Effect of Gravity on Flame Instabilities in Premixed Gases

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The effects of gravity on instabilities in laminar, premixed flames in dilute H_2-O_2-N_2 mixtures is investigated using detailed, time-dependent, two-dimensional numerical simulations. The physical processes included in the model are fluid convection, detailed hydrogen-oxygen chemistry, multispecies diffusion, thermal conduction, viscosity, and gravity. The results from the simulations show that the effects of gravity become more important as the burning velocity of a mixture is decreased by either decreasing the amount of fuel or increasing the amount of diluent. In a 1.5:1:10 hydrogen-oxygen-nitrogen mixture with a burning velocity of 9.9 cm/s, gravity plays only a secondary role in determining the multidimensional structure of the flame, and the stability and structure of the flame is controlled primarily by the thermodiffusive instability mechanism. However, in a 1:1:10 hydrogen-oxygen-nitrogen mixture, in which the burning velocity is 2.0 cm /s, gravity is more important. Here the upward-propagating flame is highly curved and evolves into a bubble rising upward in the tube; the zero-gravity flame shows a cellular structure; and the downward-propagating flame oscillates between structures with concave and convex curvatures toward the unburned mixture. These observations are explained on the basis of an interaction between the processes leading to buoyancy-induced Rayleigh-Taylor instability and the thermodiffusive instability. Flames in a 1:1:6.3 mixture, which has essentially the same burning velocity as the 1.5:1:10 mixture, show some effects of gravity, indicating that burning velocity is not the only parameter determining the sensitivity of a mixture to the effects of gravity.

Introduction

INSTABILITIES are often observed in propagating flames. In this paper, we discuss the various types of instabilities that may arise in such premixed flames and use numerical simulations to isolate and study their properties. The emphasis of this paper is on the effects of gravity on flame instabilities in gaseous hydrogen, oxygen, and nitrogen mixtures.

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; convection; and gravity. Such a detailed model allows us to investigate the multidimensional structure of flames and to evaluate the importance of various contributing physical processes. We use the numerical simulations to evaluate the relative importance of these processes in normal Earth gravity and zero-gravity conditions.

The three major instabilities that might occur in premixed flames are 1) the hydrodynamic instability, independently proposed by Landau[1] and Darrieus[2]; 2) the thermodiffusive instability investigated by Barenblatt et al.[3] and Zeldovich[4]; and 3) the buoyancy-induced instability, generally called the Rayleigh-Taylor instability.[5] A numerical model of the premixed hydrogen flame that includes all of the physical mechanisms that lead to these three types of instabilities provides an ideal test bed for studying these instabilities and their interactions.

Hydrodynamic instability was the first kind of flame instability studied theoretically. The analyses of Landau[1] and Darrieus[2] showed that a planar flame, considered as a density discontinuity that propagates normal to itself at a constant speed, is unstable at all wavelengths, with the smaller wavelengths growing faster. Physically, hydrodynamic instabilities are due to the fluid expansion and can be expected to occur in all flames in the absence of stabilizing mechanisms. The actual existence of this mode of instability has never been conclusively shown because of the difficulty in isolating and studying such an idealized flame experimentally. This difficulty in observing hydrodynamically unstable flames suggests that stabilizing mechanisms are important in real flames. For very small wavelengths that are of the order of the flame thickness, the hypothesis that a flame can be treated as a discontinuity is invalid. In fact, the phenomenological analysis of Markstein[6] has shown that introducing a characteristic length of the order of the flame thickness has a stabilizing effect on the short wavelength modes. It might still be possible to see hydrodynamic instabilities for wavelengths much greater than the flame thickness, but stable flames have been observed in systems with the characteristic dimensions even 100 times greater than the flame thickness. This suggests that other effects such as buoyancy, stretch, and heat losses play an important role in the stability or instability of real flames.

The thermodiffusive instability mechanism proposed by Zeldovich,[4] Barenblatt et al.,[3] and Sivashinsky[7,8] involves a competition between mass diffusion of the deficient reactant and diffusion of heat in the mixture. This instability can lead to the formation of cellular flames. This theory assumes an

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abundance of the one component, so that the extent of the reaction is solely controlled by the deficient component. For a simple one-step reaction in an effectively single-reactant system, this mechanism predicts the formation of cellular structure whenever the thermal diffusivity of the mixture is sufficiently smaller than the mass diffusivity of the reactant. For lean hydrogen-air mixtures, 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[9,10] of unstable lean hydrogen-air mixtures and stable rich mixtures. More recent experiments[11,12] have shown cellular flames in some rich and near-stoichiometric hydrogen-air mixtures, contradicting this simple theory. The theory has been extended[13,14] to near-stoichiometric mixtures by considering both the deficient and abundant components.

