this regime can be considered quite reactive when their burning velocities of 10-30 cm/s are compared to the 0.5-2 cm/s observed in flames that do not split¹.

11 - 9.5% Hydrogen-Air Mixtures

This next regime of mixtures consists of less vigorously burning flames which have burning velocities between 2.5 and 10 cm/s. A flame burning in a 10% hydrogen-air mixture will be examined closely, as it is representative of the flames in this regime. The first fact that is apparent from Fig. 3 is that the evolution of cellular structure is much slower than in the 12% mixture, with the large central cell becoming prominent at 40 ms. This cell begins to split after 80 ms and by 160 ms has clearly split into two. In comparison to the previous regime of mixtures, the cells are larger and far fewer in number. Even by 180 ms only two cells are clearly discernable, but there is indication from the OH concentration at this time that these cells are in the process of splitting. In this regime the formation and splitting of cells is slower and fewer cells are formed.

9 - 7.5% Hydrogen-Air Mixtures

In Fig. 4, the temperature and OH concentration profiles are shown for a flame in a 9% hydrogen-air mixture. Flames in mixtures still leaner than this also exhibit similar behavior. These flames have burning velocities less than 2 cm/s. As expected from the trend seen in the other regimes, the cellular structure evolves very slowly. However, the most striking feature of this flame is the fact that only one large cellular structure is formed. This structure, which becomes clearly visible by 40 ms, continues to grow very slowly in size as time progresses. Even by 280 ms, this single cell remains intact, and even the OH concentration at this time does not provide any indication that this cell will split. Cell splitting also does not occur in the still leaner mixtures which have been modelled.

The Cell-Split Limit

The three regimes identified above agree qualitatively with the observations from experiments done in microgravity by Ronney¹. In these experiments, cells formed but did not split further in mixtures with less than 5.5% of hydrogen. From the numerical modelling presented in this study, it appears that flames in mixtures with 9% or less hydrogen form stable cellular structures that do not split. As in the experiments, this cell-split limit is sharp, as can be seen in Fig. 5. In this figure, the OH radical concentrations are presented for flames in a 9.5% and a 9% hydrogen-air mixture. By 80 ms, the

central cell has a more pronounced structure in the 9% mixture. By 200 ms in the 9.5% mixture, this central cell begins to divide and by 280 ms has completely split in two. Indeed, there are signs that these cells are in turn ready to split. In the 9% mixture, however, the central cell remains intact even up to 280 ms.

The temperature contours for the 9% mixture (Fig. 4) indicate that the single cell can grow quite large. This gives rise to the speculation that the confinement of the cell in the 5.1 cm channel might have led to the suppression of cell splitting. So if this speculation were correct, cell splitting might occur for the 9% mixture in a slightly wider channel. On the other hand, from the OH concentration contours at 280 ms in Fig. 5, the size of the cell is not much greater than that of the cells in the 9.5% mixture, suggesting that the channel width is close to being adequate. Furthermore, it is also possible that 280 ms is too short a time for cell splitting to occur in this mixture. Both these issues can be resolved by simulating the evolution of the cellular flame in this mixture for a longer time in a larger channel. Therefore, we have repeated the calculations for the 9% mixture in a 10.2 cm channel, using a computational mesh of 144×256 grid points. These calculations have been carried out up to 400 ms and are presented in Fig. 6. Though the single central cell grows larger, it does not split, even though the channel is fully twice as wide as in the other calculations. Also, these calculations have been carried out to a much later time, allowing a longer time for the structure to evolve. We conclude that the cell-split limit obtained here is independent of the size of the channel and the length of time of simulation.

IV. Summary and Conclusions

A detailed two-dimensional numerical simulation of multi-dimensional flame structures in lean hydrogen-air mixtures has been carried out for a range of mixtures between 7.5 and 15% hydrogen. Physical processes included in the model are: fluid convection, detailed hydrogen-oxygen chemistry, species diffusion, thermal conduction, and viscosity. The simulations show the characteristic cellular structure observed in experiments and predicted by theory. Brisk formation of cells and their subsequent splitting has been found in lean mixtures with greater than 11% hydrogen. In leaner mixtures with 9.5–11% hydrogen, the formation and splitting of cells was less vigorous. For mixtures with 9% or less hydrogen, a single cell is formed which does not split at all.

A complete cessation of cell splitting was observed between 9 and 9.5%, indicating that a cell-split limit also exists in our numerical calculations. However, the mixture at which this limit occurs is higher than the 5% mixture found in experimental observations. The rea-

sons for this discrepancy are not yet known and may be due to the many differences that exist between the experimental set-up and the numerical simulations. For example, the experiments were carried out in a spherical vessel, in which the cellular phenomena is three-dimensional. The numerical calculations simulated cellular flames in a two-dimensional channel. Also, the effect of the halon additive on the cellular structures in the experiments is not fully understood.

Heat losses have been postulated as a mechanism that can cause a cell-split limit. However, the simulations in this study do not include any loss mechanism; yet such a limit is still observed. This suggests that external loss mechanisms are not required and that the cell-split limit is actually an intrinsic feature of the lean hydrogen-air mixture itself.

Acknowledgements

This work was sponsored by NASA in the Microgravity Sciences Program and by the Office of Naval Research through the Naval Research Laboratory.

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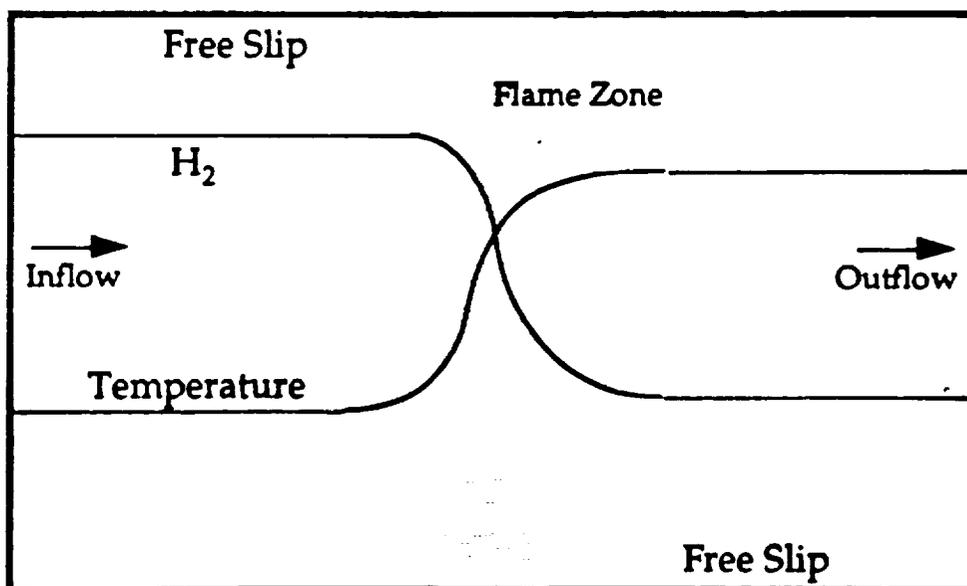


Figure 1. Initial and boundary conditions for the two-dimensional flame calculations.

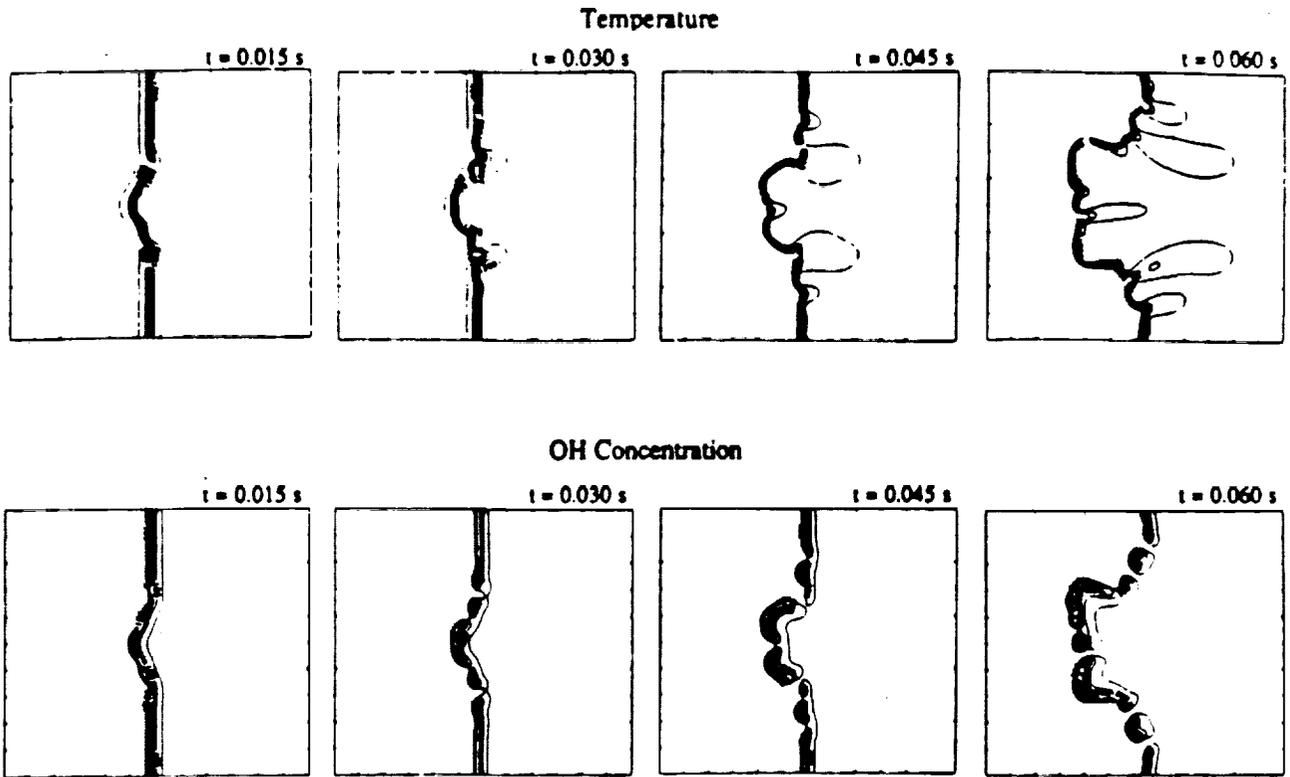


Figure 2. Cellular structures in a 12% hydrogen-air flame, 5.1 cm channel.

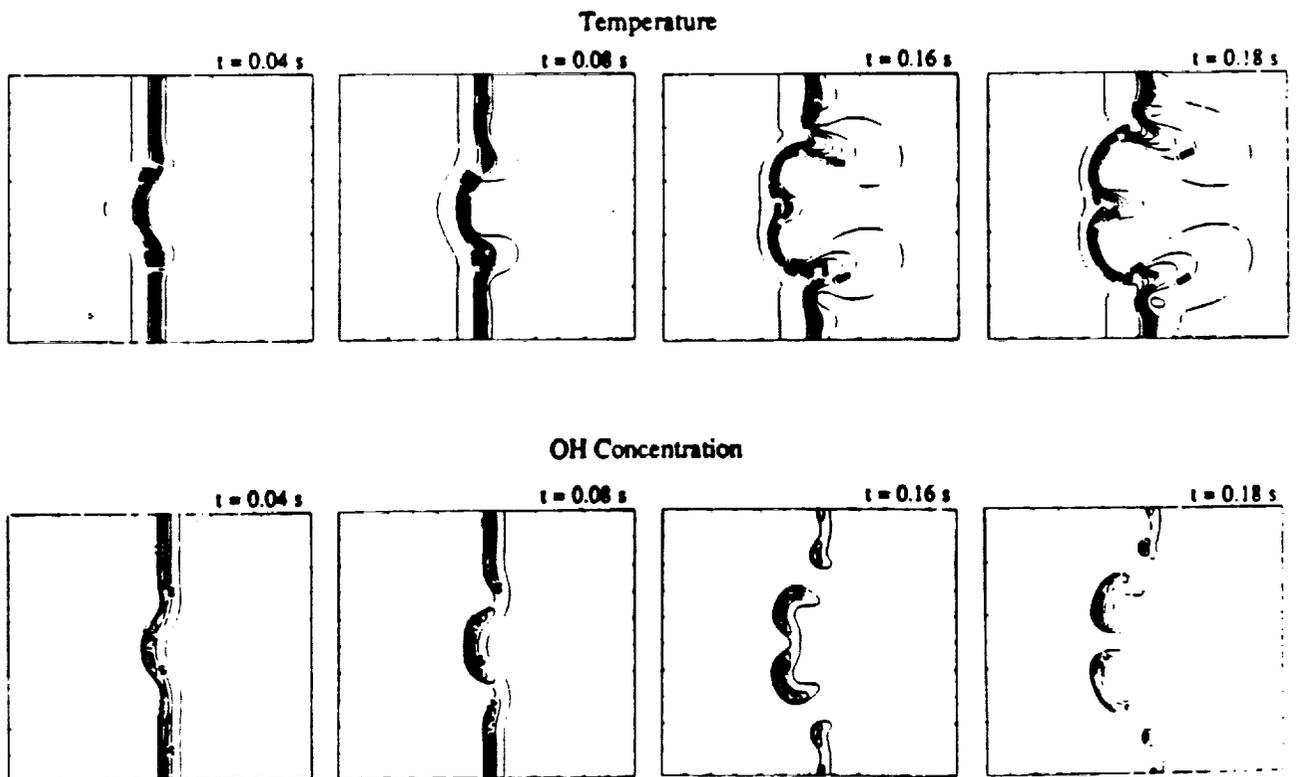


Figure 3. Cellular structures in a 10% hydrogen-air flame, 5.1 cm channel.

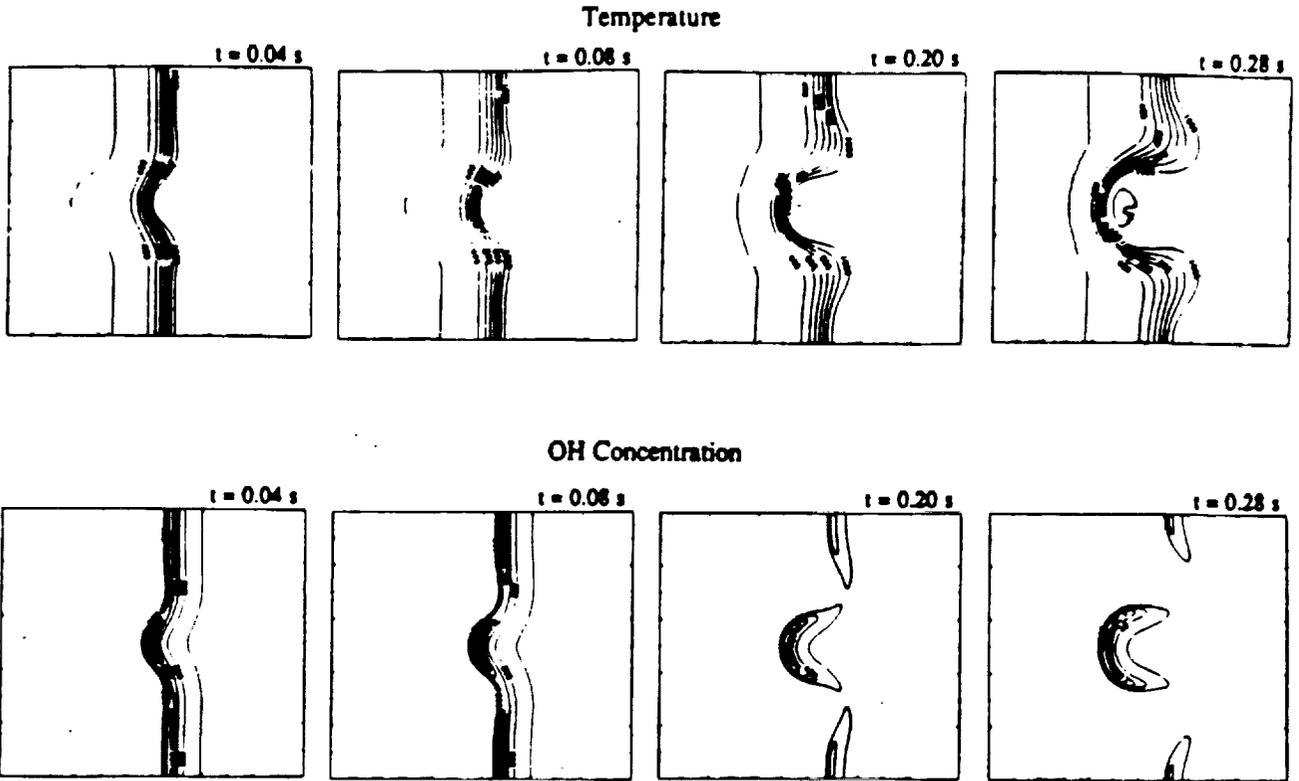


Figure 4. Cellular structures in a 9% hydrogen-air flame, 5.1 cm channel.

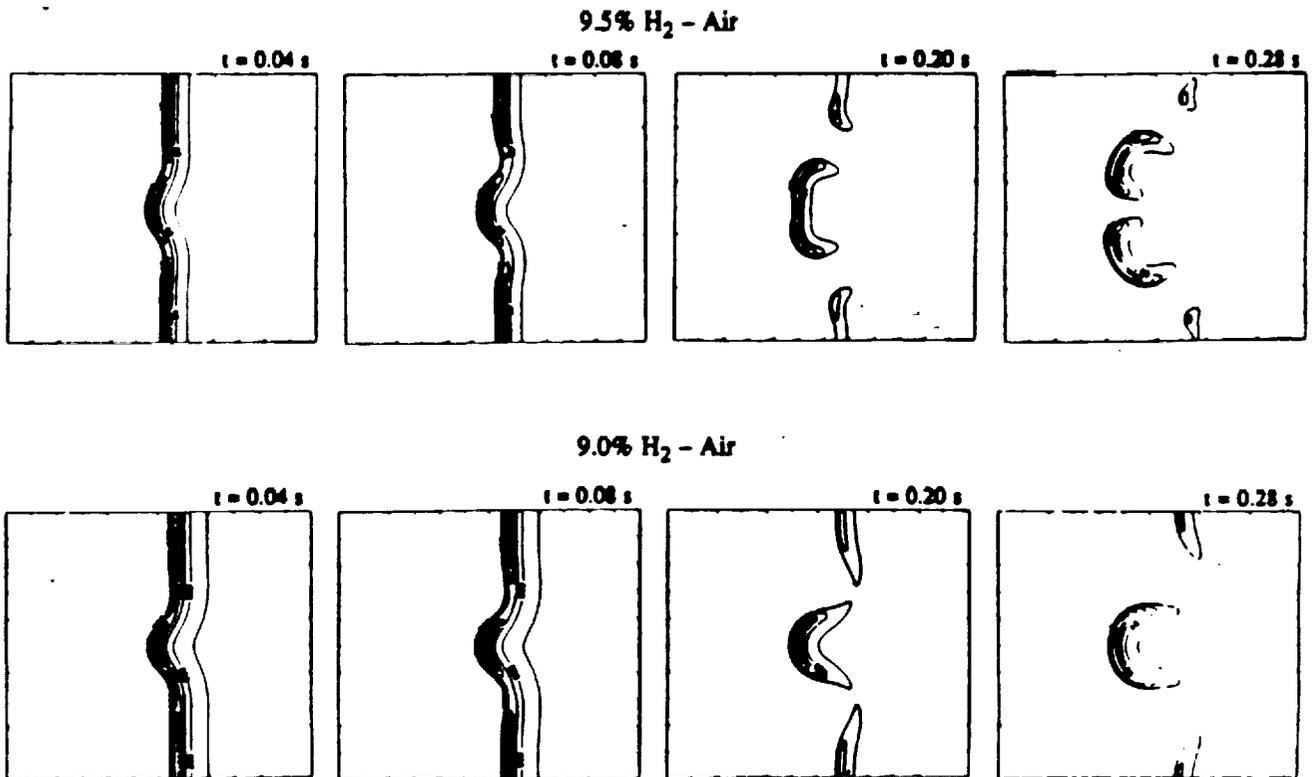


Figure 5. Comparison of OH concentration in 9.5 and 9% hydrogen-air flames.