The interface between a heavy and a light fluid is unstable if the lighter fluid is beneath the heavier one. This is the well-known Rayleigh-Taylor instability, which occurs whenever a denser fluid is accelerated into a lighter one. 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 an instability. In a downward-propagating flame, the light material is on the top and the Rayleigh-Taylor mechanism serves to stabilize the system. The physical mechanisms causing the thermodiffusive 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.[15] Dimensional arguments[16] and theoretical analysis[17] indicate that the importance of the buoyancy-induced instability increases as the burning velocity decreases and, hence, is more important when the mixture is near extinction. Thus, there should be real mixtures in which the thermodiffusive mechanism and the buoyancy-induced mechanism compete.

Cellular structures in flames have been observed in the microgravity experiments in the NASA drop tower[18,19] and in aircraft in ballistic trajectories.[20] These flames in microgravity are free from any buoyancy-induced instability, thus making it possible to examine experimentally only the thermodiffusive instability. Another approach to isolate the thermodiffusive instability for study is with a detailed numerical model of these flames.

Our numerical simulations of premixed flames contain detailed models of the physical processes that cause the various instabilities. Previously, we have used such simulations to show that, in the absence of gravity, cellular structure is caused primarily by the thermodiffusive mechanism.[21] We have shown that this mechanism correctly predicts the observed trends in lean and rich hydrogen-air mixtures. In this paper, we investigate the effect of gravity on flame instability and, in particular, focus on the interactions between the processes leading to thermodiffusive instability and the buoyancy-induced Rayleigh-Taylor instability.

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 extinction[22] 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 ρV, total energy E, and the number densities of individual species n_k, $k = 1, \ldots, n_{sp}$ according to the following:

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

$$\frac{\partial \rho V}{\partial t} + \nabla \cdot (\rho V V) = -\nabla P + F - \nabla \times \mu \nabla \times V + \nabla \left(\tfrac{4}{3} \mu \nabla \cdot V\right)$$

$$\frac{\partial E}{\partial t} + \nabla \cdot (EV) = -\nabla \cdot (PV) + \nabla \cdot (\kappa \nabla T) - \sum_{k=1}^{n_{sp}} \nabla \cdot (n_k h_k V_k) + \sum_{r=1}^{n_r} Q_r$$

$$\frac{\partial n_k}{\partial t} + \nabla \cdot (n_k V) = -\nabla \cdot (n_k V_k) + w_k$$

Here, V is the fluid velocity, P the pressure, μ the coefficient of viscosity, F the body force due to gravity, κ the thermal conductivity of the mixture of gases, h_k the enthalpy of species k, V_k the diffusion velocity of species k, Q_r the heat released from reaction r, and w_k the 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 the ratio of specific heats of the mixture γ 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 Ref. 23.

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) algorithm[24] was developed specifically to solve low-speed flow problems with high accuracy. 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 time-step 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 behavior 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[27,28] 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. In the problem considered in this paper, the time step 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 that is 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 rules.[27] The individual species dif-

fusion velocities are solved for explicitly by applying Fick's law followed by a correction procedure to ensure zero net flux.[28] This procedure is equivalent to using the iterative algorithm DFLUX[29] to second order. This method is substantially faster than one that uses matrix inversions and is well suited for a vector computer. This algorithm is also explicit, and the time-step limit imposed by H-atom diffusion requires that several integration steps need to be made for each overall time step.

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 diluent.[30] This reaction set is solved at each time step with a vectorized version of CHEMEQ, an integrator for stiff ordinary differential equations.[31] Because of the complexity of the reaction scheme and the large number of 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 time-step splitting.[32] 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.[26]