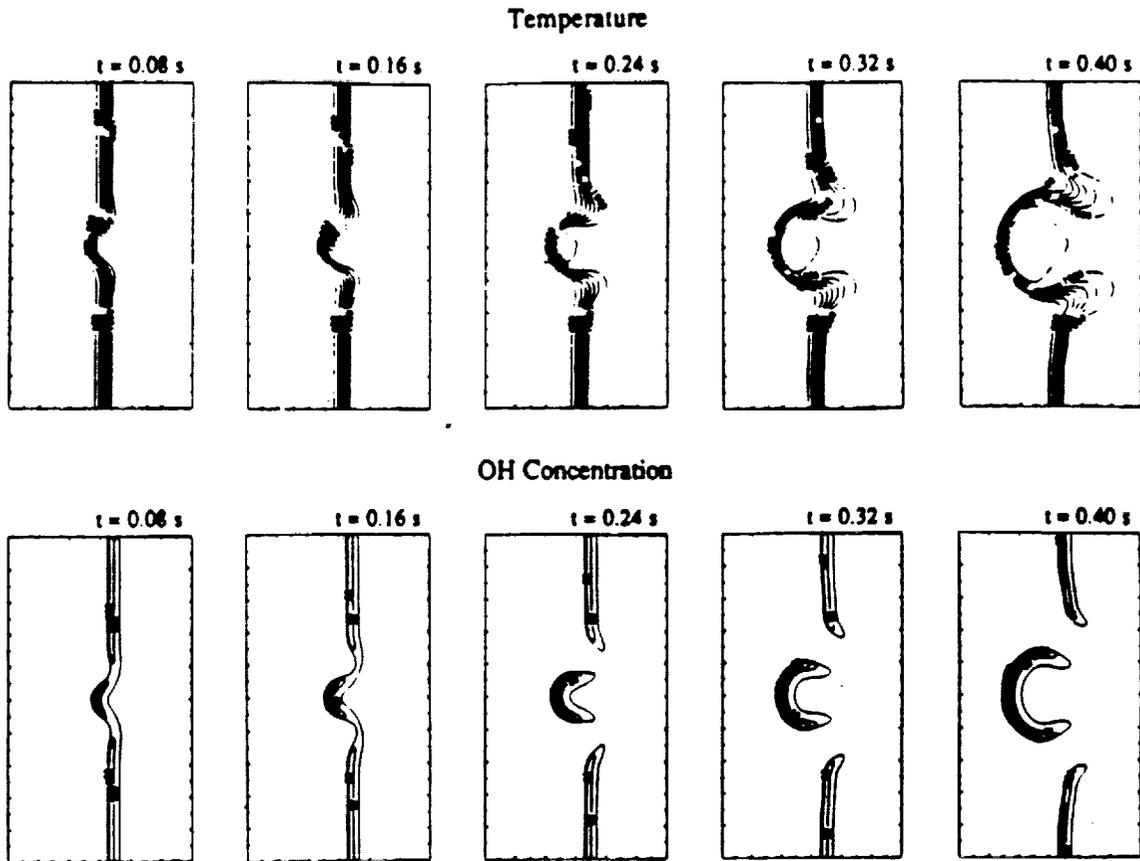
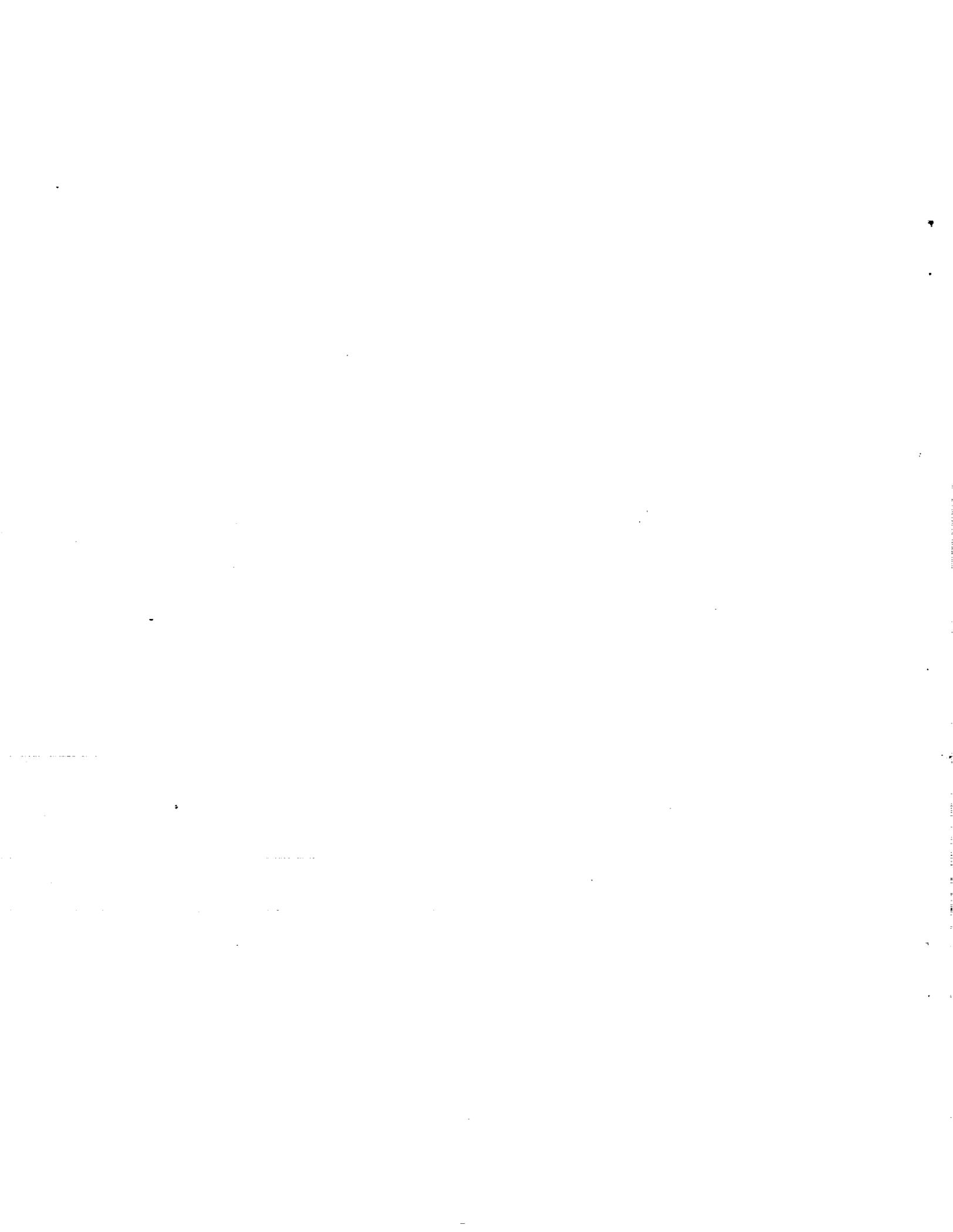


Figure 6. Cellular structures in a 9% hydrogen-air flame, 10.2 cm channel.

Appendix B

Effect of Gravity on the Stability and Structure of Lean Hydrogen-air Flames



EFFECT OF GRAVITY ON THE STABILITY AND STRUCTURE OF LEAN HYDROGEN-AIR FLAMES

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Detailed, time-dependent, two-dimensional numerical simulations with full hydrogen-oxygen chemistry are used to investigate the effects of gravity on the stability and structure of laminar flames in lean, premixed hydrogen-air mixtures. The calculations show that the effects of gravity becomes more important as the lean flammability limit is approached. In a 12% hydrogen-air mixture, gravity plays only a secondary role in determining the multidimensional structure of the flame with the stability and structure of the flame controlled primarily by the thermo-diffusive instability mechanism. However, in leaner hydrogen-air mixtures gravity becomes more important. Upward-propagating flames are highly curved and evolve into a bubble rising upwards in the tube. Downward-propagating flames are flat or even oscillate between structures with concave and convex curvatures. The zero-gravity flame shows only cellular structures. Cellular structures which are present in zero gravity can be suppressed by the effect of buoyancy for mixtures leaner than 11% hydrogen. These observations are explained on the basis of an interaction between the processes leading to buoyancy-induced Rayleigh-Taylor instability and the thermo-diffusive instability.

Introduction

Multi-dimensional structures are often observed in propagating flames, especially near the flammability limits. These structures arise from the various instabilities that can occur in premixed flames. In this paper, we examine these instabilities and use numerical simulations to isolate and study their effect on the flame structure. The emphasis of this paper is on the effects of gravity on flame instabilities and structure in gases of premixed hydrogen and air.

Linear stability analyses provide information on the roles of various processes at the onset of instability. However, the prediction of the growth of this instability to the final form is beyond the scope of these analyses. Numerical calculations can be used to help understand both the onset of the instability and the evolutionary process that produces the multidimensional structure. The two-dimensional numerical simulations of flames presented here include a multi-reaction mechanism for hydrogen combustion, molecular diffusion between the reactants, intermediates, and products, thermal conduction, convection, viscosity, and gravity. This detailed model allows us to investigate the multidimensional structure of flames and to evalu-

ate the relative importance of various instability mechanisms in normal earth gravity and zero-gravity conditions. Three-dimensional simulations may be required for a detailed quantitative determination of the cellular structure. However, three-dimensional simulations with the same level of detail as in these two-dimensional simulations are beyond the capability of current computers.

The three major instabilities that can occur in premixed flames are: the hydrodynamic instability, independently proposed by Landau¹ and Darrieus,² the thermo-diffusive instability investigated by Barenblatt³ and Zeldovich,⁴ and the buoyancy-induced instability, generally called the Rayleigh-Taylor instability.⁵ Our numerical model of the hydrogen flame includes all the physical mechanisms that lead to these instabilities and provides an ideal test-bed for studying flame instabilities. Hydrodynamic instability was the first kind of flame instability studied theoretically. The analyses of Landau and Darrieus showed that a planar flame, considered as a density discontinuity that propagates at a constant speed, is unstable to wavelengths of all sizes. Hydrodynamic instabilities can be expected to occur in all flames in the absence of stabilizing mechanisms. This instability is expected to become important in large systems which are much larger than the flame thickness. In the study presented here the hydrodynamic instability is not expected to be important because of the relatively small system size. The thermo-diffusive instability mechanism pro-

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posed by Zeldovich,⁴ Barenblatt *et al.*,⁵ and Sivashinsky⁶ involves a competition between mass diffusion of the deficient reactant and diffusion of heat in the mixture. For a simple one-step reaction, this mechanism predicts instability whenever the thermal diffusivity of the mixture is sufficiently smaller than the mass diffusivity of the reactant. For lean hydrogen-air mixtures, as examined in this study, hydrogen is the deficient reactant and its mass diffusivity significantly exceeds the thermal diffusivity of the mixture. In rich hydrogen-air mixtures, oxygen is the deficient reactant and its mass diffusivity is nearly the same as the mixture thermal diffusivity. Hence this theory agrees with early experimental observations^{7,8} of unstable lean hydrogen-air mixtures and stable rich mixtures.

The Rayleigh-Taylor instability occurs when a heavier fluid is accelerated into a lighter fluid. On earth, this acceleration is provided by gravitational attraction. In an upward-propagating flame, the light, hot burned material is on the bottom, and the dense, cold unburned material is on the top, resulting in instability. In a downward-propagating flame, the light material is on the top, and the Rayleigh-Taylor mechanism stabilizes the system. The physical mechanisms causing the thermo-diffusive instability and this buoyancy-induced instability can be important simultaneously so that under certain conditions, this interaction appears to suppress the formation of cellular structure.⁹ Dimensional arguments¹⁰ and theoretical analysis¹¹ indicate that the importance of the buoyancy-induced instability increases as the flame speed decreases, and hence is more important when the mixture is near its flammability limits.

Cellular structures in flames have been observed in the microgravity experiments in the NASA drop tower^{12,13} and in aircraft in ballistic trajectories.¹⁴ These flames in microgravity are essentially free from any buoyancy-induced instability.

We have used numerical simulations to show that, in the absence of gravity, cellular structure is caused by the thermo-diffusive mechanism.¹⁵ In this paper, we investigate the effect of gravity on flame instability and, in particular, focus on the interaction between the processes leading to thermo-diffusive instability and the Rayleigh-Taylor instability and their effect on flame structure.

Multidimensional Flame Model

Our numerical simulations of premixed flames contain detailed models¹⁶ of the physical processes that cause the various instabilities. The reactive-flow conservation equations are solved for density, momentum, energy and the species number densities. In the rest of this section, we briefly describe the algorithms used to model and couple the various

physical processes. Full details of the numerical scheme can be found in Ref. 16.

The fluid convection algorithm must be able to maintain the sharp gradients present in flames. This means that the numerical diffusion in the calculation must be considerably less than any important physical diffusion. The BIC-FCT algorithm¹⁷ was developed specifically to solve low-speed flow problems with high accuracy. BIC-FCT combines an explicit high-order, nonlinear FCT method¹⁸ with an implicit correction process. This combination maintains monotonicity and high accuracy but removes the timestep limit imposed by the speed of sound. By using BIC-FCT, spurious numerical oscillations that would lead to unphysical chemical reactions do not occur.

Thermal conductivity of the individual species is modeled by a polynomial fit in temperature to existing experimental data. Individual conductivities are then averaged using a mixture rule^{19,20} to get the mixture thermal conductivity. A similar process is used to obtain the mixture viscosity from individual viscosities. Heat and momentum diffusion are then calculated explicitly using these coefficients. All viscous terms in the compressible Navier-Stokes equation have been included.

Binary mass diffusion coefficients are represented by an exponential fit to experimental data, and the individual species diffusion coefficients are obtained by applying mixture rules.¹⁹ The individual species diffusion velocities are solved for explicitly by applying Fick's law followed by a correction procedure to ensure zero net flux.²⁰

Chemistry of the hydrogen-oxygen flame is modeled by a set of 24 reversible reactions describing the interaction of eight species, H_2 , O_2 , H , O , OH , HO_2 , H_2O_2 , H_2O , and N_2 is considered a non-reacting diluent.²¹ This reaction set is solved at each timestep with TBA, a vectorized version of CHEMEQ, an integrator for stiff ordinary differential equations.²²

All the chemical and physical processes are solved sequentially and then are coupled asymptotically by timestep splitting.²³ This modular approach greatly simplifies the model and makes it easier to test and change the model. Individual modules were tested against known analytic and other previously verified numerical solutions. One-dimensional predictions of the complete model were compared to those from the Lagrangian model, FLAME1D, which has been benchmarked extensively against theory and experiment.¹⁹

Results and Discussion

Initial conditions for the two-dimensional calculations were obtained by performing a one-dimensional calculation to provide the conditions for a

LEAN HYDROGEN-AIR FLAMES

steady, propagating flame. If the inlet velocity of the unburned mixture is set to the burning velocity of the flame, the flame is fixed in space and there is a steady, propagating flame. Thus, the transient effects arising from the ignition process can be eliminated and the one-dimensional solution provides the initial condition for the two-dimensional calculation. The computational domain for the two-dimensional calculation was $5.1 \text{ cm} \times 12 \text{ cm}$, which was resolved by a 144×128 variably spaced grid. Fine zones were clustered around the flame front. Calculations have been performed for several H_2 -air mixtures, ranging from 15% hydrogen down to 7.5% hydrogen. However, only calculations for 12, 11, and 10% hydrogen-air mixtures will be presented in detail.

12% Hydrogen-Air Flames:

The first series of calculations described here are of flames in a fuel-lean mixture of 12% hydrogen

in air. Flames propagating upward and downward in normal gravity and flames propagating in a zero-gravity environment are simulated. The initial condition is obtained by displacing the center portion of the planar flame against the direction of the flow. A smaller initial disturbance merely delayed the formation of the structures. In each case, the effect of the disturbance on the planar flame and the subsequent evolution of a multidimensional structure is shown in Fig. 1. This figure presents the OH radical concentrations for upward-, zero-gravity, and downward-propagating flames at a sequence of times. For the three cases in this figure (and in Fig. 3), the sequence of frames at various times are aligned such that time advances from the bottom to the top and the fresh, unburned mixture flows in from the top in each case. This convention in presentation has been adopted for ease in comparing the relative rate of changes in the three cases although this results in the apparent reversal of the direction of propagation of the downward-propagating flame.

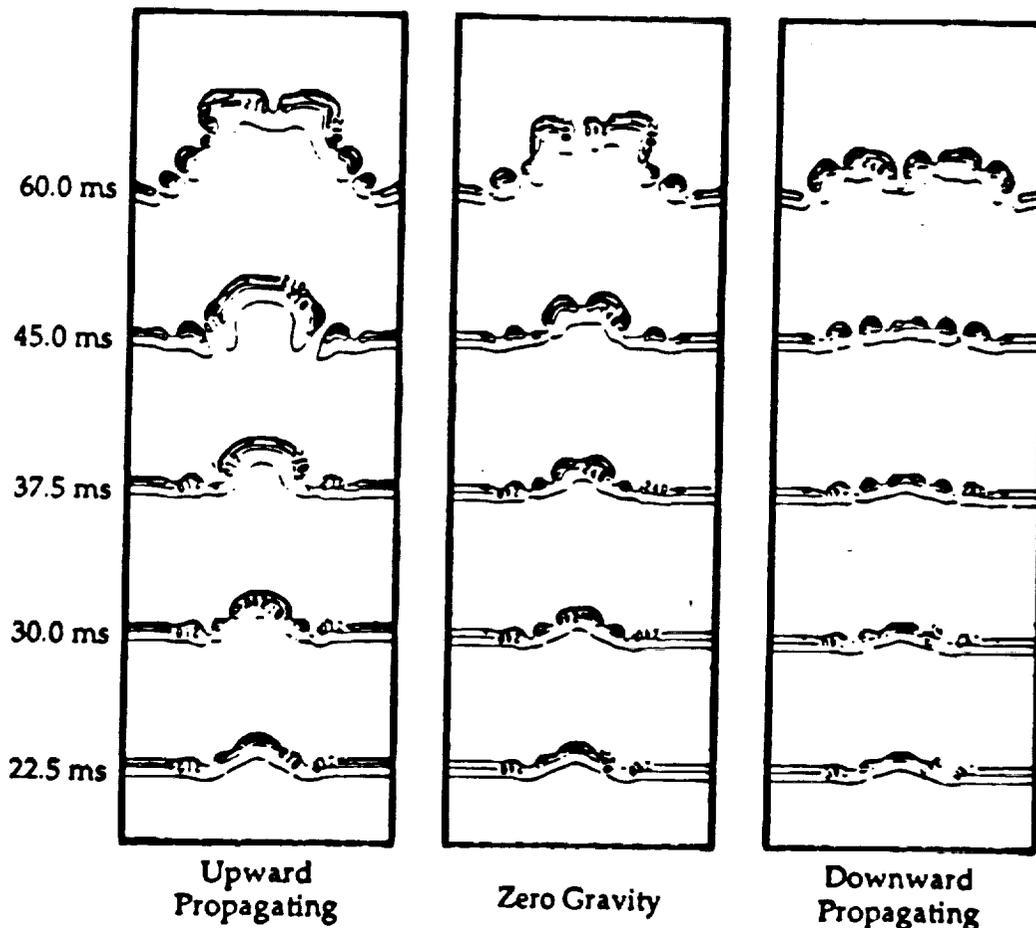


FIG. 1. OH radical number density contours for flames in a 12% H_2 -air mixture, contour levels scaled by 10^{-14} .

The first observation we make from this figure is that in all three cases, the planar flame is unstable and evolves into multidimensional structures and exhibit the characteristic features of cellular flames. To highlight the differences due to the effect of gravity, it is helpful to examine the shapes of the flames at two different length scales. The longer length scale is on the order of the size of the channel while the smaller length scale is on the order of the thickness of the flame. Using this perspective, the multidimensional structure of the flame can be described as a large-scale curvature of the flame front on which is superposed small scale cell-like structures. These small cells are due to the thermo-diffusive instability and are present in any gravity, independent of the direction of propagation. The shape and size of these cells appear to be largely unaffected by gravity.

However, gravity affects the large-scale shape of the flames. In the upward propagating flame, where the buoyancy forces are also destabilising in addition to the thermo-diffusive instability mechanism, the flame has a more curved shape than the zero-gravity flame. The downward-propagating flame has a flatter overall shape, with the large-scale curvature of the flame suppressed by the stabilizing effect of buoyancy. The most pronounced effect of gravity can be seen between 37.5 and 45 ms. During this period, the upward propagating flame

undergoes a bubble-like growth. For the downward propagating flame, the flame front flattens out.

An interesting feature of this mixture is that the cells which form grow and then split into two. These newly formed cells grow and subsequently split. This indicates that there is a preferred size for the cells, as has been observed in experiments.

The fact that a planar flame is unstable in all three cases and evolves to a cellular structure is consistent with our previous results¹⁵ that show that cellular structure is due to a thermo-diffusive instability mechanism. Our current calculations show that in this mixture, the effect of buoyancy is not substantial.

11% Hydrogen-Air Flames:

As expected, the 11% mixture exhibits a stronger effect of gravity, primarily on the overall large-scale shape of the flame and to a lesser extent on the evolution of the small scale cell-like structures. The upward propagating flame is highly curved and attains the appearance of a large, corrugated bubble rising up the channel. The zero-gravity flame is much less curved, without the bubble-like formation. The downward propagating flame has a flat overall shape throughout the calculation. The small scale cell-like structures are still present in all gravities. These cellular structures attain a larger size than in the

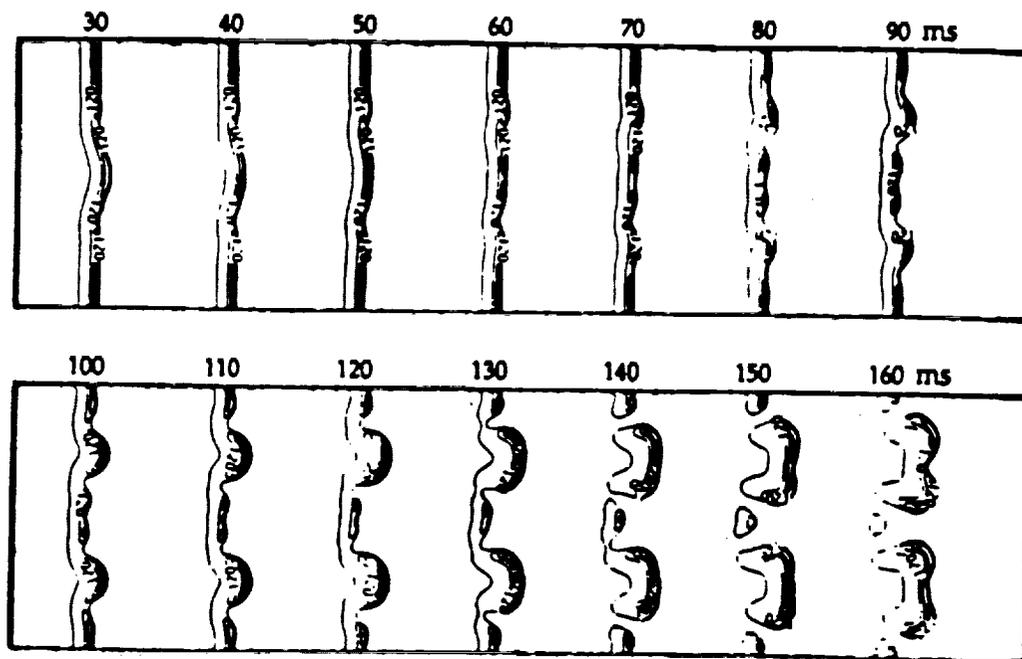


FIG. 2. OH radical number density contours for a downward propagating flame in a 11% H_2 -air mixture, contour levels scaled by 10^{14} .

LEAN HYDROGEN AIR FLAMES

12% mixture prior to splitting. The effect of gravity on the cellular structures is observed to be significant for this mixture.

Downward propagating flames are of special interest because their cellular structures have been investigated experimentally.²⁴ Therefore we have extended the calculations for this case up to 160 ms, as shown in Fig. 2. The flame is seen to be flat, with distinct cellular structures. These cellular structures split in two when they grow beyond a critical size. The newly created cells grow and split in turn. Again, a preferred cell size is established.

10% Hydrogen-Air Flames:

Figure 3 shows the OH contours for 10% hydrogen-air flames. The results for this mixture are very similar to those observed in the 11% mixture for both upward and zero-gravity flames. The evolution

of the structures is slower than in the less lean mixtures, with the effect of gravity becoming dominant later, at 80 ms. The evolution of the cellular structures and their splitting in zero gravity is also delayed. Other than the delay, both the large-scale and small-scale structures that evolve are quite similar to the 11% mixture.

However, a marked difference can be seen in the downward propagating flame. In this mixture, no small cells are formed, and the flame is quite smooth. The initial central cell is completely suppressed by the stabilizing effect of buoyancy, and unlike the less lean mixtures, no new cells are formed. The large scale structure of the flame undergoes a slow oscillation about its planar position. Extended calculations show that this oscillation is damping out. For the downward propagating flames in this mixture and in still leaner mixtures, the structures caused by the thermo-diffusive insta-

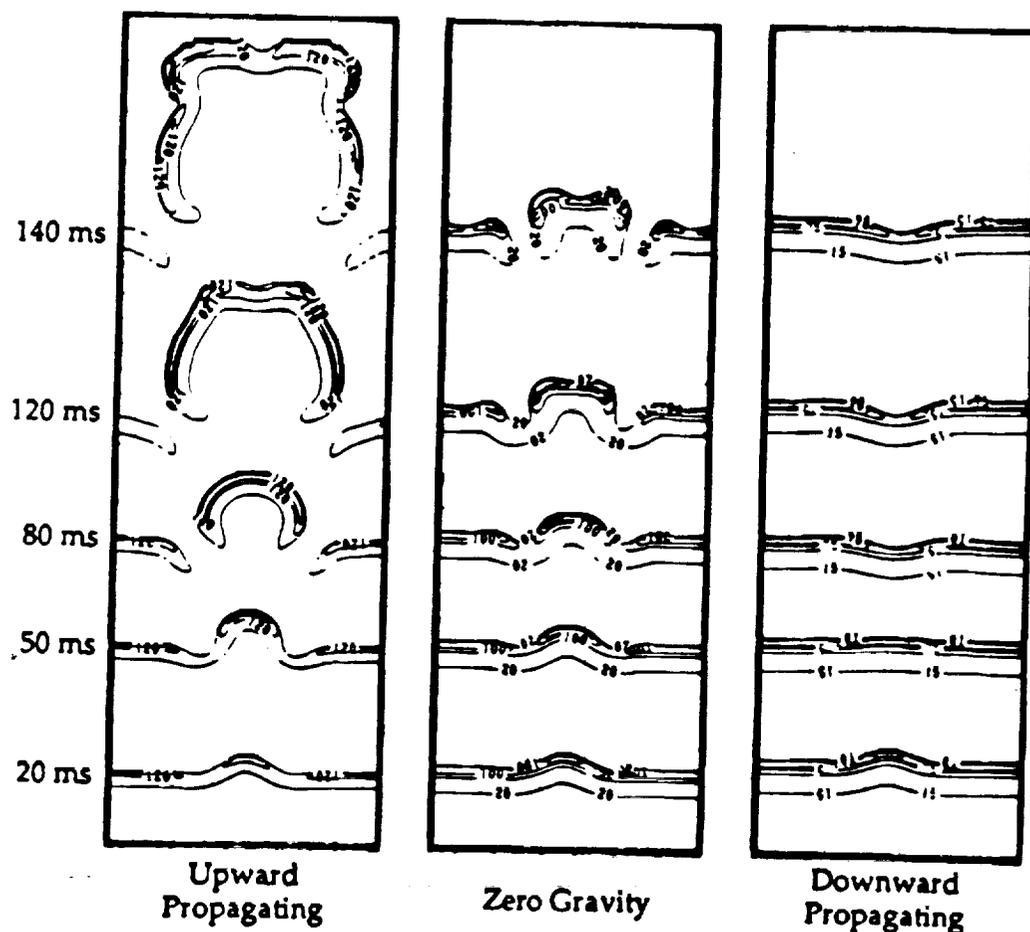


FIG. 3. OH radical number density contours for flames in a 10% H_2 -air mixture, contour levels scaled by 10^{14} .

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bility are overwhelmed by those arising from buoyancy. Any cellular structures which form early are overtaken by buoyancy effects with the result that the upward-propagating flame evolves into a bubble-like surface rising in the channel and the downward-propagating flame oscillating between mildly concave and convex flame shapes.

Conclusions

Detailed two-dimensional numerical simulations of flame instabilities in lean hydrogen-air mixtures have been carried out for upward and downward propagating and zero-gravity flames near the flammability limit. Physical processes included in the model are: fluid convection, detailed hydrogen-oxygen chemistry, multi-species diffusion, thermal conduction, viscosity, and gravity. The simulations show the characteristic cellular structure observed in experiments and predicted by theory. The thermo-diffusive instability was found to be present in the mixtures studied and at both orientations of gravity and is responsible for the small, cell-like structures. However, presence of the buoyancy-induced instability alters the large scale structure of the flame. The effect of buoyancy on the cellular structures formed by the thermo-diffusive instability is small in mixtures which have greater than 11% hydrogen. In a 11% mixture, a downward propagating flame is flat with small cells that repeatedly split. In 10% or leaner mixtures, the effect of gravity is more dramatic. In these mixtures, the upward propagating flame had the characteristic bubble shape observed experimentally⁸ and the downward propagating flame had oscillations characteristic of the Rayleigh-Taylor instability and does not have any cellular structures. These results agree with the theory¹¹ that indicates that the influence of gravity is greater for lower flame speeds and that such oscillatory behavior is possible. Our results indicate that the instability mechanisms can interact in a quite complex manner, and even though one mechanism can mask the other, in certain regimes, both can be equally important.

Calculations for still leaner mixtures are needed to address the actual extinction behavior of upward and downward-propagating flames. Loss mechanisms such as heat and radical losses to the walls as well as radiation might also play a role in determining the detailed extinction behavior of these flames. These effects will be systematically considered in further calculations. Since the experimentally observed cellular flames are three-dimensional, some three-dimensional calculations will have to be carried out to assess the role of other modes of instability that can grow in the third dimension.

However, further advances in computers are required before three-dimensional simulations can be done with the same amount of detail as in the two-dimensional simulations presented in this paper. The numerical methods used in this study can be extended readily to three dimensions.

Acknowledgments

This work was sponsored by NASA in the Microgravity Sciences Program and by the Office of Naval Research through the Naval Research Laboratory.

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LEAN HYDROGEN AIR FLAMES

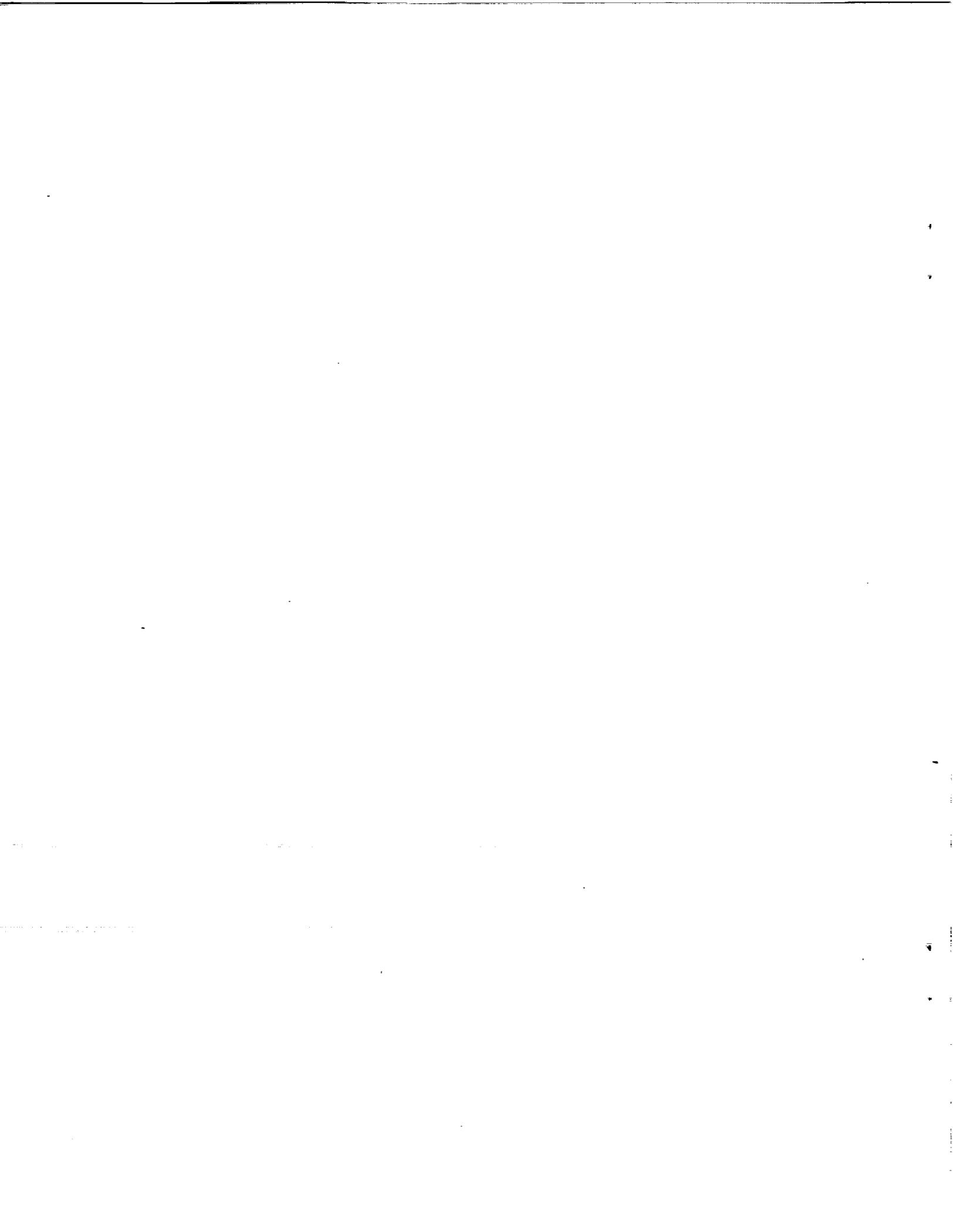
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Appendix C

Effect of Heat Losses on the Cellular Structure of Lean Hydrogen-air Flames





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**EFFECT OF HEAT LOSSES
ON THE CELLULAR STRUCTURE
OF LEAN HYDROGEN-AIR FLAMES**

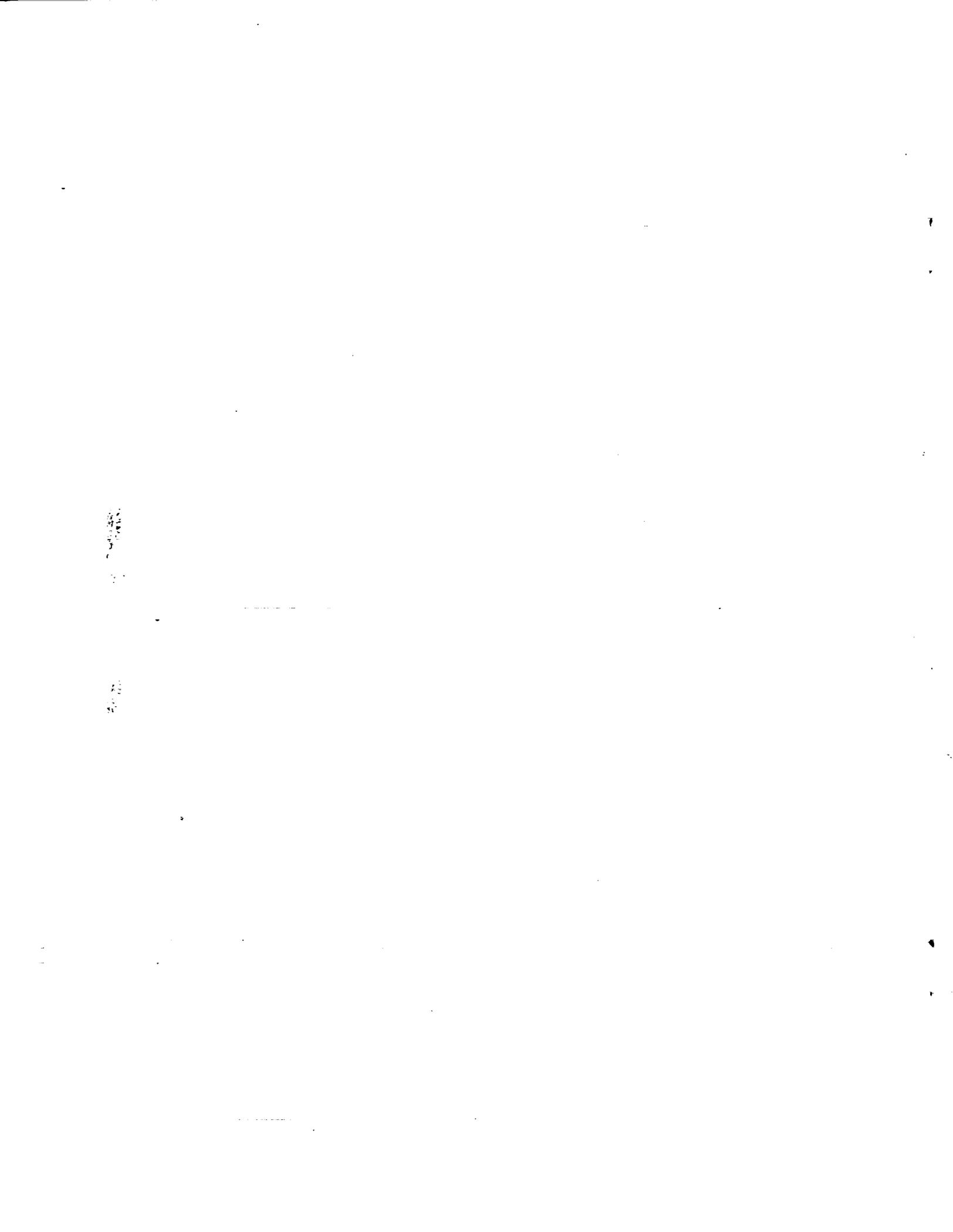
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29th Aerospace Sciences Meeting

January 7-10, 1991/Reno, Nevada



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Abstract

The effects of viscosity and heat losses to walls on the cellular structure of flames have been investigated using detailed numerical simulations. The simulations have been performed using a time-dependent, two-dimensional model that includes hydrogen-oxygen combustion with 24 elementary reactions involving eight reactive species and a nitrogen diluent, molecular diffusion of all species, thermal conduction, viscosity, convection, gravity and heat losses to the confining walls. The simulations of flames in zero-gravity show that wall heat losses act as an additional perturbation on the flames and can promote the formation of multiple cells. The effects of viscous losses to the walls creates dramatic changes in the structures of flames confined within adiabatic walls. However, the effects of viscosity are found to be secondary and can be neglected when more realistic non-adiabatic walls are used. Heat losses to the walls are also found to significantly alter some of the previous observations on the effects of gravity on flames.

I. Introduction

Planar flames in many fuel-air mixtures are unstable to disturbances and evolve into multidimensional flames with complex structures. One such flame, known as a cellular flame, has a distinctive cell-like structure. The underlying mechanism responsible for the formation of these cellular structures has been identified by simplified theoretical analyses¹⁻³ and detailed numerical simulations⁴ to be a thermo-diffusive instability mechanism. This instability mechanism involves a competition between mass diffusion of the deficient reactant and diffusion of heat in the mixture; that is, it depends on the effective Lewis number. For lean hydrogen-air mixtures, hydrogen is the deficient reactant and its mass diffusivity significantly exceeds the thermal diffusivity of the mixture and therefore planar flames are predicted to be unstable in such mixtures. Indeed, cellular flames are observed in lean mixtures both in experiments^{5,6} and in numerical simulations^{4,7}.

Recent experiments in a microgravity environment have shown that the structure and dynamics of cellular flames in lean hydrogen-air mixtures is quite complex and interesting phenomena such as multiple cell splitting and cessation of cell-splitting have been observed⁸.

These experiments were conducted in a microgravity environment in the NASA drop tower and used a large spherical combustion chamber to minimize the effects of confinement. However, gravity and confining walls are frequently encountered in most practical situations, and thus the understanding of these effects on the flame structure is important. An ideal way to isolate and study the effects of individual processes on the structure and dynamics of complex flames is by detailed numerical simulations which include all the relevant physical and chemical processes found in the flame.

The structure and dynamics of cellular flames have been studied using a two-dimensional flame model⁹ that includes detailed hydrogen-oxygen combustion with 24 elementary reactions of eight reactive species and a nitrogen diluent, molecular diffusion of all species, thermal conduction, viscosity, gravity and convection. Previous simulations with this model has shown cell splitting for sufficiently reactive mixtures and the existence of a "cell-split limit mixture" even in the absence of heat losses⁷. We have reported in a more recent paper⁸ the results of a systematic series of simulations on the effects of gravity on cellular flames in lean hydrogen-air mixtures. In this work, we now investigate the effects of viscosity and heat losses to the walls confining the flame in a two-dimensional channel. We also discuss how gravity modifies the structure and dynamics of flames in a isothermal, two-dimensional channel. We restrict our attention to a 10% H₂-air mixture, a mixture which clearly exhibits cellular structure and in which gravity can alter these structures significantly.

II. Multidimensional Flame Model

A detailed model of a flame must contain accurate representations of the convective, diffusive, and chemical processes. The individual importance of these processes varies from rich to lean flames, and is especially notable near the flammability limits¹⁰ where the exact behavior of these flames depends on a delicate balance among the processes. The reactive-flow conservation equations are solved for density, ρ , momentum, $\rho\vec{V}$, total energy E , and the number densities of individual species, n_k , $k = 1, \dots, n_p$, according to:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho\vec{V}) = 0,$$

$$\begin{aligned} \frac{\partial \rho \bar{V}}{\partial t} + \nabla \cdot (\rho \bar{V} \bar{V}) &= -\nabla P + \bar{F} - \nabla \times \mu \nabla \times \bar{V} + \nabla \cdot \left(\frac{4}{3} \mu \nabla \cdot \bar{V} \right), \\ \frac{\partial E}{\partial t} + \nabla \cdot (E \bar{V}) &= -\nabla \cdot (P \bar{V}) + \nabla \cdot (\kappa \nabla T) - \\ &\quad \sum_{k=1}^{n_s} \nabla \cdot (n_k h_k \bar{V}_k) + \sum_{r=1}^{n_r} Q_r, \\ \frac{\partial n_k}{\partial t} + \nabla \cdot (n_k \bar{V}) &= -\nabla \cdot (n_k \bar{V}_k) + w_k. \end{aligned}$$

Here \bar{V} is the fluid velocity, P is the pressure, μ is the coefficient of viscosity, \bar{F} is the body force due to gravity, κ is the thermal conductivity of the mixture of gases, h_k is the enthalpy of species k , \bar{V}_k is the diffusion velocity of species k , Q_r is the heat released from reaction r , and w_k is production of species k by chemical reaction. These equations are solved assuming that the individual species are ideal gases obeying the thermal equation of state,

$$P_k = n_k k T,$$

and that the differential relation between internal energy u and pressure P is given by

$$\delta u = \frac{\delta P}{\gamma - 1},$$

where γ , the ratio of specific heats of the mixture, is a function of its temperature and composition. In the rest of this section, we briefly describe the algorithms and input data used to model and couple the various physical processes. Full details of the numerical scheme can be found in Reference 9.

The fluid convection algorithm must be able to maintain the sharp gradients present in flames. Numerically this means that the numerical diffusion in the calculation must be considerably less than any important physical diffusion effect. Many explicit algorithms now exist that treat sharp discontinuities in flow variables accurately, but these methods are extremely inefficient at the very low velocities associated with laminar flames. The Barely Implicit Correction Flux-Corrected Transport (BIC-FCT) algorithm¹¹ was developed specifically to solve low-speed flow problems with high accuracy. BIC-FCT combines an explicit high-order, nonlinear FCT method^{12,13} with an implicit correction process. This combination maintains high-order accuracy and yet removes the timestep limit imposed by the speed of sound. By using FCT for the explicit step, BIC-FCT is accurate enough to compute with sharp gradients without overshoots and undershoots. Thus spurious numerical oscillations that would lead to unphysical chemical reactions do not occur.