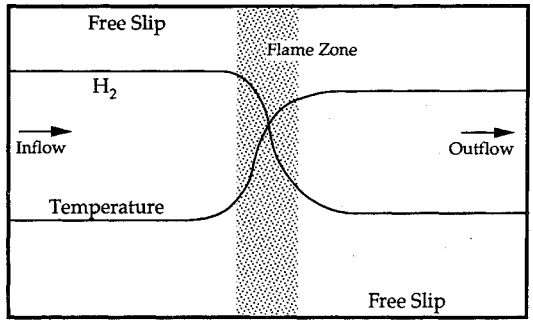


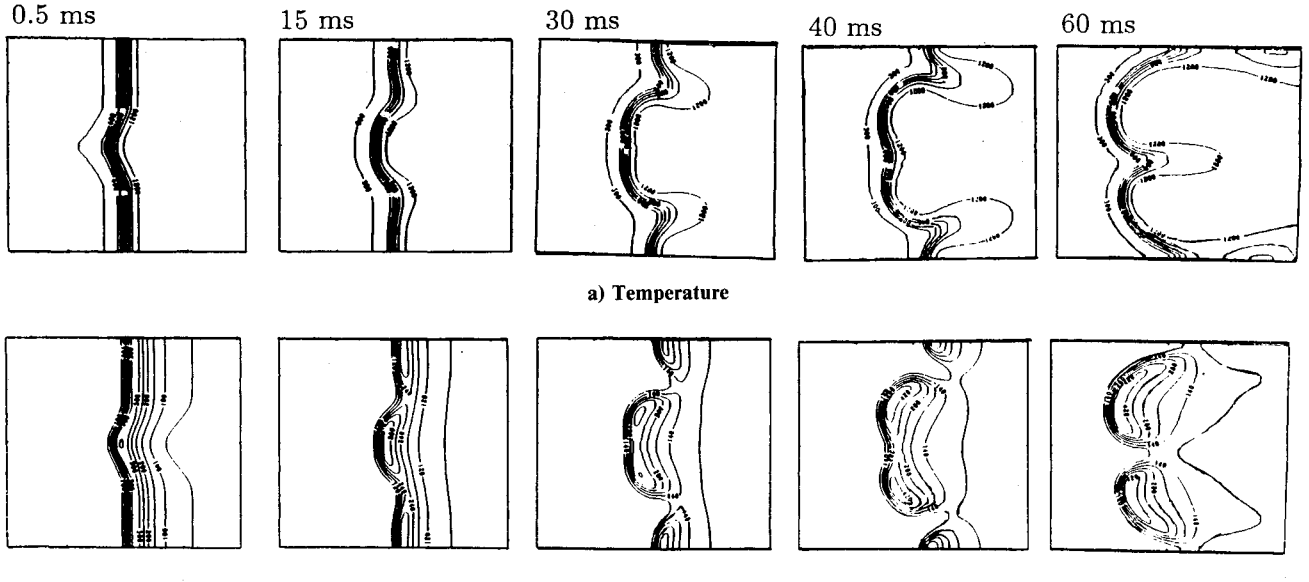
Fig. 1 Initial and boundary conditions for the two-dimensional flame calculations.

Results

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 at 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. This one-dimensional solution was compared to and found in agreement with a similar calculation performed with a totally different, highly detailed flame model, FLAME1D.[26] The computational domain for the two-dimensional calculation was 2.0×4.5 cm, which was resolved by a 56×96 variably spaced grid. Fine zones, 0.36×0.15 mm, were clustered around the flame front.

Flames in an H_2:O_2:N_2/1.5:1:10 Mixture

The first calculation is of a zero-gravity flame in a fuel-lean mixture of hydrogen and oxygen diluted with nitrogen, H_2:O_2:N_2/1.5:1:10; a flame that was multidimensional in the experiments in Earth gravity by Mitani and Williams.[12] The initial condition described by Fig. 1 is perturbed by displacing the center portion of the flame in the direction of the flow. Figure 2a shows isotherms just after the perturbation and then their evolution at subsequent times. The isotherms indicate that the temperature increases in the center portion of the flame, which is convex to the flow and decreases in the the two adjacent concave regions. This indicates that the reaction progresses more vigorously in the convex region, a conclusion corroborated by the intermediate species (OH) number-density contours shown in Fig. 2b. Figures 2 also show that the concentration of OH increases in the convex region and decreases in the concave regions. The burning velocity in the convex region appears to be slightly higher than in a planar region, whereas in the concave regions, the burning velocity has noticeably decreased. We conclude that, in this lean mixture, a planar flame is unstable and a pattern resembling a cellular structure has appeared by the time the calculation was terminated. Furthermore, additional calculations showed that no significant change was seen in the structures that were produced if a different size disturbance was used.



a) Temperature

b) OH concentration

Fig. 2 Time evolution of temperature and OH concentration contours in zero gravity for an H_2:O_2:N_2/1.5:1:10 mixture.