Thermal conductivity of the individual species is modelled by a polynomial fit in temperature to exper-

imental data. Individual conductivities are then averaged using a mixture rule^{14,15} to get the thermal conductivity coefficient of the gas mixture. A similar process is used to obtain the mixture viscosity from individual viscosities. Heat and momentum diffusion are then calculated explicitly using these coefficients. All viscous terms in the compressible Navier-Stokes equation have been included. In the problem considered in this paper, the timestep imposed by the explicit method for the diffusion terms is comparable to that used in the fluid transport step. Thus, there is no loss in efficiency sometimes associated with explicit methods.

Mass diffusion also plays a major role in determining the properties of laminar flames. Binary mass diffusion coefficients are represented by an exponential fit to experimental data, and the individual species diffusion coefficients are obtained by applying mixture rules¹⁴. The individual species diffusion velocities are solved for explicitly by applying Fick's law followed by a correction procedure to ensure zero net flux¹⁵. This procedure is equivalent to using the iterative algorithm DFLUX¹⁶ to second order. This method is substantially faster than one that uses matrix inversions and is well suited for a vector computer.

Chemistry of the hydrogen-oxygen flame is modelled by a set of 24 reversible reaction rates describing the interaction of eight species, H_2 , O_2 , H , O , OH , HO_2 , H_2O_2 , H_2O , and N_2 is considered a nonreacting diluent¹⁷. This reaction set is solved at each timestep with a vectorized version of CHEMEQ, an integrator for stiff ordinary differential equations¹⁸. Because of the complexity of the reaction scheme and the large number of computational cells in a two-dimensional calculation, the solution of the chemical rate equations takes a large fraction of the total computational time. A special version of CHEMEQ called TBA was developed to exploit the special hardware features of the CRAY X-MP vector computer.

All of the chemical and physical processes are solved sequentially and then are coupled asymptotically by timestep splitting¹⁹. This modular approach greatly simplifies the model and makes it easier to test and change the model. Individual modules were tested against known analytic and other previously verified numerical solutions. One-dimensional predictions of the complete model were compared to those from the Lagrangian model FLAME1D which has been benchmarked extensively against theory and experiment¹³.

Wall Boundary-Conditions

A variety of wall boundary conditions were used in this study to determine the effect of heat loss and viscosity on the cellular structures found in the flame. Our previous studies^{4,7-9} ignored all effects at the wall other

than confinement of the flow. The boundary conditions used in those calculations correspond to an adiabatic slip wall, that is, one at which no heat or momentum is transferred. For the present study, the wall boundary conditions were generalized to allow any combination of adiabatic or isothermal, slip or no-slip walls. Thus it is possible to investigate either individually or jointly, the effects of heat loss or viscosity at the walls. If isothermal walls are used, the wall temperature is held fixed at the inlet temperature of the unburnt gas. Because we are interested in the time-dependent dynamics of the flame, these boundary conditions are imposed such that the simulations correspond to a flame propagating in a steady manner in a channel with adiabatic slip walls and at time $t = 0$ enters a region where the walls are isothermal and/or no-slip.

III. Results and Discussion

Initial conditions for the two-dimensional calculations were obtained by performing a one-dimensional calculation to provide the conditions for steady, propagating flames in a channel with adiabatic slip walls. Figure 1 shows the configuration under study and gives the boundary conditions of the computational domain. Fresh unburned gas flows in from the right, and the products of chemical reaction at the flame front flow out to the left. If the inlet velocity is set to the burning velocity of the flame, the flame zone is fixed in space and there is a steady, propagating flame. Thus, the transient effects arising from the ignition process can be eliminated and the one-dimensional solution provides the initial condition for the two-dimensional calculation. The computational domain for the two-dimensional calculation was 5.1 cm x 10 cm, which was resolved by a 144 x 128 variably spaced grid. Fine zones were clustered around the flame front. Calculations have been performed for several hydrogen-air mixtures. However, only calculations for a 10% hydrogen-air mixture will be discussed in detail.

Flames in Channels with Isothermal, No-slip Walls

The first calculation which will be presented in detail is representative of flames in the multiple cell-splitting regime and is for a 10% hydrogen-air mixture. The initial condition described by Fig. 1 is perturbed at $t = 0$ by displacing the center portion of the flame against the direction of the flow. The top set of frames in Fig. 2 show isotherms just after the perturbation, and their subsequent evolution in time. The second set of frames show the OH radical concentrations at these same times. In each of these frames the fresh, unburnt mixture flows in from the right, and the combustion products flow out to the left. The entire width of the channel is shown in this and subsequent figures.

The large central cell which is formed early in the

calculation shows signs of splitting by 100 ms. and by 140 ms, has clearly split in two. Even as early as 60 ms, a thermal boundary layer has developed at the cold walls. At this time, a small cell begins to form near where the flame meets the wall. This cell grows quickly and is well developed by 140 ms. At 180 ms, the cells formed at the wall closely resemble those in the middle of the channel.

Let us now compare these results to those from a case in which the walls are assumed to be adiabatic and free-slip. At first glance, the temperature and OH concentration contours in Fig. 3 look quite different from the corresponding contours in Fig. 2. However, they are quite similar if the central portions (away from the walls) are compared. For example, at 140 ms, the central cells in the two cases are quite similar, but no distinct cells are formed near the walls in the case without wall losses. Again, in both cases, the central cells split into two by 180 ms. So the first conclusion of this study is that the effect of heat and momentum losses to the walls appear to affect only the region the near wall and leave the basic cellular structure and dynamics unaffected. That is, the walls effects do not seem to couple strongly with the processes occurring in the middle of the channel and modify them.

Furthermore, looking more closely at the OH concentration contours in Fig. 3, two small cells seem to be developing away from the central cell by 180 ms. It is possible that after sufficient time, these cells will grow into larger ones, and the two cases may eventually resemble each other. If this were to happen, the effect of wall losses is primarily to act as an additional perturbation and enhance the rate of formation of new cells near the walls, but this aspect of the simulations needs further study. From the above two cases, the relative importance of heat and momentum losses is also not clear. Therefore, additional simulations were performed in which the two loss mechanisms were considered individually.

Effects of Viscosity

The effect of viscosity alone acting at the walls is seen in the upper frames in Fig. 4, in which temperature contours are shown at a sequence of times. The effect at first glance is quite dramatic. Again, a cell is formed at the wall, but this cell grows very rapidly and is already as big as the central cell by 60 ms. These cells near the wall begin to split by 140 ms and form numerous cells by 180 ms. Also quite apparent is that the wall cells move upstream faster than the cells in the center of the channel. This is due to the fact that these cells are in the slow moving fluid within the boundary layer at the wall, and thus the apparent flame speed, which is the burning rate including the fluid motion, is higher.

Effects of Heat Losses

The effect of heat loss alone, without viscosity acting at the wall is examined in the lower half of Fig. 4. Viscous effects are still included in the flow within the channel. This case should be contrasted to the comprehensive case including viscosity as presented in Fig. 2. The temperature contours are remarkably similar, indicating that heat losses dominate the behavior at the wall and that the role of viscosity, despite the dramatic appearance of the upper half of Fig. 4, is only secondary. The reason for the secondary role played by viscosity when there is heat loss, is that the cells near the walls are displaced away from the walls due to heat loss. In this and in the case with heat and momentum losses discussed previously, careful examination reveals that this displacement of the cells near the walls places them just above the leading edge of the boundary layer, and not fully embedded in the boundary layer itself. Therefore when heat losses to the wall are included, the effect of the momentum loss to the wall is only minor.

Effects of Gravity and Heat Losses

One of the on-going goals of our research on flames is the simulation of flames in a flammability-limit tube. Experimental studies of near-limit flames in this tube have observed that the flame structure is different depending on whether the flame is propagating upwards or downwards. This has been attributed to gravity acting via the Rayleigh-Taylor instability mechanism. We have also observed this dramatic difference in our numerical simulations⁸. Fig. 5 shows the OH concentrations in the absence of heat loss for upward-, zero gravity, and downward-propagating flames. A large corrugated bubble-like structure forms in the upward propagating flame, while the downward propagating flame is essentially flat and oscillates about its planar position. The zero gravity flame exhibits the cell-like structures seen in the earlier calculations.

In order to more closely simulate the flammability tube, we repeated the calculations, this time with isothermal walls. In view of the results we discussed in the previous section of this paper, viscous effects at the walls were neglected. The results for this case are presented in Fig. 6. Again, results are presented for upward-, zero gravity, and downward propagating flames. The zero gravity flame exhibits cellular structures, with both near-wall and central cells. The upward-propagating flame initially exhibits a large bubble, but is quite smooth, unlike in the previous case (Fig 5). The near-wall cells, which are prominent between 60 and 100 ms, disappear completely by 180 ms. Also by 180 ms, we observe that the flame is no longer a completely closed surface like a bubble but is actually composed of two hot fingers of gas. It qualitatively appears to be a two-dimensional version

of multi-fingered flames observed in experiments with lean hydrogen-air mixtures²⁰.

The downward-propagating flame is quite flat, and both the central cell and the wall cells all but disappear by 100 ms. At later times the flame takes on a shape slightly convex to the flow. Unlike the case without heat losses, no oscillations are seen, even when the calculations are carried out to a longer duration. While difficult to discern from the figure, the peak OH concentration in the downward case is only half that in the other cases, indicating that the flame is "weaker". The "visible" flame is present only in central portion of the channel. The appearance of the flame is again similar to those observed experimentally in near-limit mixtures²¹.

IV. Summary and Conclusions

Detailed two-dimensional numerical simulations have been carried out to investigate the effects of heat and momentum losses (due to viscosity) to the walls on the structure and dynamics of flames in a 10% hydrogen-air mixture. The simulations of flames in zero-gravity show that momentum losses to adiabatic walls result in flames with very complex shapes in which the portion of the flame near the walls moves significantly faster than the central portion of the flame. This phenomena occurs because of the slower flow velocities near the walls. However, viscous effects play only a secondary role when the walls are assumed to be isothermal, a more realistic boundary condition. In this case, heat losses to the walls play the dominant role resulting in the formation of additional cellular structures close to but slightly displaced from the walls. This displacement from the walls is a key factor in the secondary role played by viscous effects because it moves the flame to the outer edges of the viscous boundary layer.

Heat losses are found to significantly modify upward- and downward-propagating flames. In the upward-propagating case, instead of a single bubble rising in the tube, a two-fingered flame is observed. This flame has some features similar to experimentally observed flames in lean hydrogen-air mixtures. The downward-propagating flame is also quite different when the effect of heat losses are considered. In this case, a flame is "visible" only in the central portion of the channel. Again, this flame is qualitatively similar to some near-limit flames in experiments. Further calculations will provide valuable insights into the dynamical behavior observed experimentally in flames near extinguishment.

Acknowledgements

This work was sponsored by NASA in the Microgravity Sciences Program and by the Office of Naval Research through the Naval Research Laboratory.

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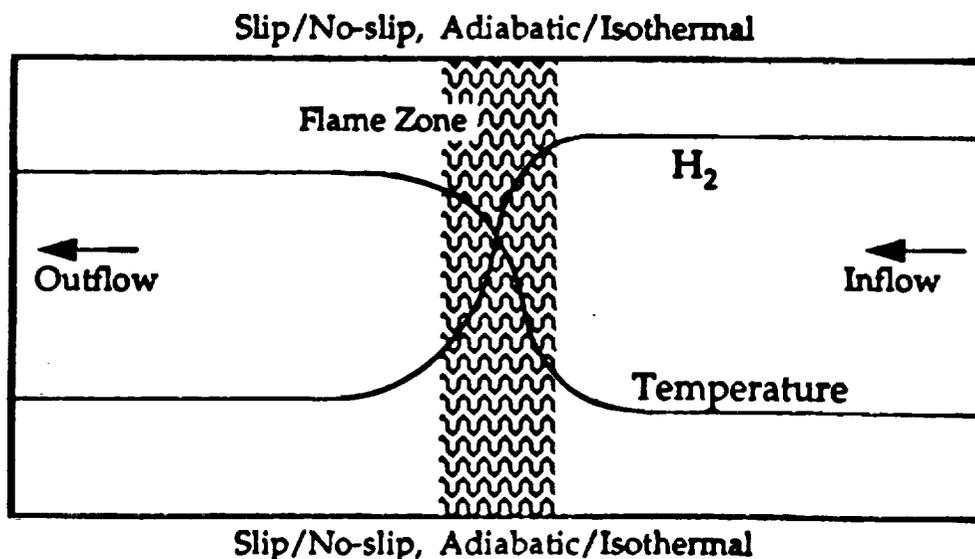
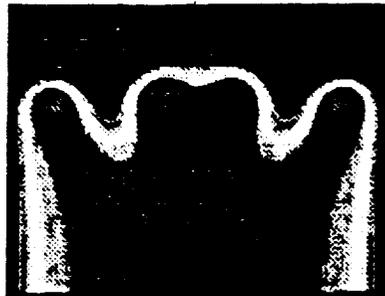
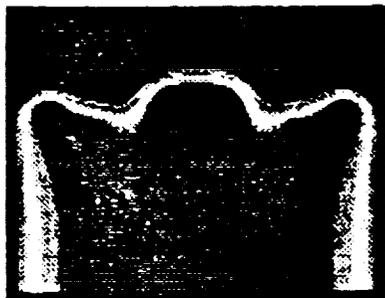
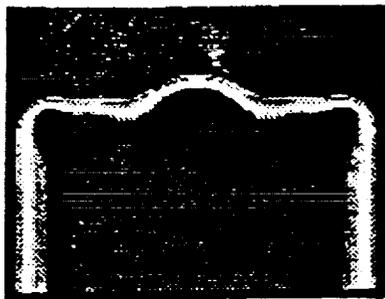
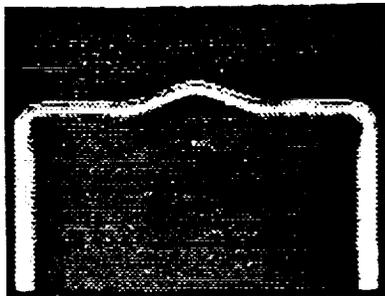


Figure 1. Initial and boundary conditions for the two-dimensional flame calculations.

Temperature



8

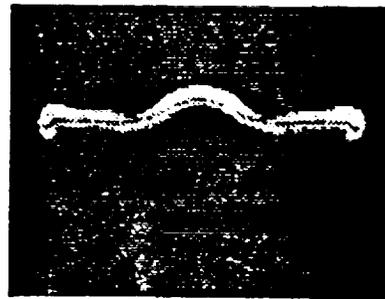
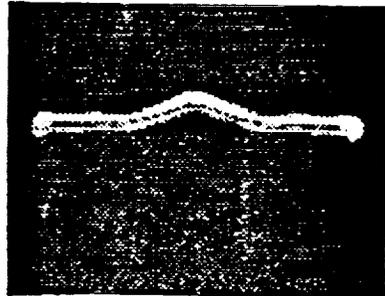
20 ms

60 ms

100 ms

140 ms

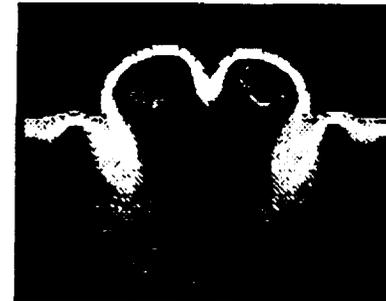
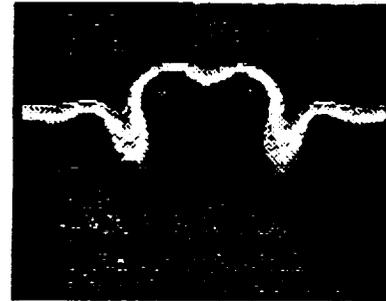
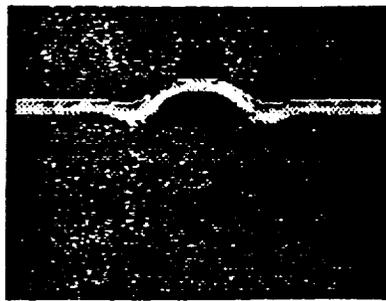
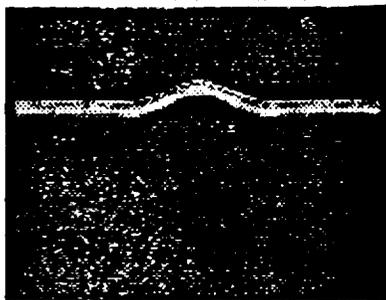
180 ms



OH Concentration

Figure 2. Cellular structures in a 10% hydrogen-air flame, isothermal no-slip walls.

Temperature



20 ms

60 ms

100 ms

140 ms

180 ms

OH Concentration

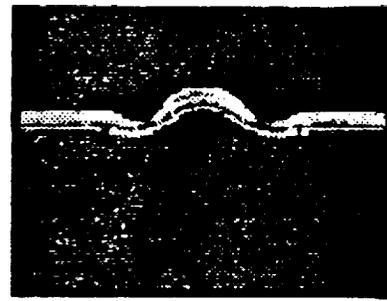
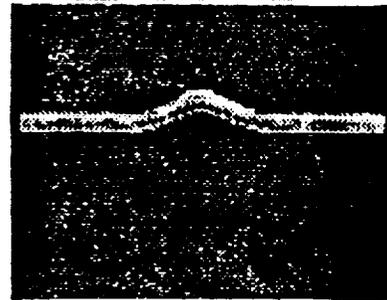


Figure 3. Cellular structures in a 10% hydrogen-air flame, adiabatic slip walls.

Adiabatic no-slip wall



Isothermal slip wall

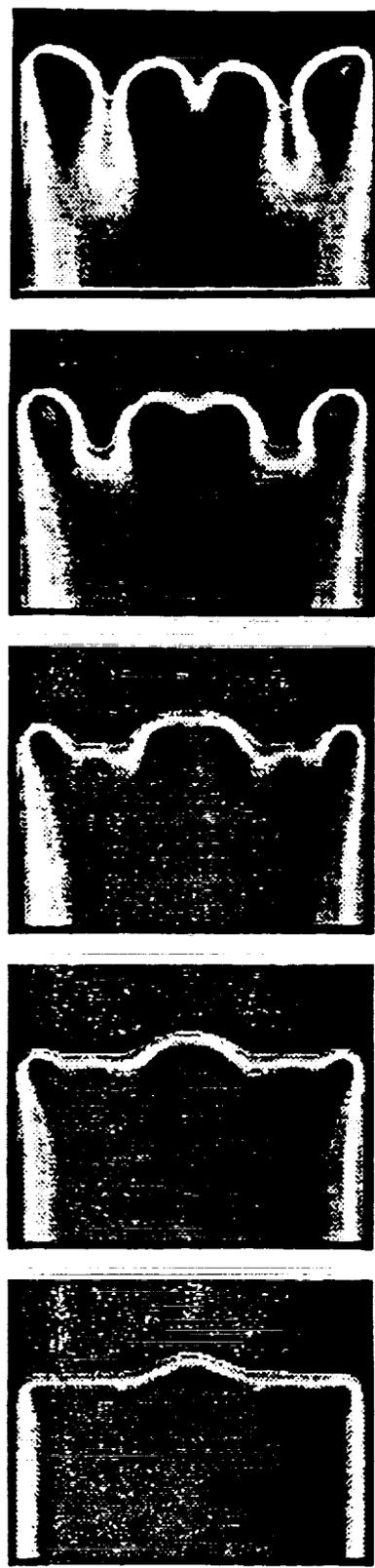


Figure 1. Temperature contours, effect of viscosity alone (above); effect of heat loss alone (below).

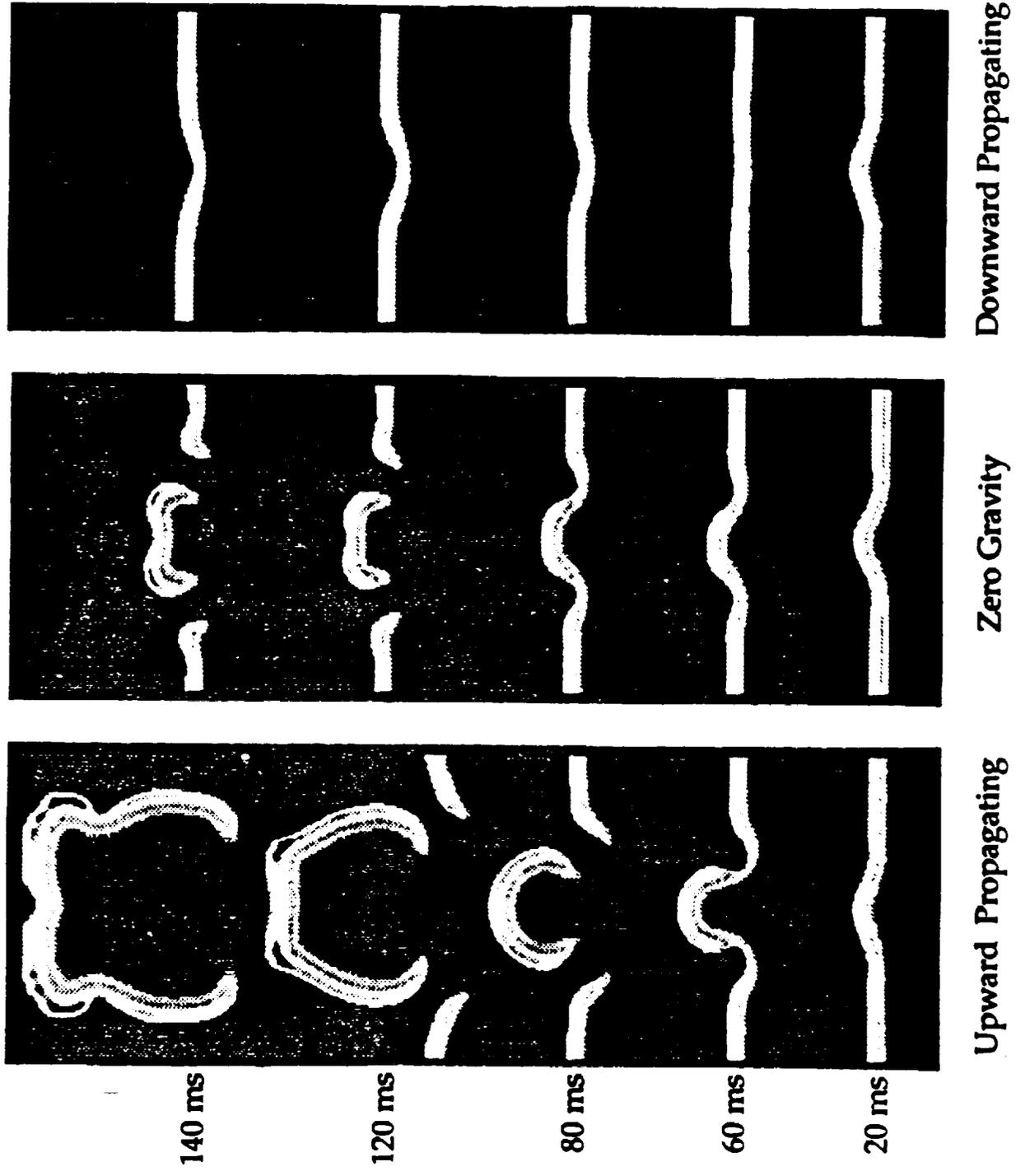


Figure 5. Comparison of OII concentration in upward-, zero-gravity, and downward-propagating flames, adiabatic walls.

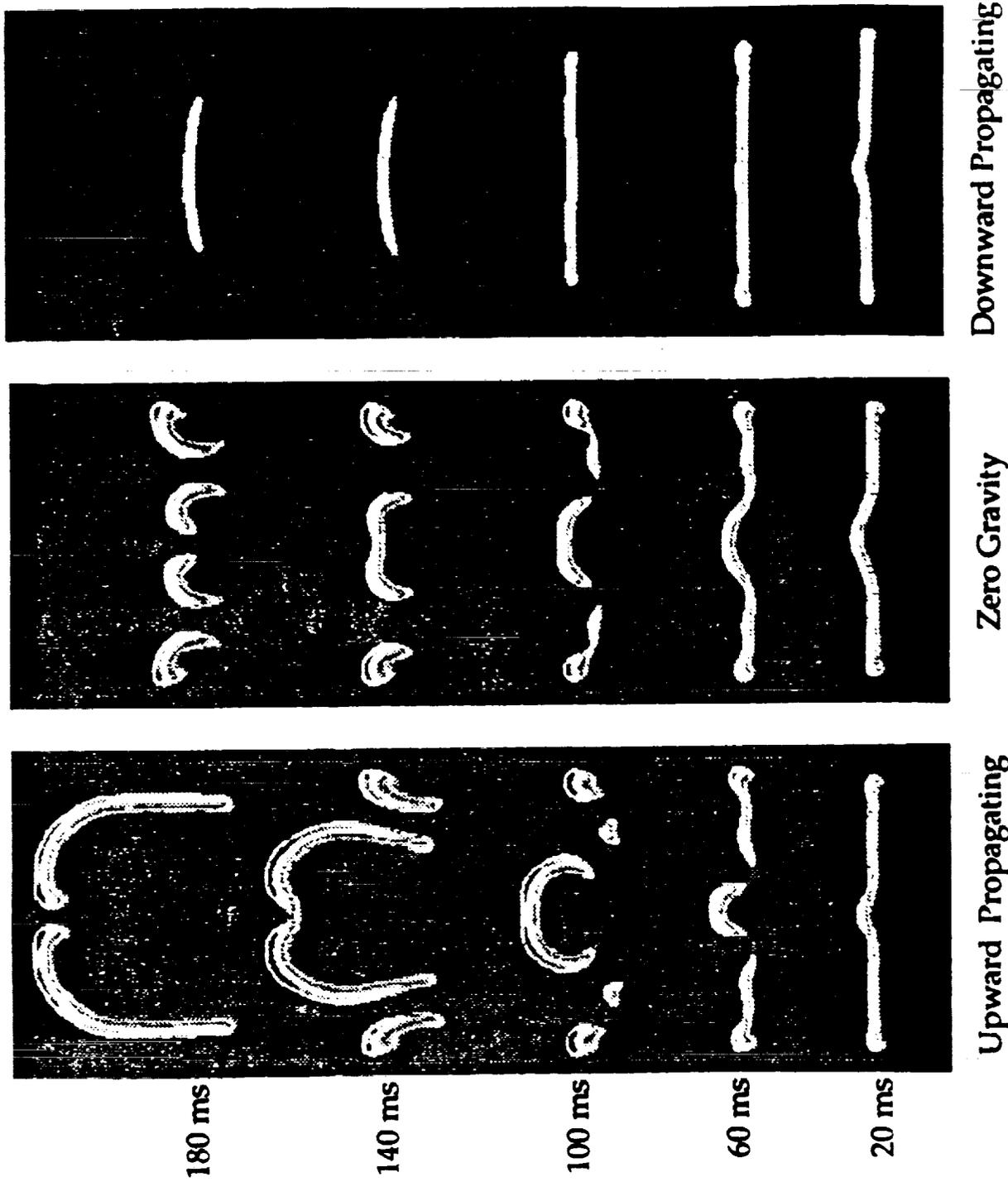


Figure 6. Comparison of Oil concentration in upward-, zero-gravity, and downward-propagating flames with heat loss to walls.

Appendix D

Lean Flammability Limit of Downward Propagating Hydrogen-Air Flames





AIAA-92-0336

**LEAN FLAMMABILITY LIMIT
OF DOWNWARD PROPAGATING
HYDROGEN-AIR FLAMES**

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Abstract

Detailed multidimensional numerical simulations that include the effects of wall heat losses have been performed to study the dynamics of downward flame propagation and extinguishment in lean hydrogen-air mixtures. The computational results show that a downward propagating flame in an isothermal channel has a flammability limit of around 9.75%. This is in excellent agreement with experimental results. Also in excellent agreement are the detailed observations of the flame behavior at the point of extinguishment. The primary conclusion of this work is that detailed numerical simulations that include wall heat losses and the effect of gravity can adequately simulate the dynamics of the extinguishment process in downward-propagating hydrogen-air flames. These simulations can be examined in detail to gain understanding of the actual extinction process.

I. Introduction

Over the years, the structure and dynamics of upward- and downward-propagating flames have been extensively studied in the laboratory and a number of review articles provide a comprehensive summary of the empirical observations^{1,2}. More recently, microgravity experiments have also added to our knowledge of the effects of the absence of gravity on near-limit flames³. Theoretical studies⁴ have examined the stability of these flames and give some insights into structure of the flame near extinguishment. In spite of all these studies, a detailed understanding of the mechanism or mechanisms of flame extinguishment and the role of factors such as gravity, stretch, chemistry and heat loss is not available from either experiment or theory alone.

Detailed multidimensional numerical simulations which include all the processes of relevance provide an ideal way to further our understanding of near-limit flames. Such numerical simulations are a valuable tool to investigate the relative importance of various processes and to identify the primary mechanism responsible for flame extinguishment under specific situations. In order to study these near limit flames several physical processes need to be included and have to be modelled to a sufficient level of detail. These processes include detailed chemistry, multi-species diffusion among many chemical species, thermal conduction, and fluid

convection. Body force due to gravity is important for these low speed flames and must be considered. Both buoyancy and fluid expansion will drive flow in the fluid, so an accurate model of compressible fluid flow is needed. Other physical processes such as radiation and soot formation are also important for many hydrocarbon fuels. The development of a numerical model which adequately includes all these processes is a formidable challenge, and this paper presents a look at our continuing efforts to solve this problem.

In the past, we have used a detailed two-dimensional numerical model to identify the primary mechanism responsible for the formation of cellular flames in hydrogen-oxygen-nitrogen mixtures⁵. The model used in that study included detailed hydrogen-oxygen kinetics with 24 elementary reactions involving eight species and a nitrogen diluent, molecular diffusion of all species, thermal conduction, viscosity, and fluid convection. Soot formation does not occur in hydrogen flames and was not included. Radiation does not play a large role in these low temperature hydrogen flames and was neglected.

Later, buoyancy driven effects were added to the model and this revised model was used to study the effects of gravity⁶ on the cellular structure and dynamics of lean hydrogen-air flames. This study showed that gravity plays a significant role in these lean flames and alters both the structure and the stability considerably. The behavior of the flame simulated by our numerical model agreed qualitatively with many experimental observations and theoretical predictions. However, quantitative agreement was not obtained, and this was due in part to the absence of heat losses in the model.

Our next study concentrated on the effect of heat losses and viscosity⁷ on lean hydrogen-air flames. At this stage, conductive heat losses and viscous effects at the confining walls were included in the model. We showed that heat losses alter the structure of these lean flames considerably. However, while viscosity acting at an adiabatic wall alters the shape of these flames dramatically, it was found to be unimportant in the presence of heat losses.

Experimental observations have shown that near-limit, upward-propagating flames are highly three-dimensional whereas downward-propagating flames are

more nearly two-dimensional; thus the use of a two-dimensional model is adequate for downward-propagating flames. Currently, the model is restricted to two-dimensions but extensions to three are planned.

In this paper, we report the results of our study on the structure and dynamics of downward-propagating flames near and at the lean-limit of hydrogen in air. After a brief description of the numerical model and the boundary conditions used, we describe a series of simulations of near-limit flames. These simulations include both the effects of buoyancy and heat loss to the wall. Finally, the observations from the various simulations are summarized and their implications on the mechanism responsible for the extinguishment of downward-propagating flames are discussed.

II. Multidimensional Flame Model

A detailed model of a flame must contain accurate representations of the convective, diffusive, and chemical processes. The individual importance of these processes varies from rich to lean flames, and is especially notable near the flammability limits⁸ where the exact behavior of these flames depends on a delicate balance among the processes. The reactive-flow conservation equations are solved for density, ρ , momentum, $\rho\vec{V}$, total energy E , and the number densities of individual species, n_k , $k = 1, \dots, n_s$, according to:

$$\begin{aligned} \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho\vec{V}) &= 0, \\ \frac{\partial \rho\vec{V}}{\partial t} + \nabla \cdot (\rho\vec{V}\vec{V}) &= -\nabla P + \vec{F} - \nabla \times \mu \nabla \times \vec{V} + \nabla \cdot \left(\frac{4}{3} \mu \nabla \cdot \vec{V} \right), \\ \frac{\partial E}{\partial t} + \nabla \cdot (E\vec{V}) &= -\nabla \cdot (P\vec{V}) + \nabla \cdot (\kappa \nabla T) - \\ &\quad \sum_{k=1}^{n_s} \nabla \cdot (n_k h_k \vec{V}_k) + \sum_{r=1}^{n_r} Q_r, \\ \frac{\partial n_k}{\partial t} + \nabla \cdot (n_k \vec{V}_k) &= -\nabla \cdot (n_k \vec{V}_k) + w_k. \end{aligned}$$

Here \vec{V} is the fluid velocity, P is the pressure, μ is the coefficient of viscosity, \vec{F} is the body force due to gravity, κ is the thermal conductivity of the mixture of gases, h_k is the enthalpy of species k , \vec{V}_k is the diffusion velocity of species k , Q_r is the heat released from reaction r , and w_k is production of species k by chemical reaction. These equations are solved assuming that the individual species are ideal gases obeying the thermal equation of state,

$$P_k = n_k k T,$$

and that the differential relation between internal energy u and pressure P is given by

$$\delta u = \frac{\delta P}{\gamma - 1},$$

where γ , the ratio of specific heats of the mixture, is a function of its temperature and composition. In the rest of this section, we briefly describe the algorithms and input data used to model and couple the various physical processes. Full details of the numerical scheme can be found in Reference 9.

The fluid convection algorithm must be able to maintain the sharp gradients present in flames. Numerically this means that the numerical diffusion in the calculation must be considerably less than any important physical diffusion effect. Many explicit algorithms now exist that treat sharp discontinuities in flow variables accurately, but these methods are extremely inefficient at the very low velocities associated with laminar flames. The Barely Implicit Correction Flux-Corrected Transport (BIC-FCT) algorithm¹⁰ was developed specifically to solve low-speed flow problems with high accuracy. BIC-FCT combines an explicit high-order, nonlinear FCT method¹¹ with an implicit correction process. This combination maintains high-order accuracy and yet removes the timestep limit imposed by the speed of sound. By using FCT for the explicit step, BIC-FCT is accurate enough to compute with sharp gradients without overshoots and undershoots. Thus spurious numerical oscillations that would lead to unphysical chemical reactions do not occur.

Thermal conductivity of the individual species is modelled by a polynomial fit in temperature to experimental data. Individual conductivities are then averaged using a mixture rule^{12,13} to get the thermal conductivity coefficient of the gas mixture. A similar process is used to obtain the mixture viscosity from individual viscosities. Heat and momentum diffusion are then calculated explicitly using these coefficients. All viscous terms in the compressible Navier-Stokes equation have been included. In the problem considered in this paper, the timestep imposed by the explicit method for the diffusion terms is comparable to that used in the fluid transport step. Thus, there is no loss in efficiency sometimes associated with explicit methods.

Mass diffusion also plays a major role in determining the properties of laminar flames. Binary mass diffusion coefficients are represented by an exponential fit to experimental data, and the individual species diffusion coefficients are obtained by applying mixture rules¹². The individual species diffusion velocities are solved for explicitly by applying Fick's law followed by a correction procedure to ensure zero net flux¹³. This procedure is equivalent to using the iterative algorithm DFLUX¹⁴ to second order. This method is substantially faster than one that uses matrix inversions and is well suited for a vector computer.

Chemistry of the hydrogen-oxygen flame is modeled by a set of 24 reversible reaction rates describing the interaction of eight species, H_2 , O_2 , H , O , OH , HO_2 , H_2O_2 , H_2O , and N_2 is considered a nonreacting diluent¹⁵. This reaction set is solved at each timestep with a vectorized version of CHEMEQ, an integrator for stiff ordinary differential equations¹⁶. Because of the complexity of the reaction scheme and the large number of computational cells in a two-dimensional calculation, the solution of the chemical rate equations takes a large fraction of the total computational time. A special version of CHEMEQ called TBA was developed to exploit the special hardware features of the CRAY Y-MP vector computer. The current version of TBA is also capable of running in parallel on multiple processors of the CRAY Y-MP.

All of the chemical and physical processes are solved sequentially and then are coupled asymptotically by timestep splitting¹⁷. This modular approach greatly simplifies the model and makes it easier to test and change the model. Individual modules were tested against known analytic and other previously verified numerical solutions. One-dimensional predictions of the complete model were compared to those from the Lagrangian model FLAME1D which has been benchmarked extensively against theory and experiment¹².

A variety of wall boundary conditions can be imposed to determine the effect of heat loss and viscosity on the cellular structures found in the flame. The boundary conditions used in these calculations correspond to an isothermal no-slip wall, where the wall temperature is held fixed at the inlet temperature of the unburnt gas. Because we are interested in the time-dependent dynamics of the flame, these boundary conditions are imposed such that the simulations correspond to a flame propagating in a steady manner in a channel with adiabatic slip walls and at time $t = 0$ enters a region where the walls are isothermal and no-slip.

In this study we restrict our attention to hydrogen flames propagating downward in a flammability tube and near the lean extinction limit. These flames have very low burning velocities which drop further during the extinction process. This makes it difficult to ensure that the flame is always in a well resolved portion of the numerical grid. Two important improvements were made to the numerical model in order to more accurately track these flames near extinction. The first is a switch to a coordinate system attached to the flame; this ensures that the flame is always within the computational domain and also inside the well resolved zone. The second is the use of an automatic regriding scheme. This scheme will concentrate grid points into regions of interest, based on various derivatives of temperature and species concentrations. This tech-

nique can ensure that the flame is well resolved even when it no longer remains planar. In this study, grid adaption is performed only in the flow direction.

III. Results and Discussion

Initial conditions for 2D calculations were taken from 1D calculations giving conditions for steady, propagating flames. Fig. 1 describes the configuration studied, and gives the boundary conditions of the computational domain. Unburnt gas flows in from the left, and reaction products of the flame flow out at the right. If the inlet velocity equals the apparent burning velocity of the flame, the flame is fixed in space yielding a steady solution. Thus, transient effects from ignition can be eliminated, and the one-dimensional solution provides initial conditions for the two-dimensional calculation. The two-dimensional computational domain was a 51 mm channel with no-slip walls held at a constant temperature of 298°K. A 144 x 128 variably spaced grid with finely spaced zones clustered around the flame was used for all the simulations. Previous grid-resolution studies have shown that this resolution is adequate for the problem at hand.

Effect of Wall Heat Losses

In a previous paper⁶, we have discussed the dynamics of downward-propagating flames in a two-dimensional channel with adiabatic walls. In this case, the effect of buoyancy was to suppress the tendency of the mixture to exhibit a cellular structure. Furthermore, the long-term evolution of the flame was a slow oscillation between a slightly convex or concave shape. The overall disc like appearance associated with near-limit downward-propagating flames was not observed in that simulation. This flame is compared in Fig. 2 with a downward-propagating flame in a channel with isothermal walls. The flames are represented by their OH concentrations at a sequence of times. The effect of wall heat losses is to quickly extinguish the flame near the walls and the extinguishment process spreads inwards for a while until a balance is attained with the heat generated by the flame. Even at the early times, the flame is quite flat and both the cellular structures at the center and near the walls disappear by 100 ms. At later times, the flame takes on a shape slightly convex to the flow. Unlike the case without heat losses, no oscillations are seen, even when the calculations are carried out to a longer duration. The overall appearance of the flame is similar to those observed experimentally in near-limit mixtures¹⁸.

Effects of Fuel Concentration

Since the above simulation with heat losses shows that simulations including the effect of wall heat losses can correctly depict the experimentally observed shape of near-limit flames, further simulations were carried out

for 10%, 9.5% and 9% hydrogen - air mixtures. Fig. 3 compares flames in 10%, 9.5% and 9% hydrogen - air mixtures. The top set of frames presents temperature contours, the bottom set presents OH mole fraction at 0.24s after initiation. The OH contours provide a useful indication of the strength of the reactions in the flame. The flame can propagate without difficulty in the 10% Hydrogen - air mixture. A high concentration of OH is formed in a thin zone which corresponds to the high temperature gradient region in the flame. The apparent burning velocity of the flame is higher than that of an adiabatic one-dimensional flame in the same mixture. In the 9.5% mixture, the flame initially does burn normally but just shortly after the time shown in the figure, the apparent flame velocity goes to zero and the flame extinguishes. It can be noted in the figure that the OH concentration is much lower and the flame is thicker, indicating a much weaker flame. The last mixture presented in Fig. 3 is 9% hydrogen - air. In a channel with adiabatic walls, a flame can burn in this mixture. However, in the the two-dimensional calculation with heat loss, the flame does not burn. The apparent flame velocity is always negative and very little OH is produced. The low levels of OH in the figure is the residue from the initially adiabatic flame. In this mixture the "flame" is very thick and resembles a thermal wave.

Details of the Extinguishment Process

Since the 9.5% hydrogen-air mixture appears to be at or just past the flammability limit, further studies were made on a 9.75% hydrogen-air mixture, in order to understand the dynamics of the extinguishment process. In Fig. 4, the centerline temperature is shown at a sequence of times for this mixture. If a flame is propagating at a steady rate, the temperature profile at successive times will fall on top of each other because of the coordinate system used which moves with the flame. Indeed for the first 160 ms, the flame appears to be steadily propagating. It then slows down and the temperature profile begins to spread out. After about, 240 ms, the characteristic "flame" profile is lost and the sharp distinction between the burnt and unburnt gases disappears. This is again in agreement with experimental observations.

Next, the cause of this rapid change in the temperature profile is investigated. Fig. 5 takes a detailed look at the 9.75% hydrogen - air mixture. Contours of the mole fraction of water (in white) are superimposed on the OH radical contours. In the first frame, which is at an early time, 0.08 sec., these two indicators depict a "normal" flame. By 0.16 sec., water adjacent to the walls moves ahead of the rest of the flame. By 0.24 sec. these contours clearly show tongues of cooler gas moving down toward the unburnt mixture, in accordance with the experimental observations. At later times, af-

ter 0.24 and 0.32 sec., significant amounts of water can be seen well ahead of the flame. By 0.32 sec. the low levels of OH indicate that the flame has extinguished. When the mole fraction contours of hydrogen radical and water were superposed, again large amounts of water were observed in front of the hydrogen radical contours, and H radical also decreases dramatically as the flame extinguishes.

These observations indicate that products do indeed get in front of the flame and contribute to its extinguishment. From, the above simulations, it appears that heat losses to the walls, first extinguish the flame near the walls and this creates a lower temperature (higher density) region near the walls when compared to the central regions. Buoyancy acts on this difference in densities and can cause the central regions to rise up while the mixture near the walls flows down. This should result in a recirculatory flow pattern near the walls. In Fig 6, the instantaneous streamlines at 320 ms are shown superposed on the water mole-fraction contours. Large recirculation zones are clearly apparent and such a flow pattern is sufficient to move the burnt products into the unburnt mixture ahead of the original flame. One-dimensional calculations confirm that a self-sustained flame does not propagate downward in the diluted fuel-air mixture observed in the two-dimensional simulations.