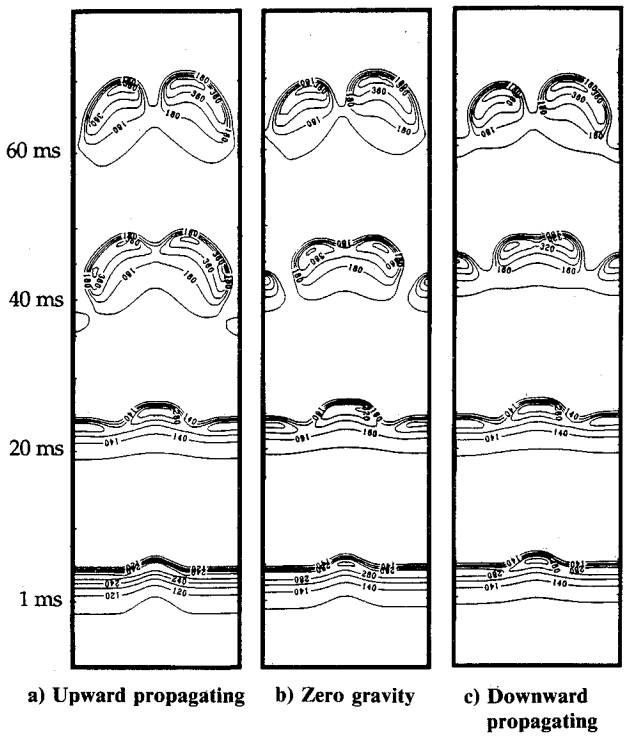


Fig. 3 Concentration contours of OH showing the effect of gravity in an H_2:O_2:N_2/1.5:1:10 mixture.

Figures 3 compare this zero-gravity flame to flames propagating upward and downward in this same mixture, so that the flames are propagating opposite to and in the direction of gravity. Initially (up to 20 ms) all of these flames are similar; they are clearly cellular with noticeable, though not very significant, differences. By 60 ms, the thickness of the two-dimensional flame is affected by gravity, with the upward-propagating flame being thicker than the downward-propagating flame. These figures also indicate that the upward-propagating flame transitions to a cellular flame more rapidly than the downward-propagating flame, and the zero-gravity results are intermediate. These observations suggest that the buoyancy-induced instability enhances the growth of the initial disturbance in the upward-propagating flame and retards the growth in the downward-propagating flame.

The fact that a planar flame is unstable in all three cases and evolves to a cellular structure is consistent with our previous results that show that cellular structure in this mixture is primarily due to a thermodiffusive instability mechanism. Our current calculations show that, in this mixture, the effect of buoyancy through the Rayleigh-Taylor instability is not substantial. In the case of downward propagation, if the mixture were not prone to thermodiffusive instability, we would expect the buoyancy-induced instability to damp the initial disturbance and return the flame to a planar configuration.

The dimensional arguments presented by Williams[16] and the theoretical analysis of Clavin[17] indicate that the buoyancy-induced instability becomes more important as the burning velocity decreases. Though the 1.5:1:10 mixture is lean, it has a burning velocity of 9.9 cm/s. Therefore, we have performed computations for a leaner mixture that has a still lower burning velocity.

Flames in an H_2:O_2:N_2/1:1:10 Mixture

A similar comparison to the one in Figs. 3 has been made in Figs. 4 for flames propagating in an H_2:O_2:N_2/1:1:10 mixture. For this mixture, the differences among the three cases are more dramatic. The zero-gravity case again shows a cellular structure but with a larger cell size than in the previous mixture. At 20 ms, the shape and size of the three flames are comparable. The zero-gravity flame exhibits a seemingly stable cellular structure with little change from 20 to 100 ms. The upward-propagating flame becomes more and more curved and the central portion of the flame moves more rapidly than the sides. In this case, the buoyancy-induced and the thermodiffusive mechanisms are both destabilizing. At 100 ms, the upward-propagating flame exhibits a bubble-like appearance characteristic of some near-limit upward-propagating flames. In downward propagation, buoyancy tends to stabilize and return a perturbed flame to its initial planar configuration. However, at later times, the flame front begins to oscillate around the planar configuration. The fact that early in the calculations, at 20 ms, all three flames appear similar suggests that the thermodiffusive instability is growing more rapidly than the buoyancy-induced instability. By 100 ms, the buoyancy-induced instability has had enough time to strongly interact with the thermodiffusive instability, and in the case of the downward-propagating flame, it essentially counteracts the effects of the thermodiffusive instability.