IV. Summary and Conclusions

The computational results presented here show that a downward propagating flame in an isothermal channel has a flammability limit of around 9.75%. The experimentally observed value is between 9 and 10%. This is in excellent agreement with our results. Also in excellent agreement are the detailed observations of the flame behavior at the point of extinguishment. Indeed, the description of experimental results could apply verbatim to the the simulation of the 9.75% mixture. For example, the flame is first observed to halt its downward propagation and then actually move back into the burnt products. The flame is quenched at the walls and tongues of colder gases, comprised mainly of burnt products flow down the sides. At the same time, there is an upward motion in the gases at the center of the channel, causing the flame to rise up into the burnt products. So the primary conclusion of this work is that detailed numerical simulations that include wall heat losses can adequately simulate the dynamics of the extinguishment process in downward-propagating hydrogen-air flames. Such simulations can now be used to further explore the relative importance of various competing mechanisms that cause the observed behavior.

Further simulations indicate that a flame propagates downward steadily in a 9% mixture in a channel

with adiabatic walls and propagating cellular flames are observed in a zero-gravity environment. Therefore, our detailed simulations indicate that both heat losses and gravity are simultaneously required to cause the observed limit.

Even in the 10% mixture, heat losses to the walls shorten the flame and differential buoyancy should cause the gases near the wall to have a downward motion relative to the gases near the center. This does occur but a figure such as Fig. 6 shows that the water does not get ahead of the flame. Therefore, we conclude that the dilution of the unburnt mixture with the products of combustion is an essential step in the extinguishment process.

Acknowledgements

This work was sponsored by NASA in the Microgravity Sciences Program and by the Office of Naval Research through the Naval Research Laboratory.

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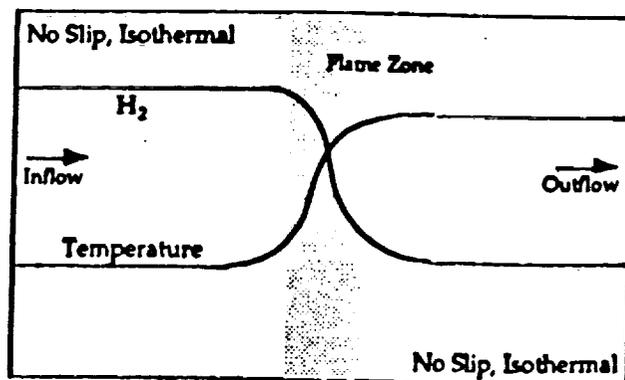


Figure 1. Initial and boundary conditions for the two-dimensional flame calculations.

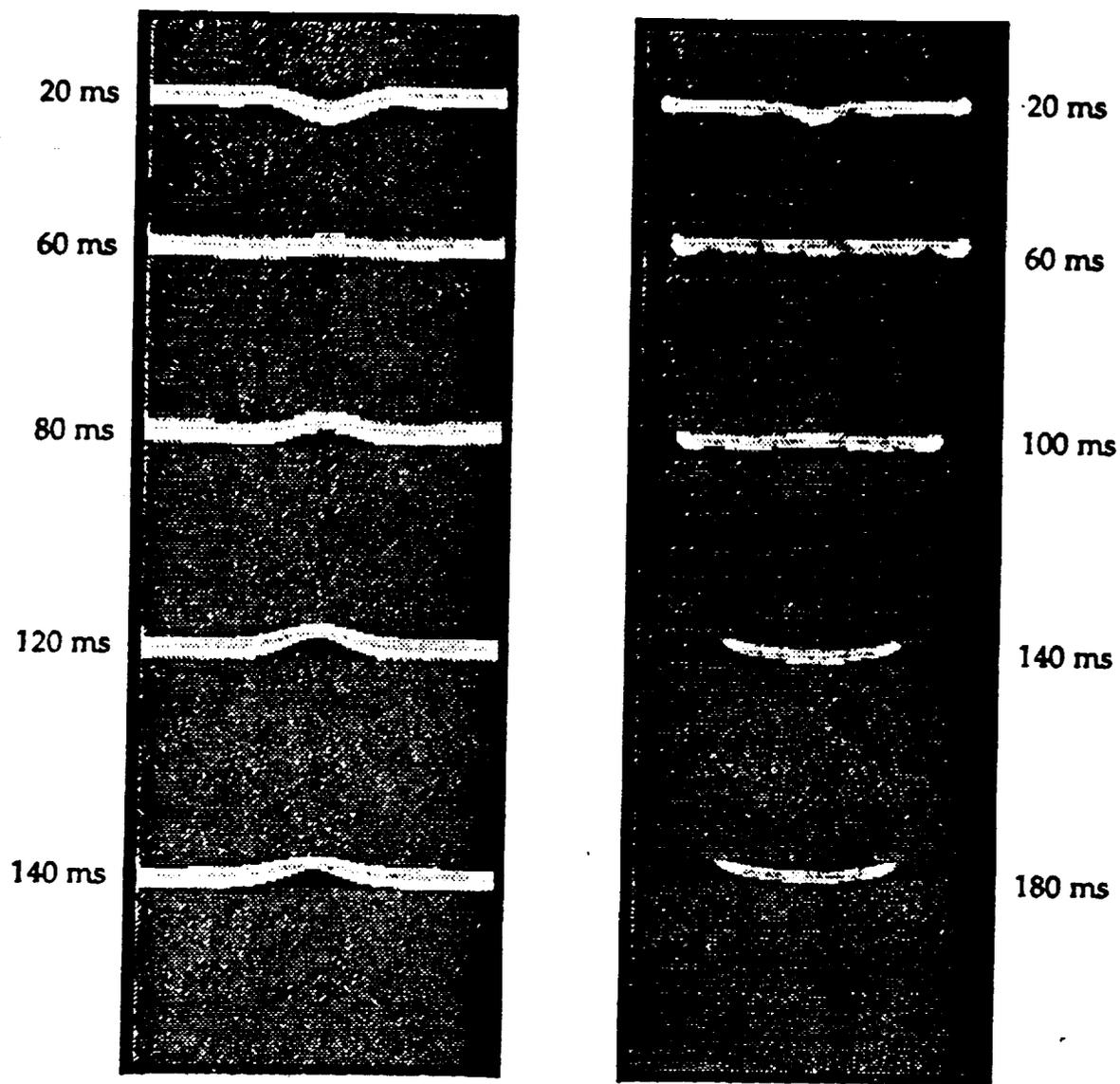


Figure 2. Comparison of flames in 10% H₂ - air mixture propagating downward with heat losses (right) and without losses (left).

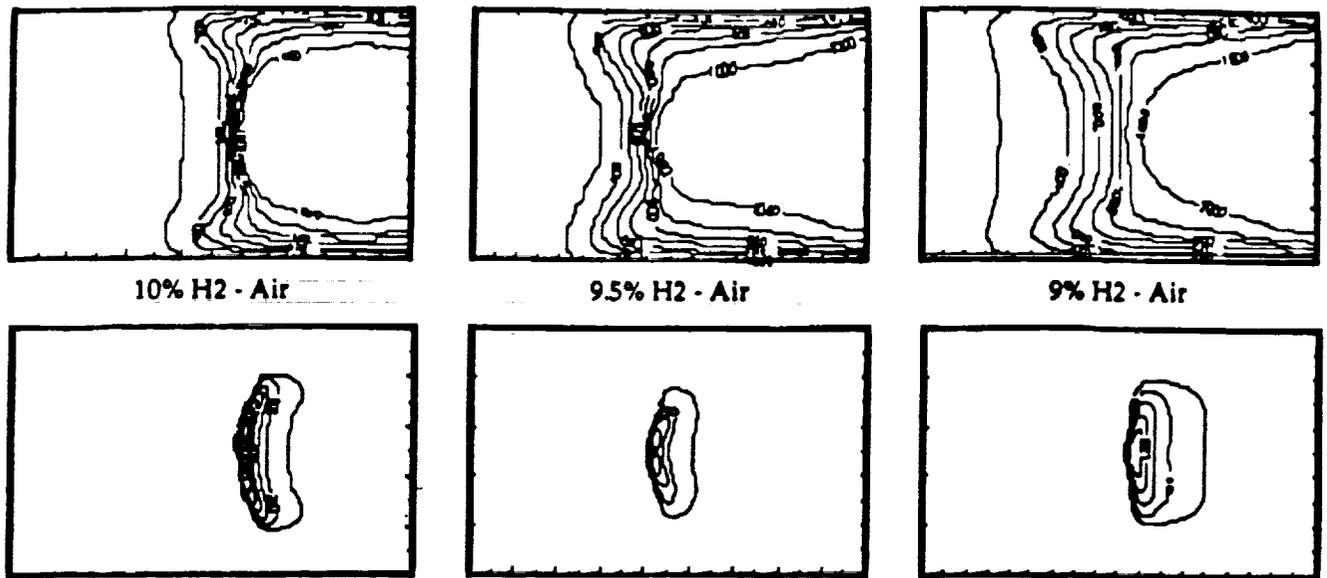


Figure 3. Temperature (top) and OH mole fraction (bottom) in flames in 10%, 9.5%, and 9% H₂ - air mixtures at 0.24 sec. after initiation.

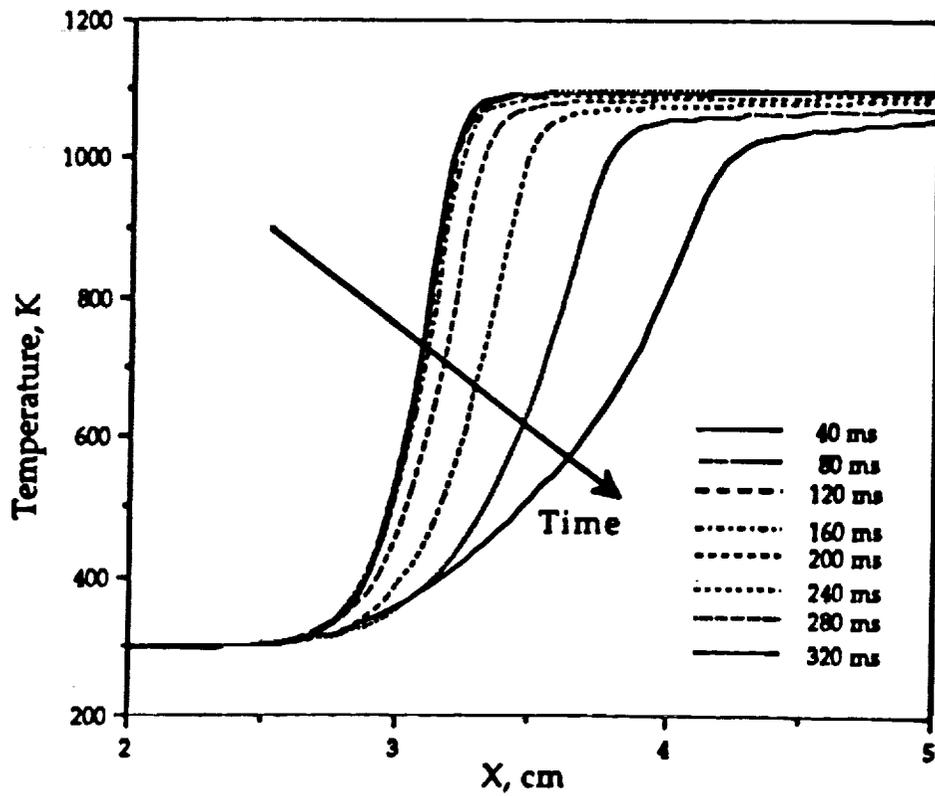


Figure 4. Centerline temperature profiles at various times in 9.75% H₂ - air mixture.

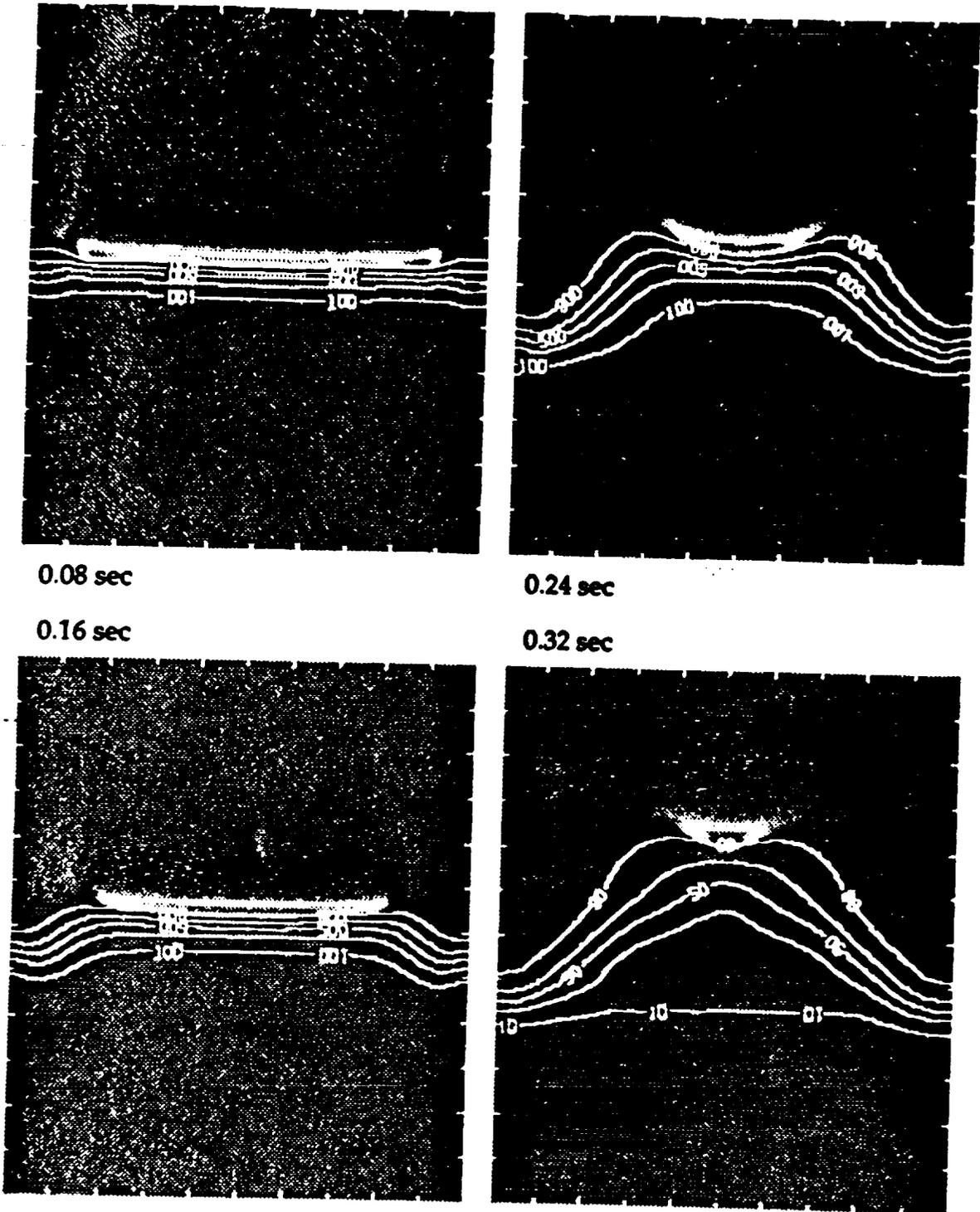


Figure 5. H₂O mole fraction contours superposed on OH mole fraction contours in a 9.75% H₂ - air mixture.

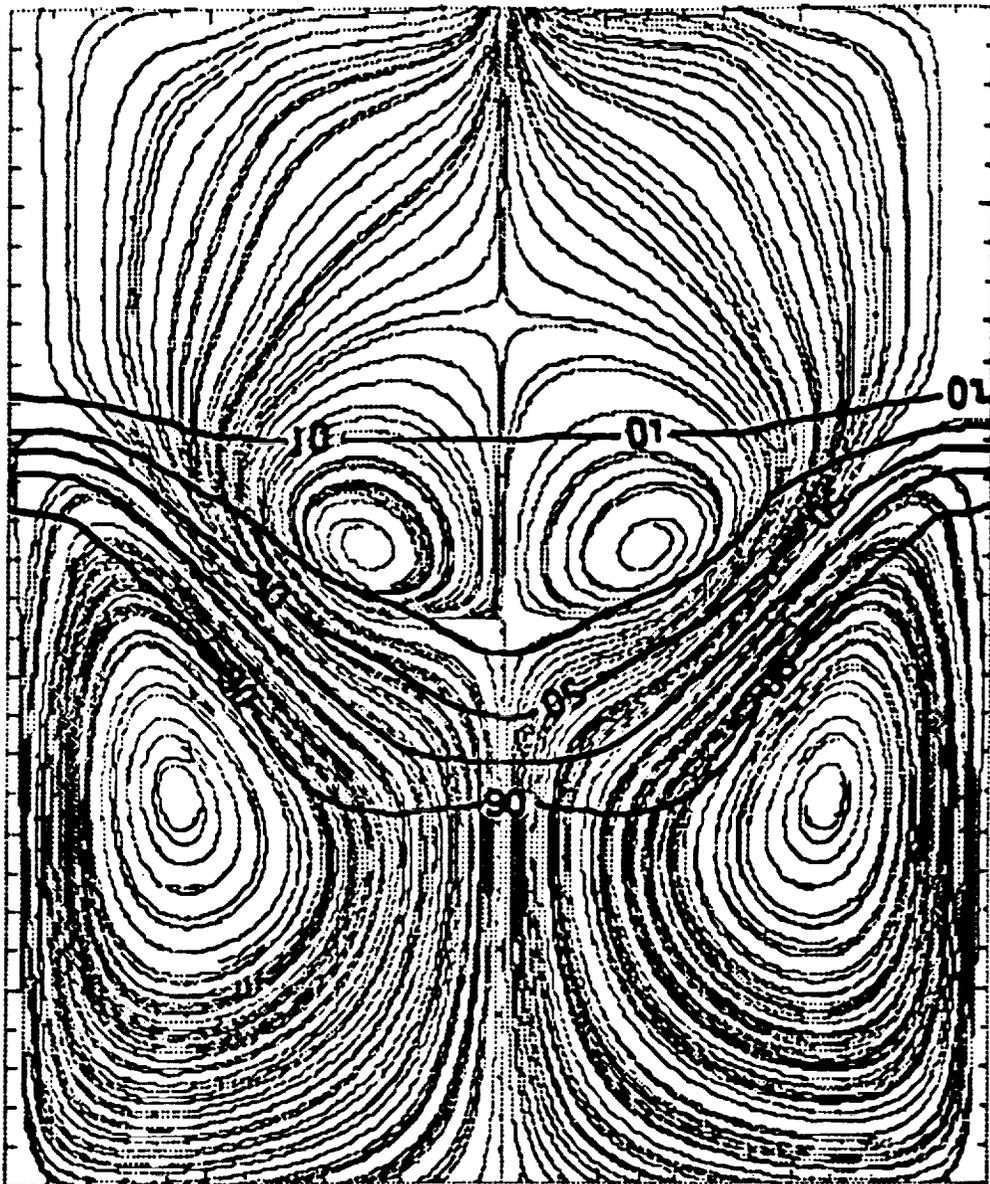


Figure 6. Instantaneous streamlines superposed on H_2O mole fraction contours at late time (0.32 sec) in 9.75% H_2 - air mixture.

Appendix E

Dynamics of Flames near the Rich-Flammability Limit of Hydrogen-Air Mixtures

Dynamics of Flames Near the Rich-Flammability Limit of Hydrogen-Air Mixtures

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Abstract

Flames near the rich-flammability limit of hydrogen-air mixtures are studied using a detailed, time-dependent, one-dimensional Lagrangian model. Results from the numerical simulations indicate that a steady burning velocity is not obtained for very rich hydrogen-air mixtures. As the amount of hydrogen is increased, at first a damped oscillation is observed in the flame and burning velocities, and then with further increase in the amount of hydrogen, an undamped oscillation with a complex set of frequencies is observed. Simulations with a simplified one-step irreversible chemical reaction do not show these oscillations, suggesting that chemical kinetics plays a strong role in inducing these oscillations. Further analysis shows that the oscillations are due to a competition for H atoms between chain branching and chain-terminating reactions. Simulations of spherically expanding flames suggest that stretch effects (due to curvature) will cause the oscillations to occur in less rich mixtures than that observed for planar flames. The implications of these oscillations on the rich-flammability limit as well as the role of chemical kinetics in creating a fundamental flammability limit is discussed.

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Introduction

Flammability limits can be defined as the limits of concentration or pressure beyond which a fuel-oxidiser mixture does not support flame propagation. A difficulty with this definition is that there are significant variations in the observed limits depending on the experimental set-up as well as the method of observation. After reviewing the early empirical observations of flammability limits, Coward and Jones¹ proposed a "standard" apparatus and procedure for determining the limits. They defined a mixture as flammable if the flame propagates all the way to the top of the tube when ignited at the bottom. If the flame fails before it reaches the top, the mixture is considered nonflammable. Since then, this procedure and apparatus have been used to obtain "standard" flammability limits. However, it must be remembered, that there are mixtures beyond these standard limits which can sustain a flame under specific conditions. In this paper, we investigate the characteristics of flames propagating in hydrogen-air mixtures near the standard rich-limit.

Various explanations for the observed flammability limits have been based on factors such as buoyancy forces, preferential diffusion, flame chemistry, conductive and radiative heat losses, the aerodynamics of burnt gases, and flame stretch²⁻⁶. In practical situations, many of these factors are present simultaneously and may interact with each other, making it very difficult to evaluate the actual role of these individual factors. Numerical simulations provide an ideal way to systematically isolate and evaluate the role of various processes.

In an earlier study on the effects of curvature and dilution on flame propagation^{4,9}, it was shown that a flame can be extinguished with less dilution in one geometry than in another, even in the absence of external heat losses and buoyancy effects. Specifically, we showed that a planar flame can propagate steadily in a dilute mixture which does not support a spherically expanding flame. Because this behavior is related to the effects of flame stretch and Lewis number, it raises the fundamental question of whether there is an extinguishment limit in the absence of stretch effects. This question can be addressed by carefully studying the behavior of planar flames in the absence of external heat losses and gravitational effects. Our preliminary results¹⁰ showed that there is such a limit. However, a more

recent numerical study of lean methane-air flames suggests that there may not be any limit in the absence of heat losses¹¹.

In this paper, we present results of a systematic study of flames in rich hydrogen-air mixtures. First, we simulate the propagation of planar adiabatic-flames in mixtures with different amounts of hydrogen. We observe unsteady flame propagation in certain very rich hydrogen-air mixtures. After investigating the source of these oscillations, we consider the effects of stretch (due to curvature) on the near-limit behavior of flames by simulating spherically symmetric flames. Finally, we discuss the effects of other processes such as heat losses and buoyancy, on the propagation of near-limit flames and flammability limits.

The Numerical Model

The results presented here are derived from numerical simulations using a time-dependent, one-dimensional, Lagrangian model¹² which solves the conservation equations for mass, momentum, energy and individual species densities coupled to equations describing the detailed chemical kinetics reactions, molecular diffusion, thermal conduction and thermal diffusion of the various species involved in hydrogen-air combustion. The model permits a wide variety of geometric, initial, boundary, and time-varying energy input conditions and was specifically developed to study the various time-dependent physical and chemical processes which control flame initiation, propagation and extinction. An adaptive gridding technique is used to ensure adequate numerical resolution and resolve the details of the flame structure.

In the calculations presented below, the model was configured with an open boundary at one end to simulate an unconfined system. Most of the calculations were performed in a planar (Cartesian) geometry and gravitational and non-adiabatic effects were ignored in all calculations. Some of the calculations considered a spherically expanding one-dimensional flame. The initial temperature and pressure of the mixtures considered were 298 K and 1 atm, respectively.

The burning velocity of a flame is usually defined as the flame velocity with respect to the unburned gases ahead of the flame. Therefore, it can be calculated from the flame velocity if we know the velocity of the unburnt gases ahead of the flame. For planar flames the velocity of the unburnt

gases ahead of the flame is constant. Hence the burning velocity can be unambiguously determined as the difference between the flame velocity and the flow velocity ahead of the flame,

$$V_{\text{burn}} = V_{\text{flame}} - V_{\text{fluid}} \quad (1)$$

However, for spherically propagating flames of finite thickness, the fluid velocity attains a maximum within the flame and then decreases ahead of the flame (e.g. see ref. 9). A minimum value for the burning velocity can be calculated by subtracting the maximum fluid velocity from the flame velocity. A definition more appropriate for comparison with the planar case is to consider the fluid velocity at the foot of the flame. A convenient definition for the foot of the flame is the first location where the temperature reaches a value of 300 K. In the results presented here, the burning velocity of a spherical flame has been defined as the difference between the flame velocity and the fluid velocity at the first location where the temperature reaches a value of 300 K.

Results and Discussion

In these calculations, the amount of hydrogen in the mixture was systematically increased and with each concentration of hydrogen, the calculations were carried out until a steadily propagating flame (if any) was observed. The burning velocities were calculated as discussed above. The simulations of planar flames are presented first and then the spherically propagating flames are discussed.

Rich Hydrogen-Air Flames

Fig. 1 shows the flame velocity, fluid velocity and the burning velocity for a 78% hydrogen-air mixture. All the velocities are independent of time and a steady burning velocity of about 30 cm/s is obtained. When the amount of hydrogen is increased to 79%, the burning velocity begins to show very small amplitude oscillations but is essentially constant at about 23 cm/s. However, for a 80% hydrogen-air mixture, well defined oscillations are observed initially for both the flame and fluid velocities as shown in Fig. 2. The oscillations are damped and for longer times (greater than 0.12 s), a steady burning velocity of about 18 cm/s is observed. From the simulations, the adiabatic flame temperature for this steady-state flame is about 975 K.

For a 81% hydrogen-air mixture, undamped oscillations are observed which persist for very long times (Fig. 3). This calculation was carried out to 60,000 timesteps to reach a physical time of nearly 1 sec and more than ten "cycles" of the oscillations are observed. During each cycle, the burning velocity drops from a peak of about 28 cm/s to a trough of about 6 cm/s. When the amount of hydrogen is further increased to 82%, the burning velocity goes nearly to zero following a peak in the burning velocity oscillations. However, after more than half a second, a second peak is observed in the burning velocity indicating that a flame can still propagate in this mixture. The burning velocities for the 80-82 % hydrogen-air mixtures are summarized in Fig. 4. From a comparison of the flames in the three mixtures, it appears that as the amount of hydrogen is increased, the frequency of the oscillations decreases rapidly and near-zero burning velocities can be attained during the oscillations.

Oscillatory Flames in Detailed Simulations

Oscillations in the flame velocity have been observed in previous detailed flame simulations^{11,13}. Carter et. al¹³ noted that they could not obtain a steady burning velocity for mixtures with more than 76% hydrogen and suggested that the onset of an instability may be a precursor of flammability limits. However, there was some question if the numerical resolution used in their studies was adequate. Furthermore, in a previous study¹⁰, we also observed oscillations in the mixtures simulated by Carter et. al¹³, but were able to get rid of the oscillations by sufficiently increasing the numerical resolution. In a more recent study of lean methane flames, Lakshmisha et.al¹¹ observed irregular oscillations in near-limit mixtures but concluded that the oscillations were numerical in origin. Therefore, we did a number of studies to find out if the oscillations we observe are numerical in origin.

The tests we did included varying the spatial and temporal resolutions by upto a factor of three, changing the convergence criteria on the chemistry, varying the ignition source, varying initial conditions, changing the size of the system, changing the rezoning parameters and many more. In all cases the results were perfectly reproducible, indicating that the oscillations are indeed a result of the physics represented by the model and not a numerical artifact.

Oscillatory Flames in Simplified Analyses

Since the oscillations are not numerical in origin, there must be a mechanism involving diffusive, convective or chemical processes or combinations of these processes. The Lewis number (defined as thermal conduction divided by mass diffusion) for the flames in which the oscillations are observed is greater than one. Oscillations have been predicted by theoretical and numerical studies¹⁴⁻¹⁶ when the Lewis number is greater than one and the activation energy of the single-step irreversible reaction used is very large. The physical mechanism causing these oscillations has not yet been clearly identified. It is not thermo-diffusive in nature because the thermo-diffusive instability mechanism is stabilising for these Lewis numbers. To explore if the oscillations we observe is due to the same mechanism, the simulations for the 80% hydrogen-air mixture were repeated using a single-step Arrhenius reaction which gives the correct steady-state burning velocity and flame temperature. The technique of Coffee et al.¹⁷ was used to obtain the parameters for the single-step reaction. Oscillations were not observed in these simulations, suggesting that some aspect of the detailed chemical-kinetic reaction mechanism used in our simulations may be responsible for the oscillations. Further calculations were then done to isolate the reactions which may be responsible for the observed behavior.

Chemical Instabilities

The flame temperature for a 80% mixture is just 975 K. With increases in fuel concentration the temperature decreases further, but it is difficult to define an adiabatic flame temperature for these flames because of the oscillatory behavior. An important set of reactions at these low temperatures is the following in which there is a competition for H atoms between a chain branching and a chain terminating reaction:



At low temperatures (less than 950 K), it is possible that the H atoms are consumed faster in chain terminating reactions than produced in chain branching reactions. This could eventually lead to a situation where the energy released in exothermic reactions is not sufficient to balance the energy

diffused by the transport processes and hence the flame velocity decreases with time. However, over a sufficient period of time, it is possible that enough H atoms accumulate for significant energy release to occur and cause the flame velocity to increase again. Such a series of events can lead to oscillations in the burning velocity. To test if this hypothesis can explain the observed oscillations, a series of simulations were performed in which the importance of the chain terminating reaction (Eqn. 3 above) was systematically varied by modifying its pre-exponential factor. The burning velocities in three such cases are compared in Fig. 5. Either increasing or decreasing the overall chain-termination reaction rate by even a small amount (10%) has a substantial effect on the time history of the burning velocity. When the termination reaction rate is decreased by 10%, the frequency of the oscillations increases and the amplitude decreases. When the termination reaction rate is increased by 10%, the pattern of the oscillations changes and consists of two peaks of different amplitudes. Furthermore, the minimum value attained for the burning velocity is quite low (3 cm/s). This result suggests that increasing the termination reaction rate might have the same effect as increasing the amount of hydrogen in these near-limit mixtures.

To explore this possibility further, the termination reaction rate was doubled. The resulting time history of the burning velocity is compared to the actual time history of the burning velocity in a 82% hydrogen-air mixture (Fig. 6). The striking similarity between the two results confirms the importance of the balance between the chain-branching and chain-terminating reactions in near-limit hydrogen-air mixtures.

If the amount of hydrogen is further increased to 83%, the time between the peaks in the burning velocity increases to more than 2 seconds and the burning velocity is essentially zero between the peaks (Fig. 7). Note that the flame continues to be convected along with the fluid when the burning velocity goes to zero (see the plot of the time history of flame position in Fig. 8). In non-burning cases, the flame position ceases to advance and then recedes with increasing time as thermal conduction diffuses the temperature profile.

In Fig. 9, the time histories of the flame and fluid velocities in a 84% mixture are shown on an expanded scale. In this case, peaks in the burning velocity are not observed and the flame and fluid velocities seem to be

asymptotically approaching each other. When these two velocities become equal, the burning velocity will become zero. The time taken to attain this state depends on the amount of initiation energy added at the beginning of the calculation. Further simulations in which the amount of ignition energy is increased makes the flame propagate for a while longer but it eventually appears to die. If the amount of hydrogen is increased further, a zero burning velocity is attained earlier. These simulations suggest that an intrinsic flammability limit might have been attained.

On Flammability Limits

Several interesting observations can be made from the results presented above. First of all, the calculated limit is beyond the experimentally observed standard rich-flammability limit under normal gravity conditions. There are two major possibilities for the differences between the experimental observations and these calculations. First of all, the calculated limit may depend on the chemical reaction rates used. As noted in Fig. 5, the frequency, amplitude and other characteristics of the oscillations are noticeably affected by even small changes in the termination reaction rate. Furthermore, if large changes are made in the termination reaction rate, the characteristics of the oscillation can be drastically modified. For example, if the termination reaction rate is halved, the oscillations completely disappear in the 81% mixture (see Fig. 10). This does not mean that the oscillations are an artifact of the particular kinetic rates used. With the termination rate halved, the oscillations will appear in a richer mixture. Another example where the termination reaction rate is changed significantly has been shown in Fig. 6. Here, the 81% mixture behaves like a 82% mixture when the termination reaction rate is doubled. That is, when this rate is increased significantly, the chemical instabilities occur in a less-rich mixture. Therefore, we conclude that the observed instabilities are not an artifact of the reaction rates used though the actual limiting mixture will depend on the specific reaction rates used.

Another possibility for the differences noted between the numerical and experimental observations may be due to the effects of gravity, stretch, conductive or radiative heat losses in the experiments or multidimensional effects. As mentioned earlier, these effects have not been included in these numerical simulations. Further simulations in which these effects are sys-

tematically included will provide valuable information on observed flammability limits. As a first step in this direction, the effects of stretch due to curvature have been considered.

Spherically Propagating Flames

Figure 11 shows the time variation of the flame, fluid and burning velocities for a spherically propagating flame in a 80% hydrogen-air mixture. Recall that a planar flame in this mixture exhibited very little oscillations and quickly attained a steady burning velocity of about 18 cm/s. The spherically propagating flame shows stronger oscillations which persist for a longer time than in the planar case. A low burning velocity of about 6 cm/s is attained during the oscillations. Of course, at longer times, the behavior of the spherically propagating flame is similar to that of a planar flame since the effects of curvature diminish with increasing radii. The stretch effect due to curvature can be minimized by increasing the radius of energy deposition and the amount of energy deposited. For the simulation presented in Fig. 11, 18 J of energy was deposited in a Gaussian profile with a constant radius of deposition of 2 cm and over a period of 100 microseconds.

Typical flame, fluid and burning velocities in a 81% hydrogen-air mixture are shown in Fig. 12. Notice that a flame does not propagate for more than 0.3 sec even when 200 J of energy are deposited in a volume with a radius of 4 cm. An initial spike in the velocities is observed followed by a rapid drop in the burning velocity to zero. Other initial conditions tried also did not result in a propagating spherical flame in this mixture. This suggests that a flammability limit is attained in a less rich mixture when stretch effects are included.

Concluding Remarks

The simulations presented in this paper have shown that a steady burning velocity is not attained even for planar flames in very rich hydrogen-air mixtures. The unsteady behavior observed in the flame and burning velocities have been shown to be caused by a competition between a chain-branching and a chain-terminating reaction. These two observations imply that time-accurate numerical simulations with multi-step kinetics are required to computationally study the rich-limit behavior of hydrogen-air

mixtures.

In very rich hydrogen-air mixtures, the burning velocity goes to zero, suggesting a flammability limit determined by chemical kinetics. The details of the observed oscillations and the actual limit depend on the specific values used for the reaction rates. However, the observed features do not depend on the actual values of the reaction rates but require that there be two competing reactions. A similar limit which depends on two competing reactions has recently been reported by Law and Egolfopoulos⁶.

The limiting mixture observed in the planar simulations is outside the experimentally observed flammability limit. Simulations of spherical flames show that stretch effects due to curvature can cause unsteady burning velocities in different mixtures and shift the observed limit to less rich mixtures. Other effects not included in the current simulations such as radiative and conductive losses may also modify the predicted flammability limit. Therefore further simulations are planned to investigate if including small amount of heat losses or other factors will produce the experimentally observed limit. A possibility is that during one of the oscillations, when a very low burning velocity (less than 1 cm/s) is attained, a small amount of heat loss will be sufficient to prevent the flame from further propagation.

Acknowledgements

This work is sponsored by NASA through the Microgravity Science and Applications Program and by the Office of Naval Research through the Naval Research Laboratory. The authors would like to acknowledge many fruitful discussions on flammability limits with Drs. J.P. Boris, E.S. Oran at the Naval Research Laboratory and Profs. C.K. Law and P. Ronney at Princeton University.

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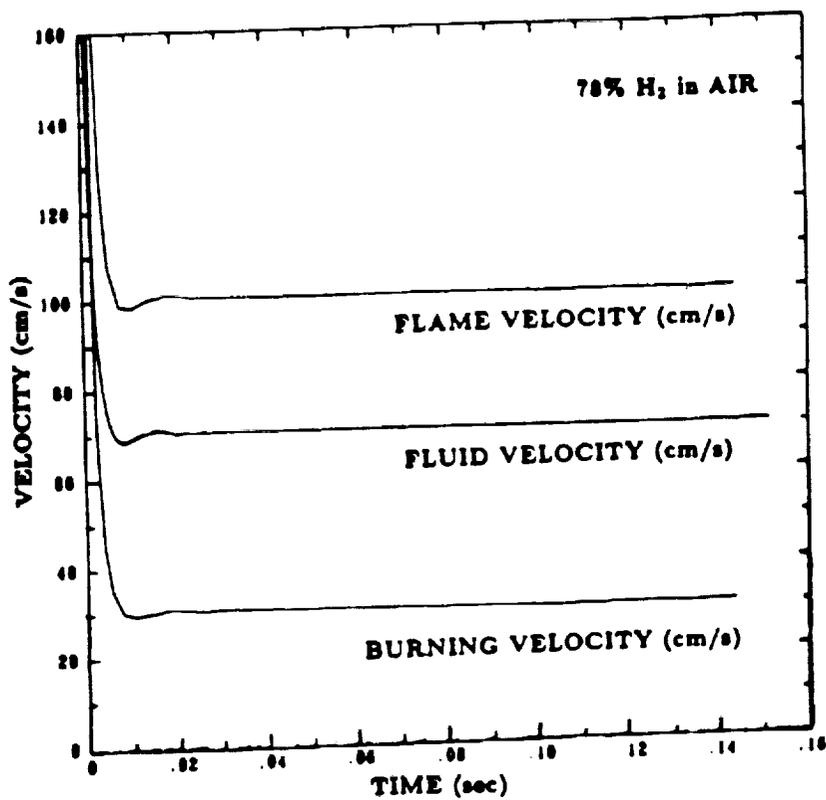
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FIGURES CAPTIONS

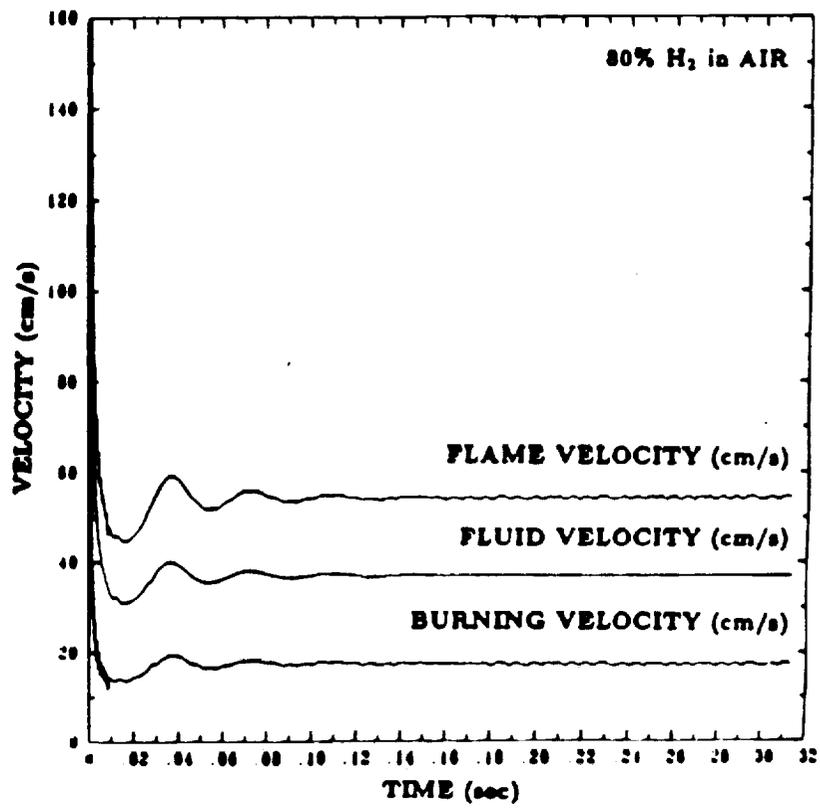
- Fig. 1 Flame velocity, fluid velocity and burning velocity in a 78% Hydrogen-Air mixture.
- Fig. 2 Time evolution of the flame velocity, fluid velocity and burning velocity in a 80% Hydrogen-Air mixture.
- Fig. 3 Time evolution of the flame velocity, fluid velocity and burning velocity in a 81% Hydrogen-Air mixture.
- Fig. 4 Oscillations in the burning velocity of a series of Hydrogen-Air mixtures.
- Fig. 5 The effects of modifying the termination reaction rate in a 81% Hydrogen-Air mixture.
- Fig. 6 Comparison of the time evolution of the burning velocity in a 82% Hydrogen-Air mixture to that in a 81% mixture with the termination reaction rate doubled.
- Fig. 7 Time evolution of the flame velocity, fluid velocity and burning velocity in a 83% Hydrogen-Air mixture.
- Fig. 8 Time history of the position of the flame in a 83% Hydrogen-Air mixture.
- Fig. 9 Time evolution of (a) the flame velocity and (b) the fluid velocity in a 84% Hydrogen-Air mixture.
- Fig. 10 Flame velocity, fluid velocity and burning velocity in a 81% Hydrogen-Air mixture when the termination reaction rate is halved.
- Fig. 11 Time evolution of the flame, fluid and burning velocities of a spherically expanding flame in a 80% Hydrogen-Air mixture.
- Fig. 12 Time evolution of the flame, fluid and burning velocities of a spherically expanding flame in a 81% Hydrogen-Air mixture when 200 J of energy are deposited with a radius of 4 cm and over a time period of 100 microseconds.