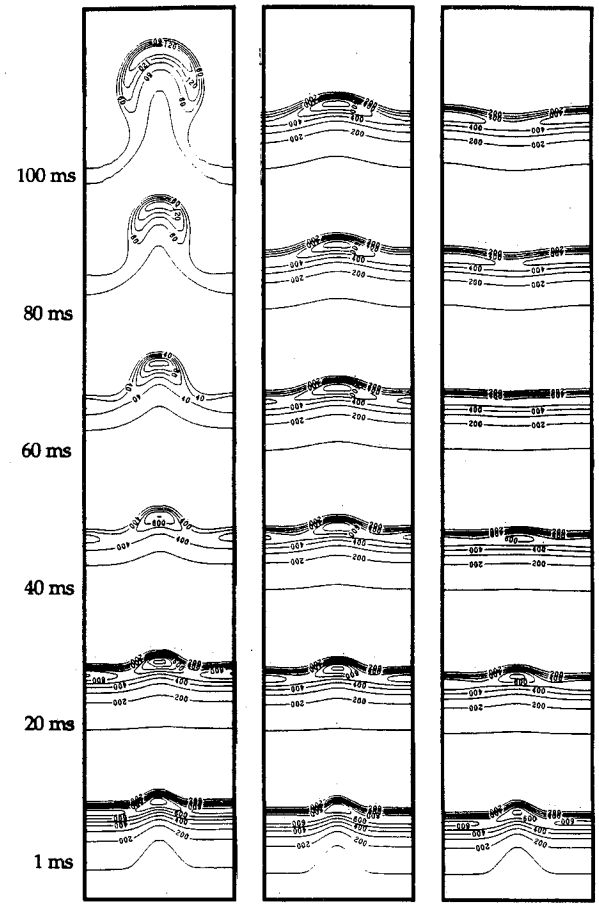
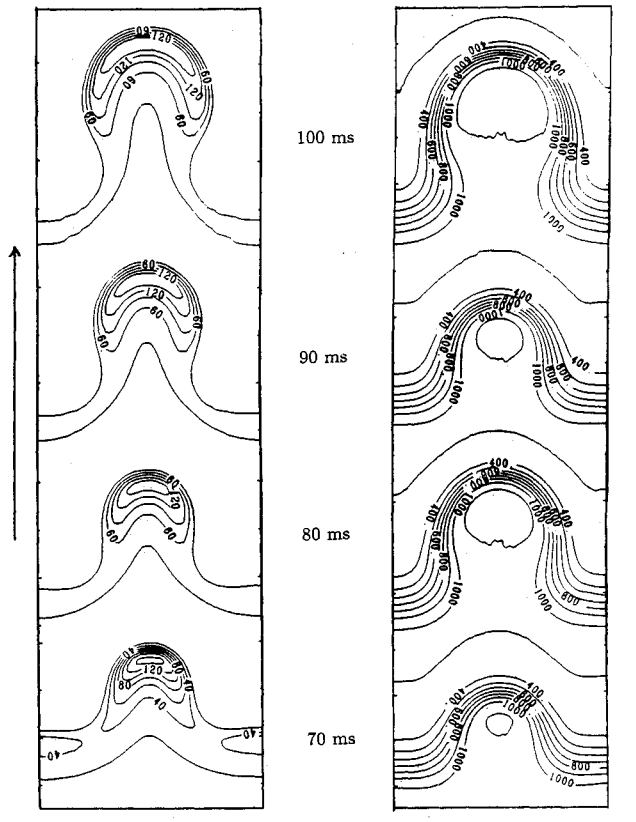


Fig. 4 Concentration contours of OH showing the effect of gravity in an H_2:O_2:N_2/1:1:10 mixture.

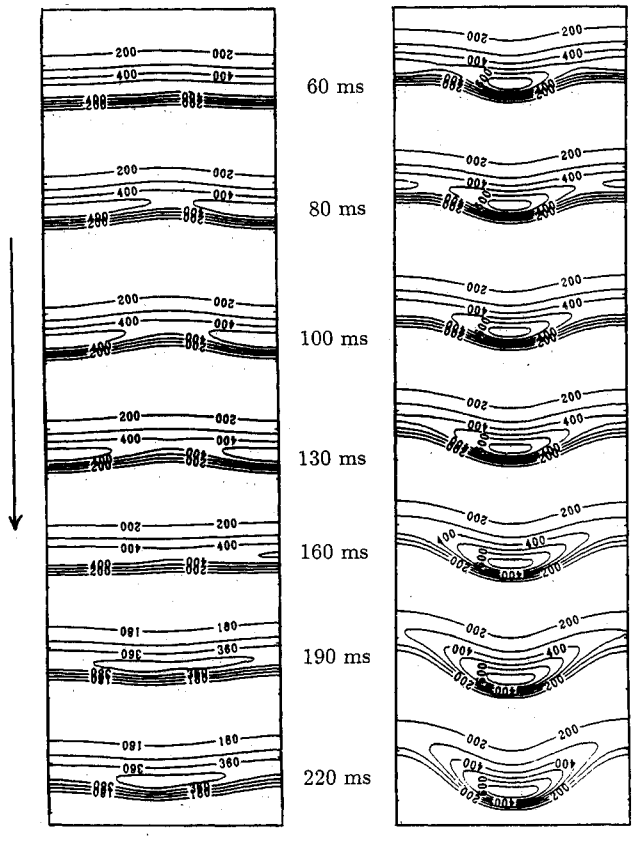
Upward-Propagating Flames

In Figs. 5, temperature and OH concentration profiles are shown at four late times in the evolution of the upward-propagating flame. The reaction zone, as indicated by OH profiles, is more vigorous in the center of the flame, where the initial perturbation was, than near the walls. With time, this difference in the reaction rates increases and the flame appears to be stretched as the central portion moves more rapidly than the sides. The temperature contours substantiate this observation because they show that the overall thickness of the flame is greater at the walls than in the center. For flames in lean premixed hydrogen-oxygen-nitrogen mixtures, a larger flame



a) OH concentration b) Temperatures

Fig. 5 Time evolution of the structure of an upward-propagating flame in an H_2:O_2:N_2/1:1:10 mixture.



a) Downward propagating b) Zero gravity

Fig. 6 Detailed comparison of the late time evolution of the structure of a downward-propagating and zero-gravity flame in an H_2:O_2:N_2/1:1:10 mixture—OH concentration contours.

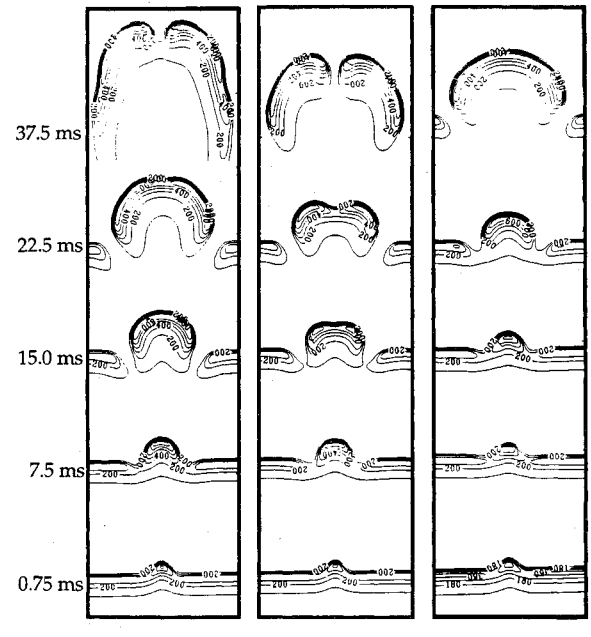
thickness corresponds to a slower flame.[33] With further dilution and perhaps for later times even at this dilution, these results suggest that the central portion of the flame might break away and rise as a bubble. Experimental observations of such a phenomenon has been observed near the lean flammability limit.[34] The effects of heat and radical losses to the walls might also play a role in the actual extinction of these flames.

Downward-Propagating Flames

The long-term evolution of the downward-propagating flame is compared to that of the zero-gravity flame in Figs. 6. The zero-gravity flame is cellular and remains so with time. The downward-propagating flame is nearly planar at 60 ms, develops a concave front toward the unburned mixture by 100 ms, and appears to show a cellular structure again by 200 ms. At an intermediate time (160 ms), it goes through a nearly planar stage. The structure changes from 160 to 220 ms because a planar flame in this mixture is unstable to the thermodiffusive instability. However, as discussed earlier, the buoyancy-induced instability tends to suppress a multidimensional structure for downward-propagating flames. The flame fronts at later times than shown here would probably show the curvature decreasing and the flame becoming nearly planar, then the front becoming concave and the cycle repeating itself. Downward-propagating flames in Earth gravity sometimes oscillate near the flammability limits,[35] a behavior characteristic of waves in fluids stabilized by buoyancy.

Flames in an H_2:O_2:N_2/1:1:6.3 Mixture

Dimensional arguments and theoretical analyses[16,17] have shown that the burning velocity is an important parameter in determining the role of buoyancy. A one-dimensional flame propagating in zero gravity in an H_2:O_2:N_2/1:1:6.3 mixture has a burning velocity of 10.0 cm/s, which is almost the same as in a 1.5:1:10 mixture. At the same time, this mixture has the same equivalence ratio as the 1:1:10 mixture. A comparison of flames in this mixture with those in the other two mixtures will determine whether the burning velocity or the equivalence ratio plays the more important role in determining the flame



a) Upward propagating b) Zero gravity c) Downward propagating

Fig. 7 Comparison of the structures of upward-propagating, zero-gravity, and downward-propagating flames in an H_2:O_2:N_2/1:1:6.3 mixture—OH concentration contours.

shape. Figures 7 compare the structure of the flame in the 1:1:6.3 mixture when propagating upward, downward, and in zero gravity. These figures should be compared to Figs. 3 and 4, which depict flames in 1.5:1:10 and 1:1:10 mixtures, respectively. These calculations show that flames in the 1:1:6.3 mixture do not resemble flames in either of the other two mixtures exactly. The structures in this mixture evolve much more quickly than in either of the other mixtures, and the simulation was ended at 37.5 ms. The structure and dynamics of the flames in the 1:1:6.3 mixture are completely different from those in the 1:1:10 mixture, but some resemblance can be seen to the upward-propagating and zero-gravity flames in the 1.5:1:10 mixture. However, in the downward-propagating flame in the 1:1:6.3 mixture, no cell splitting is observed, unlike in the 1.5:1:10 mixture. Thus, it can be concluded that gravity affects flames in this mixture more strongly than in the 1.5:1:10 mixture. From these comparisons, it appears that the equivalence ratio plays a lesser role than the burning velocity. However, the burning velocity alone does not determine the flame shape and other factors, such as mass diffusion and heat conduction, still remain important.

Linear Stability Analysis

Significant progress has been made in recent years in the theoretical analysis of the dynamics of premixed flames,[17] making it possible to obtain predictions of the stability of the flames for the cases investigated in this paper and qualitatively compare them to the predictions from the numerical simulations. Quantitative comparisons between our numerical calculations and the theoretical predictions are not yet possible because of many simplifying assumptions that have been made to make the theoretical analysis tractable. A theoretical analysis combining the effects of the hydrodynamic, thermodiffusive, and the buoyancy-induced instability has been carried out by Pelecé and Clavin.[35,17] This analysis gives a dispersion relation for the initial growth rate of a disturbance as a function of its wave number. Among the assumptions made in the analysis are that the fuel is converted to products in a single step, the Zeldovich number is high, and the transport properties are constant. Since the simulations used a detailed set of chemical reactions involving multiple species and temperature-dependent transport properties, we have evaluated the growth rate using the transport properties at the initial temperature and for a series of Zeldovich numbers. The details are presented in the Appendix.

In spite of the simplifying assumptions used in the theoretical analysis, many predictions of the theory are in qualitative agreement with those from our detailed numerical simulations. For example, in the 1:1:10 mixture, the theory correctly predicts the oscillatory behavior found in downward propagation and also indicates that the effect of gravity would be stronger than in the 1.5:1:10 mixture. However, the theory incorrectly predicts damped oscillations for downward propagation in the 1.5:1:10 mixture. The disagreement between the two predictions suggests that some of the simplifying assumptions used in the theoretical analysis needs to be looked at more closely. Perhaps, a two-step chemical reaction model or simple temperature dependences for the transport properties may improve the comparisons.

Summary and Conclusions

Detailed two-dimensional numerical simulations of flame instabilities in dilute H_2-O_2-N_2 mixtures have been carried out for three lean mixtures with mixture ratios of 1.5:1:10, 1:1:6.3, and 1:1:10 with burning velocities of 9.9, 10.0, and 2.0 cm /s, respectively. Physical processes included in the model are fluid convection, detailed hydrogen-oxygen chemistry, multispecies diffusion, thermal conduction, viscosity, and gravity. The simulations show the characteristic cellular structure observed in experiments and predicted by theory. The thermodiffusive instability was found to be present in all of the mixtures studied and at both orientations of gravity. However, presence of the buoyancy-induced instability can alter the structure of the flame. The effect of buoyancy was found to be small in the 1.5:1:10 mixture, though its effect on the 1:1:10 mixture was more dramatic. In this leaner mixture, the upward-propagating flame had the characteristic bubble shape observed experimentally and the downward-propagating flame had oscillations characteristic of the Rayleigh-Taylor instability. These results agree with the theory[17] that indicates that the influence of gravity is greater for lower burning velocities. The results for this leaner mixture indicate that these instability mechanisms interact in a quite complex manner, and even though one mechanism can mask the other, in certain regimes both can be equally important.

Flames in the 1:1:6.3 mixture have the same burning velocity as in the 1.5:1:10 mixture and do show some similarities. However, the effect of gravity is more important in the former mixture indicating that the burning velocity alone does not determine the sensitivity of a mixture to the effects of gravity. There are significant differences between the 1:1:6.3 mixture and the 1:1:10 mixture, leading to the conclusion that the equivalence ratio is less important than the burning velocity in determining the flame structure.

An evaluation of the theoretical dispersion relation of Pelecé and Clavin, which predicts the growth rate of instabilities, was carried out at conditions approximating those in our numerical calculations. Some qualitative agreement was observed: the theory predicts oscillatory behavior in downward propagating flames and that the effect of gravity is more marked in the leaner mixture. However, other theoretical predictions disagreed with the results from the numerical calculations. These discrepancies may arise from the sensitivity of the dispersion relation to input parameters or from limitations of the theoretical analysis due to the many simplifying assumptions. Considering the differences in the theory and simulations, the qualitative agreement observed in many cases is considered encouraging.

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 considered systematically in further calculations. These numerical calculations will be extremely expensive and will require tens of hours on a CRAY supercomputer. Thus, the test matrix of cases must be considered carefully, and a well-developed theory can help select the interesting cases. Microgravity experiments can also provide criteria for this selection.

The actual cellular structures are three dimensional. It is possible that the evolution of these structures are different than in two dimensions. Fully three-dimensional calculations will be needed to investigate this. The algorithms used in this study will extend to three dimensions in a straightforward manner. These three-dimensional calculations, if carried out to the same level of detail as the calculations presented here, will require hundreds of hours on today's supercomputers. Appropriate simplifications in the numerical model and significant advances in supercomputer technology are needed for doing affordable three-dimensional simulations of the structure of cellular flames.

Appendix: Theoretical Stability Analysis

We have evaluated the dispersion relation for the combined effects of the hydrodynamic, thermodiffusive, and buoyancy-induced Rayleigh-Taylor instabilities of flame structure[17] at conditions in our numerical simulations. The relation, expressing the dependence of the growth rate σ to the wave number k of the disturbance, is

$$A(k)\sigma^2 + B(k)\sigma + C(k) = 0$$

Table A1 Growth rates for $k = 2\pi$

E	Upward	Zero G	Downward
		H_2:O_2:N_2/1:1:10	
10	103.0	15.8	$0.9 \pm 100.0i$
20	90.1	15.6	$2.2 \pm 85.8i$
30	85.2	15.5	$2.7 \pm 80.5i$
		H_2:O_2:N_2/1.5:1:10	
10	95.2	52.8	$-14.3 \pm 54.7i$
20	83.1	44.5	$-5.4 \pm 53.5i$
30	78.2	40.6	$2.4 \pm 52.7i$

where

$$A(k) = (2-\gamma) + \gamma\left(\frac{L}{d} - \frac{1}{\gamma}\log\frac{1}{1-\gamma}\right)\frac{\rho_b D_{th}}{\rho_u u L}\,k$$

$$B(k) = 2u_L k + \frac{2}{1-\gamma}\left(\frac{L}{d} - \log\frac{1}{1-\gamma}\right)\frac{\rho_b D_{th}}{\rho_u}\,k^2$$

$$C(k) = \frac{\gamma}{1-\gamma}\,u_L^2 k\left[\frac{1}{2}\,k_c - k\left(\delta - \frac{k}{k^*}\right)\right]$$

$$\delta = 1 + \frac{gd}{u_L^2}(1-\gamma)\left(\frac{L}{d} - \frac{1}{\gamma}\log\frac{1}{1-\gamma}\right)$$

$$k^* = \left(1 + \frac{2+\gamma}{\gamma}\frac{L}{d} - \frac{2}{\gamma}\log\frac{1}{1-\gamma}\right)^{-1}\frac{\rho_u u L}{\rho_b D_{th}}$$

$$k_c = \frac{2g(1-\gamma)}{u_L^2}$$

The Markstein length L is given in terms of the flame thickness d by

$$L = \left[Ze\,\frac{(Le-1)}{2} + 1\right]d$$

where Ze is the Zeldovich number, and Le the Lewis number.

The following numerical values were used for the physical properties: $D_{th} = 0.293$; $Le = 0.378$; with $\rho_b/\rho_u = 0.265$, $\gamma = 0.739$, $d = 0.55$ cm, $u_L = 2.0$ cm/s for the 1:1:10 mixture. The values for ρ_b/ρ_u, γ, d, and u_L for the 1.5:1:10 mixture are 0.240, 0.760, 0.35 cm, and 9.9 cm/s, respectively. Three values of the Ze corresponding to activation energy E of 10, 20, 30 kcal/mole were used. This is the only number for which there is no exact correspondence to the numerical model, which includes a full chemical kinetics mechanism.

Table A1 gives the value obtained for σ for $k = 2\pi$, which corresponds to a wavelength of 1 cm. Negative values of σ have not been presented because this mode is expected to decay rapidly.

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