Figure 1



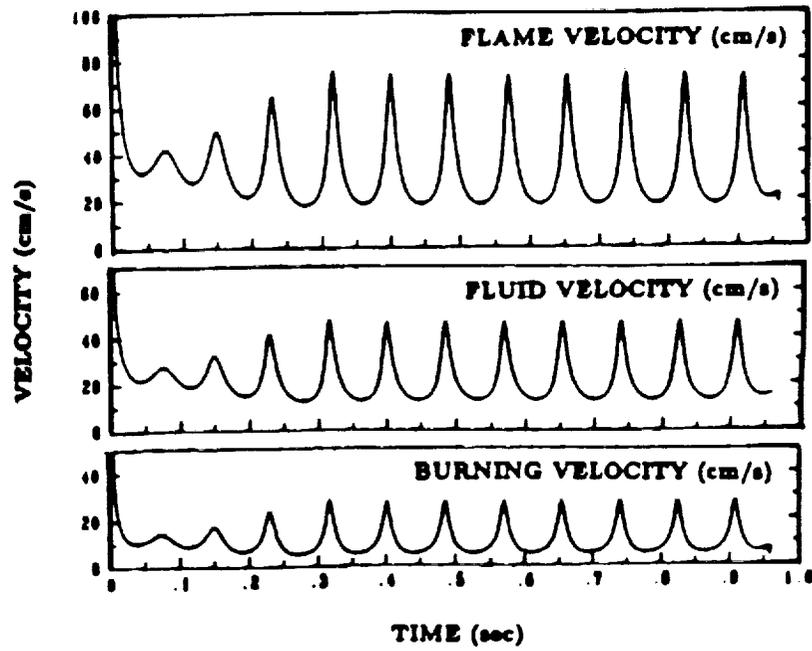
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Figure 2



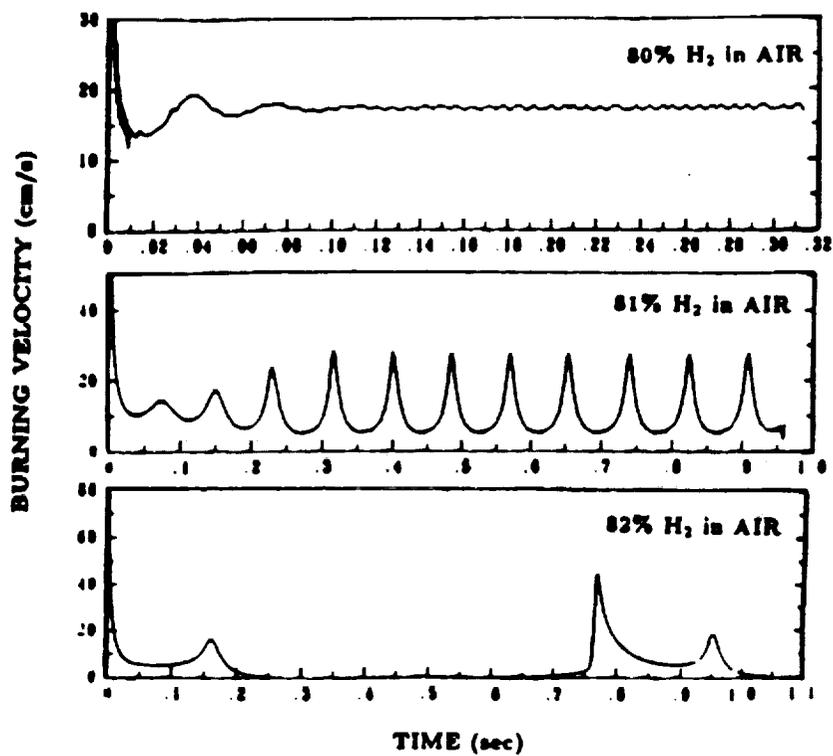
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Figure 3



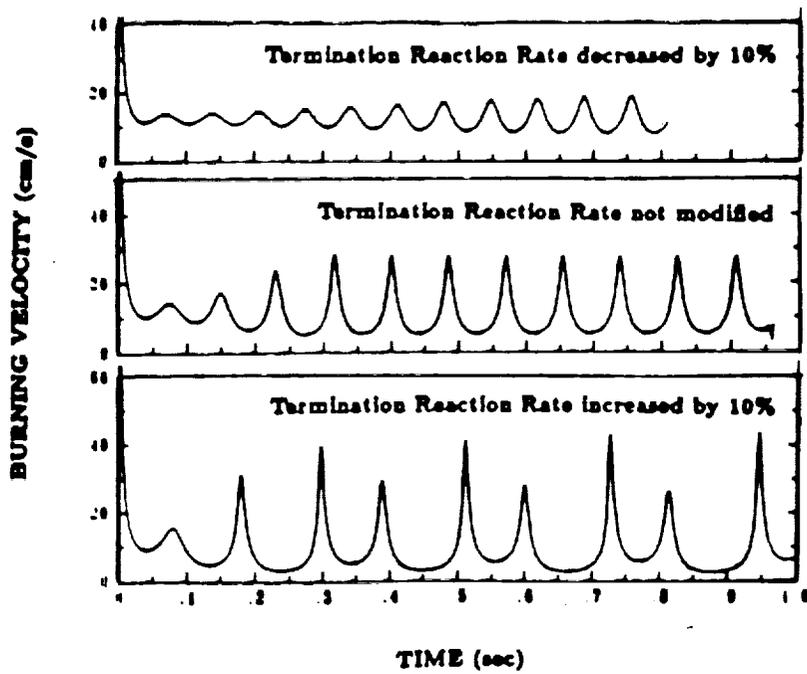
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Figure 4



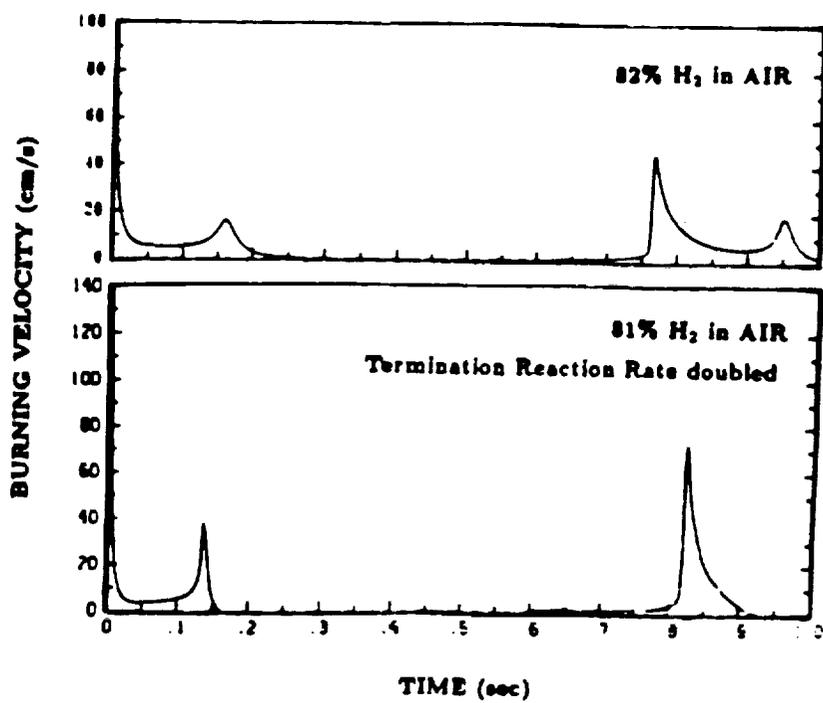
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Figure 5



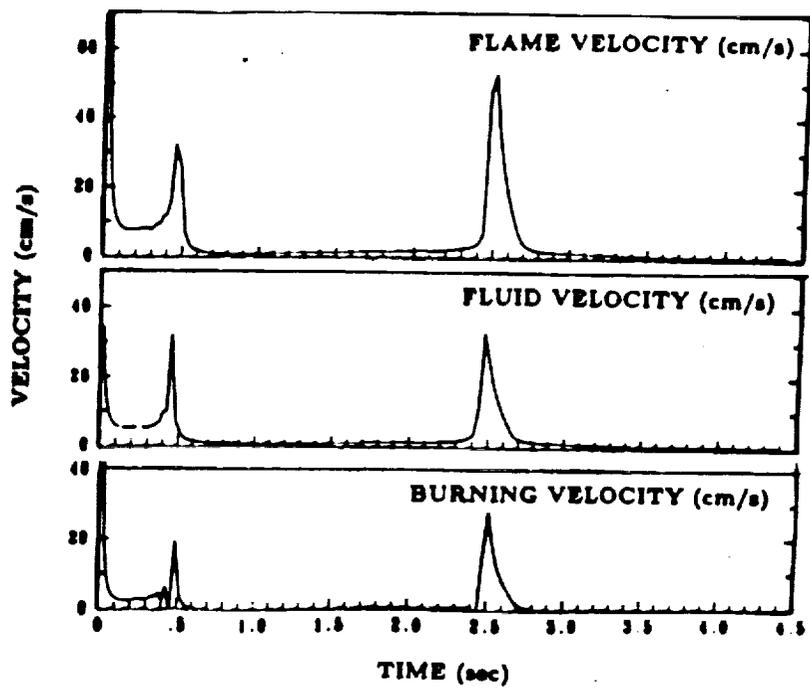
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Figure 6



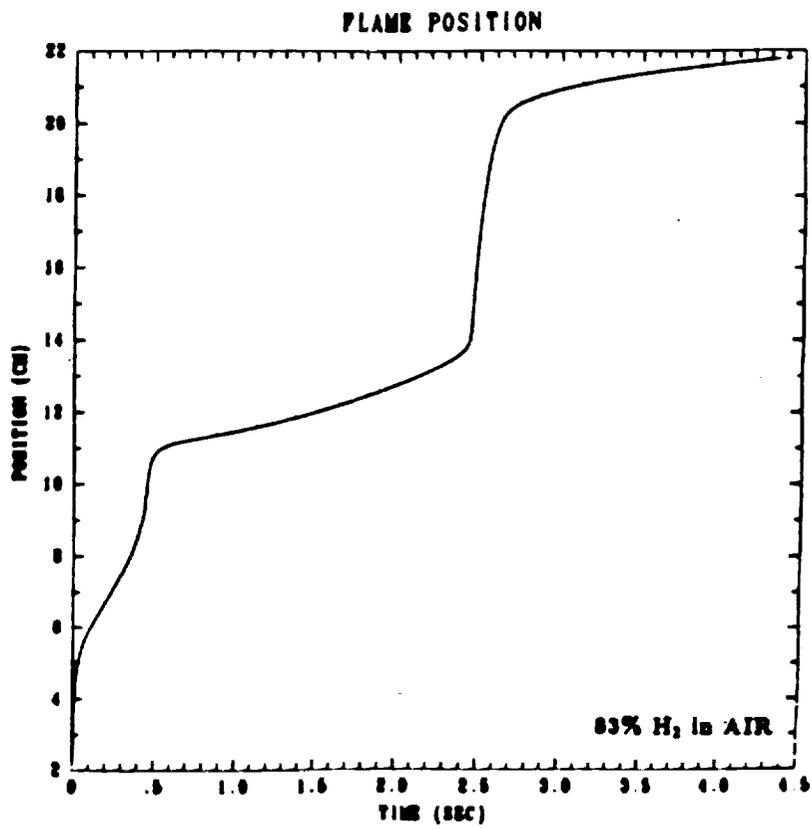
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Figure 7



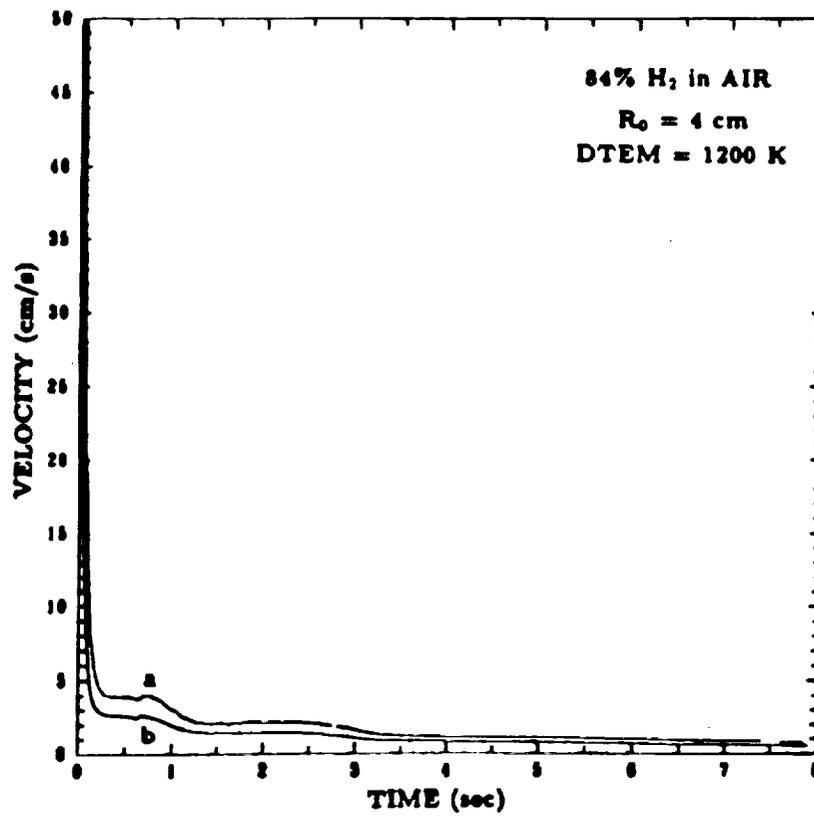
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Figure 8



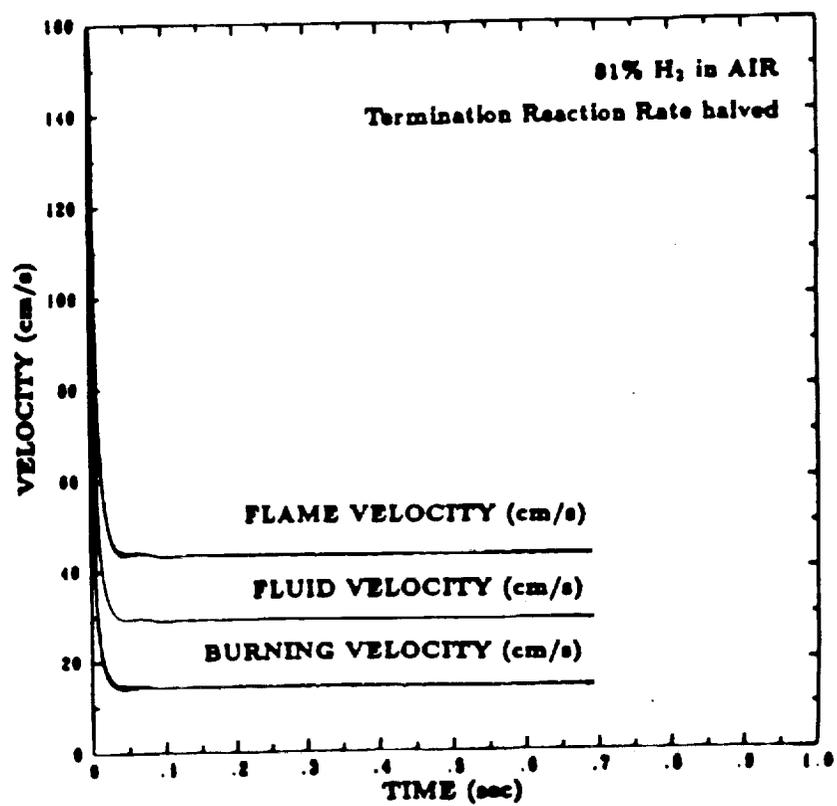
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Figure 9



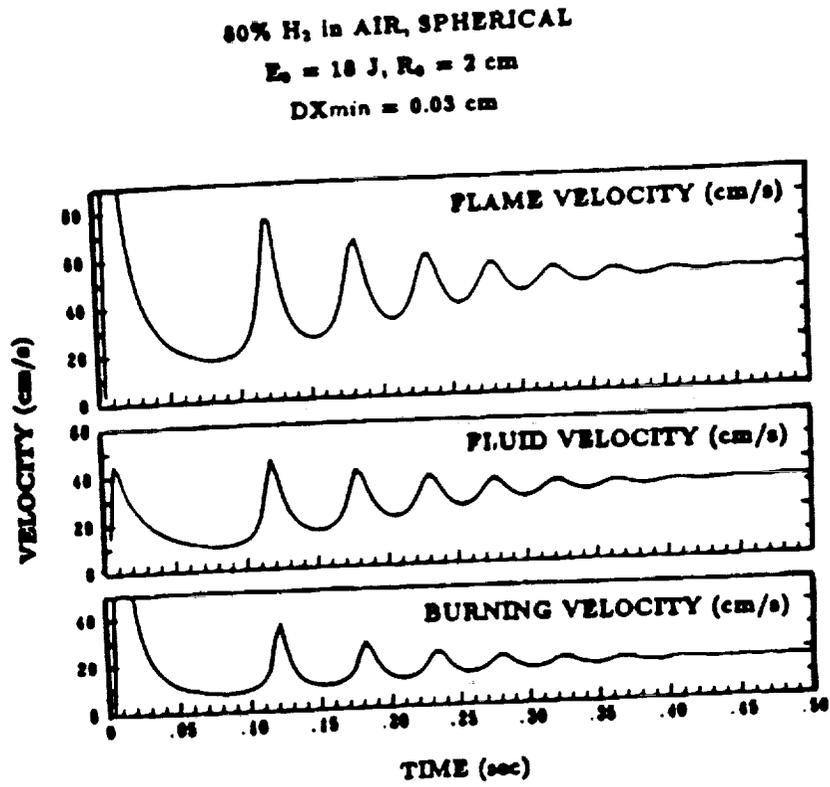
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Figure 10



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Figure 11



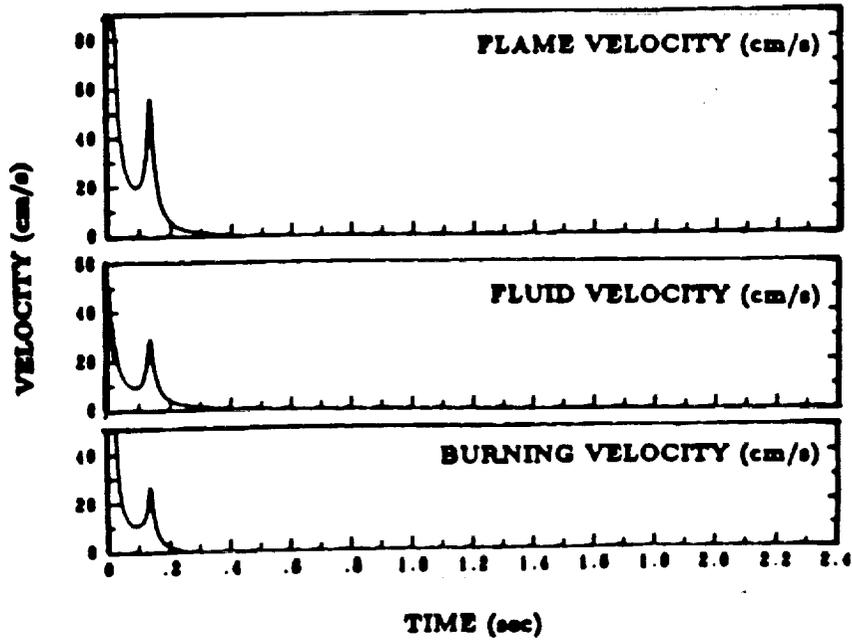
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Figure 12

81% H₂ in AIR, SPHERICAL

$E_0 = 200$ J, $R_0 = 4$ cm

$DX_{min} = 0.025$ cm



REPORT DOCUMENTATION PAGE

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OMB No. 0704-0188

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1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE January 1993	3. REPORT TYPE AND DATES COVERED Final Contractor Report	
4. TITLE AND SUBTITLE Time-Dependent Computational Studies of Premixed Flames in Microgravity		5. FUNDING NUMBERS WU-674-22-05 C-32001-M	
6. AUTHOR(S) K. Kailasanath, G. Patnaik, and E.S. Oran		8. PERFORMING ORGANIZATION REPORT NUMBER E-7517	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) University of Toledo Mechanical Engineering Department Toledo, Ohio 43606		9. SPONSORING/MONITORING AGENCY REPORT NUMBER NASA CR-191051	
8. SPONSORING/MONITORING AGENCY NAMES(S) AND ADDRESS(ES) National Aeronautics and Space Administration Lewis Research Center Cleveland, Ohio 44135-3191		10. SPONSORING/MONITORING AGENCY REPORT NUMBER NASA CR-191051	
11. SUPPLEMENTARY NOTES Project Manager, H.D. Ross, Space Experiments Division, NASA Lewis Research Center, (216) 433-2562.			
12a. DISTRIBUTION/AVAILABILITY STATEMENT Unclassified - Unlimited Subject Category 29		12b. DISTRIBUTION CODE	
13. ABSTRACT (Maximum 200 words) This report describes the research performed at the Center for Reactive Flow and Dynamical Systems in the Laboratory for Computational Physics and Fluid Dynamics, at the Naval Research Laboratory, in support of the NASA Microgravity Science and Applications Program. The primary focus of this research is on investigating fundamental questions concerning the propagation and extinction of premixed flames in earth gravity and in microgravity environments. Our approach is to use detailed time-dependent, multispecies, numerical models as tools to simulate flames in different gravity environments. The models include a detailed chemical kinetics mechanism consisting of elementary reactions among the eight reactive species involved in hydrogen combustion, coupled to algorithms for convection, thermal conduction, viscosity, molecular and thermal diffusion, and external forces. The external force, gravity, can be put in any direction relative to flame propagation and can have a range of values. Recently more advanced wall boundary conditions such as isothermal and no-slip have been added to the model. This enables the simulation of flames propagating in more practical systems than before. We have used the numerical simulations to investigate the effects of heat losses and buoyancy forces on the structure and stability of flames, to help resolve fundamental questions on the existence of flammability limits when there are no external losses or buoyancy forces in the system, to understand the interaction between the various processes leading to flame instabilities and extinguishment, and to study the dynamics of cell formation and splitting. Our studies have been able to bring out the differences between upward- and downward-propagating flames and predict the zero-gravity behavior of these flames. The simulations have also highlighted the dominant role of wall heat losses in the case of downward-propagating flames. Details of the flame-extinguishment process during downward propagation at the flammability limit have been elucidated. The simulations have been able to qualitatively predict the formation of multiple cells and the cessation of cell-splitting. Our studies have also shown that some flames in a microgravity environment can be extinguished due to a chemical instability and without any external losses. However, further simulations are needed to more completely understand upward-propagating and zero-gravity flames as well as to understand the potential effect of radiative heat losses.			
14. SUBJECT TERMS Microgravity; Flammability; Flame; Combustion		15. NUMBER OF PAGES 92	16. PRICE CODE A05
17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT