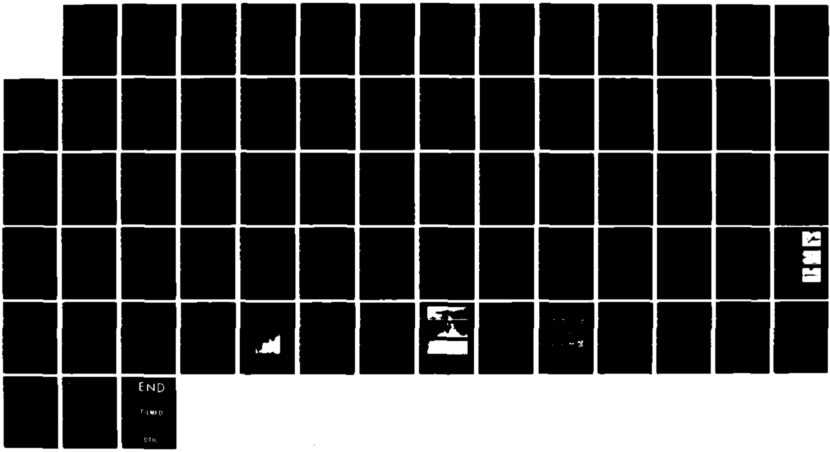
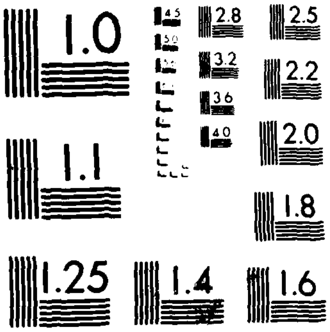
AD-A153 857 CHEMICAL-ACOUSTIC INTERACTIONS IN COMBUSTION SYSTEMS 1/1
(U) NAVAL RESEARCH LAB WASHINGTON DC E S ORAN ET AL.
24 APR 85 NRL-MR-5554

UNCLASSIFIED F/G 21/2 NL





MICROCOPY RESOLUTION TEST CHART
NATIONAL BUREAU OF STANDARDS 1963-A

②

NRL Memorandum Report 5554

Chemical-Acoustic Interactions in Combustion Systems

E. S. ORAN AND J. H. GARDNER

Laboratory for Computational Physics

April 24, 1985

This research was supported by the Office of Naval Research
and the Department of Energy.

NAVAL RESEARCH LABORATORY
Washington, D.C.

SECURITY CLASSIFICATION OF THIS PAGE

REPORT DOCUMENTATION PAGE

1a REPORT SECURITY CLASSIFICATION UNCLASSIFIED	1b RESTRICTIVE MARKINGS
2a SECURITY CLASSIFICATION AUTHORITY	3 DISTRIBUTION / AVAILABILITY OF REPORT Approved for public release; distribution unlimited.
2b DECLASSIFICATION / DOWNGRADING SCHEDULE	
4 PERFORMING ORGANIZATION REPORT NUMBER(S) NRL Memorandum Report 5554	5 MONITORING ORGANIZATION REPORT NUMBER(S)
6a NAME OF PERFORMING ORGANIZATION Naval Research Laboratory 6b OFFICE SYMBOL (If applicable) Code 4040	7a NAME OF MONITORING ORGANIZATION
6c ADDRESS (City, State, and ZIP Code) Washington, DC 20375-5000	7b ADDRESS (City, State, and ZIP Code)
8a NAME OF FUNDING / SPONSORING ORGANIZATION Office of Naval Research 8b OFFICE SYMBOL (If applicable)	9 PROCUREMENT INSTRUMENT IDENTIFICATION NUMBER
8c ADDRESS (City, State, and ZIP Code) Arlington, VA 22217	10 SOURCE OF FUNDING NUMBERS PROGRAM ELEMENT NO 61153N PROJECT NO RR-024-03-01 TASK NO. WORK UNIT ACCESSION NO DN291-086

11 TITLE (Include Security Classification)
Chemical-Acoustic Interactions in Combustion Systems

12 PERSONAL AUTHOR(S)
Oran, E.S. and Gardner, J.H.

13a TYPE OF REPORT	13b TIME COVERED	14 DATE OF REPORT (Year, Month, Day)	15 PAGE COUNT
Interim	FROM ________ TO ________	1985 April 24	70

16 SUPPLEMENTARY NOTATION
This research was supported by the Office of Naval Research and the Department of Energy.

17 COSATI CODES			18 SUBJECT TERMS (Continue on reverse if necessary and identify by block number)
FIELD	GROUP	SUB-GROUP	Acoustic coupling Acoustics Combustion Combustion instabilities

19 ABSTRACT (Continue on reverse if necessary and identify by block number)

A review is presented of chemical-acoustic coupling in terms of its role as a basic interaction which can alter the behavior of combustion systems. Effects resulting from this interaction include sound amplification, changes in sound speed and frequency, sound-induced changes in reaction rates, and acoustic stimulation of chemical oscillation and instabilities. Such effects are important in a variety of problems including combustion instability in jet and rocket engines, the structure of propagating detonations, and turbulence in chemically reacting flows. Background material is presented starting with the early work Lord Raleigh and continuing into a discussion of the relevant properties of sound waves. This leads to a discussion of the influence of energy release on sound waves and the influence of sound waves on chemical reactions. The conclusion is a discussion of chemical-acoustic coupling in combustion environments.

20 DISTRIBUTION / AVAILABILITY OF ABSTRACT ☒ UNCLASSIFIED/UNLIMITED ☐ SAME AS RPT ☐ DTIC USERS	21 ABSTRACT SECURITY CLASSIFICATION UNCLASSIFIED
22a NAME OF RESPONSIBLE INDIVIDUAL E. S. Oran	22b TELEPHONE (Include Area Code) (202) 767-2960 22c. OFFICE SYMBOL Code 4040

DD FORM 1473, 84 MAR 83 APR edition may be used until exhausted
All other editions are obsolete

SECURITY CLASSIFICATION OF THIS PAGE

CONTENTS

CHEMICAL-ACOUSTIC INTERACTIONS IN COMBUSTION SYSTEMS

I. INTRODUCTION

Chemical-acoustic coupling is concerned with the interactions between sound waves and processes involved in chemical reactions. The sound waves may be natural modes of a system, be generated by energy deposited in the system by an external source, or be generated by the chemical reactions within the system. The chemistry may involve changes in number of species or release of energy. The overall effect is a nonlinear coupling of the chemical kinetic processes with the sound waves. This coupling is potentially important in that it can alter chemical reaction times from unperturbed values, generate unwanted frequencies through nonlinear effects which may impede the operation of a system, or change the characteristics of the naturally occurring or imposed sound waves in the system.

This review is concerned with chemical-acoustic coupling as a basic interaction which can alter the behavior of combustion systems. Effects resulting from this interaction include sound amplification, changes in sound speed and frequency, sound-induced changes in reaction rates, and acoustic stimulation of chemical oscillation and instabilities. Such effects are important in a variety of problems including combustion instabilities in jet and rocket engines, the structure of propagating detonations, and turbulence in chemically reacting flows. It is an appealing subject of study because it isolates two interacting effects, well studied on their own and by their own scientific communities.

The earliest studies are by Lord Rayleigh whose criterion explained how an oscillating system would respond to energy input. This work, discussed below in Section III, is a fundamental guide when looking at a complex systems. Many combustion systems show important effects at least partly

---

Manuscript approved January 28, 1985.

attributable to chemical-acoustic interactions. These include, for example, solid rocket motors and ramjets, which are susceptible to combustion instabilities. Such systems, however, are complex geometrically and involve complex flow patterns and combustion processes. Thus to date, most of the research which isolates the chemical-acoustic coupling effects has been theoretical. This is because it is relatively difficult to isolate the interaction physically without bringing into play various dissipative or hydrodynamic processes. Notable exceptions are the experiments described in Section IV. However, the effects predicted by the theory, and verified by those experiments that have been done, show that the coupling process is important and can have a strong effect on a combustion system.

In the review presented below we are emphasizing the physical mechanism and not the mathematical analyses and techniques. References to the original papers should make the derivations easily available. Our goal here is to explain how relevant the timescales of the physical processes can be altered and how new effects can arise due to the coupling. In Section II we introduce the equations describing the interactions of sound waves and chemical reactions. We then proceed to discuss the effects of energy release on sound waves in Section III and the influence of sound waves on chemical reactions in Section IV. We conclude by discussing some applications of these phenomena in combustion environments.

II. PROPERTIES OF SOUND WAVES

A. Basic Equations

We consider first the coupled, inviscid conservation equations

$$\frac{\partial \rho}{\partial t} = -\underline{\nabla} \cdot \rho \, \underline{v} \tag{1}$$

$$\frac{\partial n_j}{\partial t} = -\underline{\nabla} \cdot n_j \underline{V}_j - \underline{\nabla} \cdot n_j \underline{v} + w_j \tag{2}$$

$$\frac{\partial \rho \, \underline{v}}{\partial t} = -\underline{\nabla} \cdot (\rho \underline{v} \underline{v}) - \nabla P \tag{3}$$

$$\frac{\partial E}{\partial t} = -\underline{\nabla} \cdot (\underline{v} E) - \underline{\nabla} \cdot P \underline{v} - \underline{\nabla} \cdot \underline{Q} \tag{4}$$

where ρ, $\rho \underline{v}$, P and E are the density, momentum, pressure, and total energy ($E = \frac{1}{2}\rho v^2 + P/\gamma - 1$), respectively, and $\underline{v}$ is the fluid velocity. The $\{n_i\}$ are number densities of individual reacting species, which are controlled by chemical production and loss terms $\{w_i\}$ and diffusion velocities $\{V_i\}$. Throughout we assume the ideal gas law, so that pressure P and temperature T are related simply by

$$P = \rho R T \tag{5}$$

and the density is related to the $\{n_i\}$ by

$$\rho = \sum_j n_j m_j \tag{6}$$

The heat flux Q, where

$$\underline{Q} = -\lambda\underline{\nabla}T + \sum_j n_j h_j V_j + D(\{V_j\},\{n_j\}), \tag{7}$$

is a function of a number of variables including λ, the thermal conductivity coefficient, $\{h_i\}$ the specific enthalpies, and D, which represents the molecular and thermal species diffusion processes.

These equations have been derived and discussed extensively by Williams (1965) and their numerical solution has been discussed, for example, by Oran and Boris (1981). They are the nonlinear, compressible partial differential equations describing a gas phase combustion system. Depending on the initial and boundary conditions, they represent the time evolution of a reacting, flowing system which can contain, generate and suppress sound waves.

At each point in the fluid, sound waves cause alternate compressions and rarefactions. That they are present in this set of equations can be seen simply (see, e.g. Landau and Lifshitz (1959)) by considering only Equations (1) and (3), and assuming small fluctuations about P, v, and ρ. Below we show how the sound wave equation, including the presence of an energy release term, can be derived from the conservation equations given above.

B. Sound Wave - Energy Release Coupling

The simplest way to explicitly show how sound waves can couple to energy release is to reduce the complexity of the problem from that presented in Equations (1)-(7) and look at a simplified system, as suggested by Toong (1972). Let us we assume that we are dealing with a stationary, ideal, one-dimensional gas with constant specific heats. The gas very slowly releases energy according a reaction rate w, and thermal conduction and diffusion are not important. Then using the relations

$$E = \rho\varepsilon + \frac{1}{2}\rho v^2 \tag{8}$$

$$\rho\varepsilon = H - P \tag{9}$$

$$c_p = \frac{\partial\varepsilon}{\partial T}\Big|_{P,\{\alpha_i\}} \tag{10}$$

where $\{\alpha_i\}$ are the mass fractions of the constituents. Rearranging of terms in Equations (1)-(4) we obtain

$$\frac{\partial\rho}{\partial t} = -\frac{\partial}{\partial x}\rho v \tag{11}$$

$$\rho\frac{\partial v}{\partial t} = -\rho v\frac{\partial v}{\partial x} - \frac{\partial P}{\partial x} \tag{12}$$

$$\rho c_p\frac{\partial T}{\partial t} - \frac{\partial p}{\partial t} = -\rho c_p v\frac{\partial T}{\partial x} + v\frac{\partial p}{\partial x} + wH. \tag{13}$$

To isolate the sound waves in these equations, we expand the quantities into a mean quantity, indicated with a bar, and fluctuating quantity, indicated by a prime:

$$\begin{aligned} T &= \bar{T} + T' \\ \rho &= \bar{\rho} + \rho' \\ v &= \bar{v} + v' \\ w &= \bar{w} + w'. \end{aligned} \tag{14}$$

We also assume that mean quantities are, on the average, spatially uniform, so that when Equation (14) is inserted into Equations (11)-(13), we obtain the linearized equations for the fluctuations

$$\frac{\partial \rho'}{\partial t} = - \bar{\rho} \frac{\partial v'}{\partial x} \tag{15}$$

$$\bar{\rho} \frac{\partial v'}{\partial t} = - \frac{\partial p'}{\partial x} \tag{16}$$

$$\bar{\rho} c_p \frac{\partial T'}{\partial t} + \rho' c_p \frac{\partial \bar{T}}{\partial t} - \frac{\partial p'}{\partial t} = w'H \ . \tag{17}$$

For a perfect gas with constant specific heat, Equations (17) and (15) can be combined to give

$$\frac{\partial p'}{\partial t} + c_s^2 \bar{\rho} \frac{\partial v'}{\partial x} = (\gamma-1)w'H. \tag{18}$$

where c_s^2 is equal to $\gamma\bar{p}/\bar{\rho}$.

In order to relate the unknown fluctuations to one unknown quantity, we introduce the velocity potential ϕ,

$$v' = \frac{\partial \phi}{\partial x} \ , \tag{19}$$

Then Equation (16) becomes

$$p' = -\bar{\rho}\frac{\partial\phi}{\partial t} \tag{20}$$

which when substituted into Equation (18) gives

$$\frac{\partial^2\phi}{\partial t^2} - c_s^2\frac{\partial^2\phi}{\partial x^2} = -\frac{(\gamma-1)}{\bar{\rho}}w'H. \tag{21}$$

This is the standard one-dimensional wave equation on the left hand side and a perturbation term on the right hand side. Now in this simplified equation, the coupling is obvious. It occurs through c_s^2 and through the right hand side of the equation.

We note, however, that much of the interaction has been simplified. In order to reduce the full set of equations to this simple model equation a number of simplifying assumptions had to be made. First a perfect gas with no viscosity or thermal conductivity was assumed. In addition, the mean flow is uniform and the mean density and chemical composition are assumed homogeneous. This restricts the results to either a premixed homogeneous combustion problem or a problem with an external energy deposition source. Perhaps more important is that nonlinear effects of convection and wave steepening have been supressed so that transition to shock waves cannot be treated. However, this formulation has allowed to isolate the coupling of acoustic waves and chemical reactions and to isolate the two fundemental time scales, the chemical timescale and the acoustic timescale.

When the chemical reaction time is long compared to the acoustic time, the chief coupling is through fluctuations in the reaction rate on the right hand side of Equation (21). In the other limit, when the reaction rate is fast compared to the acoustic time, the primary coupling is through changes in the mean sound speed c_s. These limits are discussed below in more detail.

C. Relevant Timescales

Here we define a number of important timescales that will be used thoughout this paper.

First consider

t_s, the period of a sound wave

and the associated frequency, ω_s,

$$\omega_s = \frac{2\pi}{t_s} = \frac{2\pi c_s}{L} \tag{22}$$

where L is the wavelength.

Then there are a number of chemical timescales. We can consider a chemical reaction to take place in two stages: an induction period when radicals build up but there is little energy release, and an energy release period when the largest amount of energy is being released. Consider the chemical time t_c, defined in a static, homogeneous mixture as

$$t_c = t_i + t_{\Delta T}.$$

Then we define

t_i, a static chemical induction time,

in which radicals are built up, but little energy is released (in some cases, the overall reaction could actually be endothermic during this time), and

t_i', a perturbed chemical induction time arising as a result of chemical-acoustic coupling.

In addition we define

$t_{\Delta T}$, a time in which energy is released and the system temperature can rise

and

$t_{\Delta T}'$, a perturbed $t_{\Delta T}$.

These times are shown qualitatively in Figure 1. Depending on the particular temperature, pressure and mixture composition of a reactive system, t_i could be essentially zero or much larger than $t_{\Delta T}$. In general, systems where the energy release term can be written as a single Arrhenius rate have $t_{\Delta T} \gg t_i$.

Another chemical time, t_r, a chemical relaxation time, is often defined in the theoretical analyses referenced throughout this paper. This is related to $t_{\Delta T}$ and is a measure of the comsumption of a reactant when the consumption rate can be written in Arrhenius form:

$$w = K n_r^m e^{-E_a/RT} \tag{23}$$

where m is the order of the reaction, E_a is the activation energy which reflects the temperature dependence, and K is a constant. Then

$$t_r = n_r/w. \tag{24}$$

This is of the same order of magnitude as $t_{\Delta T}$. It is related to the energy release time by the thermodynamic quatities of heat capacity and enthalpy of reaction by

$$t_{\Delta T} = t_r \frac{1}{\gamma} \frac{\rho c_p T}{H} \tag{25}$$

D. The Sound Speed

For the perfect gas described in Section B above, c_s can be evaluated explicitly,

$$c_s^2 = \frac{\gamma p}{\rho}. \tag{26}$$

However, in a chemically reactive medium, evaluating the speed of sound is not so staightforward. An unambiguous definition (Brinkley and Richardson, 1953; Chu, 1958) is

$$c_f^2 = \frac{\partial}{\partial \rho} P(\rho,s,\{\alpha_i\}) \Big|_{\{\alpha_i\},s} \tag{27}$$

where the derivative is evaluated keeping s, the entropy, and $\{\alpha_i\}$, the mass fraction of the constituents, constant. This definition, called the frozen sound speed, gives the velocity of the leading edge of a wave front. However, an equilibrium sound speed, most appropriately used to describe a fast reacting medium, can be defined as

$$c_e^2 = \frac{\partial}{\partial \rho} P(\rho,s,\{\alpha_i(\rho,s)\}) \Big|_s \tag{28}$$

where $\{\alpha_i(\rho,s)\}$ are the equilibrium mass fractions, and the derivative is evaluated along an equilibrium isentrope. In most materials, the frozen sound speed is slightly larger than the equilibrium sound speed.

The applicability of each of these sound speeds has been clarified by Chu (1958, 1970) who showed how, in a reacting medium with finite reaction times, c_s varies from the wave front back in through the wave itself. At the wave front, $c_s = c_f$. Then c_s approaches c_e moving back through the wave front as the reactions are occurring. The value of c_s reaches c_e instantly if the reactions are infinitely fast. Experimentally the distinction between c_e and c_f may be impossible to resolve: for a very fast reaction rate, the size of the disturbance propagating at the frozen sound speed becomes infinitessimally small and all that can be seen is the finite size disturbance associated with the equilibrium front. In the case of very slow reactions, the measured sound speed is the same as the frozen sound speed.

The phase velocity, v_p, is the velocity at which a fixed amplitude of the wave travels. When there are chemical reactions occurring, it is the velocity at which a fixed fraction of completion of the reaction moves. It is important to note that the phase velocity, v_p, of a given part of a reaction wave does not travel at either c_s or c_e, but at some speed in between. This speed is determined by the rate of reaction and is responsible for the dispersive effect of wave propagation in a chemically reactive medium. A reacting medium has different harmonic components travelling at different speeds. Figure 2, based on one by Chu (1970), shows a qualitative plot of v_p as a function of frequency. As $\omega \to 0$, then $v_p \to c_e$. In the other limit, $\omega \to \infty$, and $v_p \to c_f$. One needs to be careful here about these limits: as $\omega \to \infty$ with a finite chemical reaction time, a sound wave travelling at c_f is damped. On the other hand, no damping occurs when ω is fixed and the chemical reaction time goes to infinity.

III. THE INFLUENCE OF ENERGY RELEASE ON SOUND WAVES

An important problem is how the medium through which a sound or pressure wave propagates affects the wave itself. In general, as sound waves propagate through a medium, they can decay due to geometric expansion or energy absorption, damp due to frictional forces, or disperse due to nonequilibrium chemistry. The effects of the background medium on sound waves can be significant. As sound waves propagate through a nonequilibrium medium, or through a medium which becomes nonequilibrium due to interactions with the sound waves, their shape changes in time, producing frequency dispersion. When the amplitude of the wave is large, there are nonlinear effects leading to mode-coupling, which we can call amplitude dispersion. This is essentially the same process by which finite amplitude compressional sound waves steepen into shock waves. Sound waves propagating through a medium may be altered by a competition between both frequency and amplitude dispersion.

In trying to understand these effects it is important to have a good physical understanding for the effects of energy release on sound waves. We first examine the relevant work of Lord Rayleigh who gave the first insights into how energy sources can effect the amplitude and frequency of a sound wave. We then consider the effects of a nonequilibrium background on a sound wave. Finally, we consider the effects of chemical energy release on sound waves.

A. Rayleigh's Criterion

The earliest work on the effects of energy release on sound waves was done by Lord Raleigh (Rayleigh, 1878). Rayleigh observed that periodically adding or taking heat from some location in a gas alters acoustic oscillations in that gas. The type of effect produced depends on the phase of the oscillation at the time when the heat transfer takes place. If heat is added at the moment of highest pressure, or taken from the system at the moment of lowest pressure, the oscillations are amplified. That is, the sound wave amplitude is increased or if there are damping forces, the heat release helps to overcome them. Similarly, if heat is added at the moment of lowest pressure, or taken away at the moment of highest pressure, the oscillation is damped.

Thus when the transfer of heat takes place at the moment of either highest or lowest pressure, the frequency of the sound wave is not affected. However, if the gas is at its normal density at the moment when the heat is transfered, i.e., at a node of the pressure, the amplitude is neither enhanced or decreased, but the frequency is changed. The frequency is raised if heat is added a quarter period before the highest pressure occurs, and the frequency is lowered if heat is added a quarter period after the highest pressure occurs. In general, both kinds of effects are produced by periodic heat input and both the frequency and the amplitude may be changed. This is summarized qualitatively in Figure 3, which is based on one given by Wood (1966).

Rayleigh's criterion was put on solid mathematical grounds by Chu (1956, 1965), who derived a generalized version. In his model he took into account such nonideal effects as viscous losses. In addition to the heat sources considered by Lord Rayleigh, Chu also included the source terms due to body forces and mass addition. When these effects are externally imposed, resonances can occur in an analogous way to putting in heat in phase or out of phase such as shown in Figure 3. Modifications to the criterion are also required when the average flowfield is not zero and when the ratio of specific heats is not constant or the amplitude of the oscillation is not low. Chu (1965) also points out the interesting complication which occurs when the driving functions are fucntions of the flow variables themselves. This is a situation in which the disturbance, the sound waves or pressure waves, can reinforce or damp themselves through the effects of local driving terms. Such a reinforcement could lead to instability.

B. Sound Waves in a Nonequilibrium Medium

Two cases of sound waves propagating through nonequilibrium media have been discussed in the literature. In the first case, the background is assumed to be initially in equilibrium, and this can be driven out of equilibrium by a propagating sound wave. In the second case, the background is not initially in equilibrium, such as a chemically reacting flow or one driven out of equilibrium by an external energy source.

Einstein (1920) and Clarke and McChesney (1964) showed that sound waves are attenuated when they propagate in a dissociating mixture slightly perturbed from equilibrium by a propagating sound wave. Clarke (1978a) shows that if the background is sufficiently out of equilibrium, sound wave amplification is possible. Otherwise, the sound waves are damped. In a review of the theories of propagation of real gases, Lick (1967) considers in detail the effects of various dissipative mechanisms. For the simplest one-dimensional linear acoustic theories for propagation of the wave through a uniform medium, Lick also points out that the common approximation assumes the disturbance is propagated at a constant velocity, the shape of the wave does not change, and the entropy of the fluid does not change as the wave propagates. More realistic approaches modify this by considering the effects of dissipative processes and by the nonlinear effects caused by convection. These nonlinear effects occur because the sound speed, which depends on the local state of the fluid, varies and because the sound waves are convected with the fluid. To properly consider effects of convection, we need a nonlinear theory. Below we summarize the results of a number of authors who have shown that the presence of initial nonequilibrium in the background gas is an essential criterion for a sound wave or a pressure wave to be amplified.

Toong et al. (1965) experimentally observed that sound waves may be amplified or suppressed through their interactions with flames. Clarke (1973, 1974) theoretically investigated the response of diffusion flames to sound waves. Here the acoustic disturbance passes through a laminar diffusion flame with a zone within which the medium is not in equilibrium. The most important effect is the interaction of the sound waves with the chemical reactions, and not with the mass diffusion, viscosity, or heat conduction. We can summarize this work by noting that the flame modelled shows a chemically induced unstable response to the presense of the sound waves. Departure from chemical equilibrium is a prerequisite for instability to occur and the magnitude of the departure depends on the activation energy and the energy of reaction. Sound wave amplification, which takes energy from the reaction process and transfers energy into the sound wave modes, is a consequence of the instability and is a potential mechanism for flame extinction. Clarke also notes that the observations of Melvin et al. (1971) indicate flame extinction at temperatures in line with the predictions of the theory.

Srinivasan and Vincenti (1975) considered the related problem of the interaction of acoustic waves with a gas maintained in a nonequilibrium state by an external energy source. Again, the ambient gas acts like an energy source for the acoustic waves and, depending on the relaxation rates, can amplify them. The work of Bauer and Bass (1973) demonstates this phenomena in terms of an ambient gas maintained at vibrational and radiation nonequilibrium by thermal radiation. This work is similar to the diffusion flame studies mentioned above, although in the flame studies the energy maintaining the disequilibrium is locally provided by the material. It differs from the earlier studies mentioned (Einstein, Clarke and McChesney) which consider nonequilibrium in the ambient material caused by the sound wave. This situation, in contrast, would have a tendency to damp the sound wave. The conclusion from these studies is that a disturbance may be amplified when the nonequilibrium in the background is the result of a source external to the sound wave itself.

C. The Effects of Energy Release on Sound Waves

The problem more relevant to combustion is how sound waves change as they propagate through ambient nonequilibrium, chemically reacting mixtures. The studies of such systems, carried out by Toong and associates, Clarke, Gilbert and associates, and Blythe, are summarized below. An excellent, tutorial review has been given by Clarke (1984).

Toong (1972) used a perturbation method to examine the effects on sound waves when the background medium is undergoing a chemical change described by a power-law expression for the chemical reaction rate. Consider a one-dimensional medium through which travelling sound waves are propagating. Then the whole medium is allowed to react chemically by an irreversible chemical rate characterized by an order, a chemical time, and an activation energy, as in Equation (23). As shown in Equation (21), there are basically two kinds of effects which occur: one type in which there are mean changes in the averaged background state and the other type due to fluctuations in reaction rates. We compare two timescales, the sound wave period, t_s, and the chemical time, t_r, defined in Equation (24). Then when the amplitude of the sound wave is small, the ratio of a characteristic chemical to acoustic time controls which effect is the most important. Figure 4 is a graph of temperature as a function of time for typical cases in the regimes of short and long chemical types compared to the acoustic time. For example, if there are many periods of a sound wave during the time of chemical change, so that $t_r >> t_s$, the main effects are from fluctuations in the chemical rates due to variations in external conditions imposed by the sound waves. Sound wave amplification is predicted for exothermic reactions and sound wave dampening for endothermic reactions. When the reaction rate is independent of temperature, no effect is predicted.

However, if the chemical time scale is very short compared to the sound wave period, $t_r << t_s$, the mean changes due to chemistry are most important. These occur primarily through the time-dependent variations in the sound speed due to the time-dependent temperature vatiations. In this case, just the opposite effects can occur than in the case of fluctuations described above: amplification of sound waves can occur for endothermic reactions and dampening for exothermic reactions.

At intermediate values of the ratio of t_r to t_s, the results also depend on the activation energy of the chemical reactions. When this quantity is small, the results are similar to the effect of constant reaction rate. However, when there is a strong temperature dependence, the effects due to the fluctuations in the reaction rate become pronounced and lead to amplification for exothermic reactions and attentuation for endothermic reactions. The conditions leading to sound wave amplification in exothermic materials are reaction rates increasing with temperature, a reaction order greater than zero, and a large activation energy in the Arrhenius rate. Conversely, a constant chemical rate, order equal to zero, or small activation energy promotes attenuation. Note that when the amplitude of the sound wave becomes large, nonlinear coupling effects become important and much of the classification of possible interactions breaks down.

Figure 5, taken from Toong et al. (1974), shows the calculated variation of acoustic energy during the course of the chemical reaction time for an exothermic system with $t_r >> t_s$. We observe that the energy released tends to go into acoustic energy as the system departs from chemical equilibrium. As the system relaxes back to equilibrium, the acoustic amplifications are damped.

Another important aspect of this problem (see Garris et al., 1975) can be summarized as: once chemical energy is released, not only can the existing acoustic waves be amplified, but other waves are generated which remain in the system and complicate the overall picture. These waves are chemical modes, which mean that when the chemical reaction takes place, there is a spatially non-uniform entropy variation in the system. There is also a reflected acoustic mode, travelling in the opposite direction from the original sound wave. If only acoustic modes were generated, the pressure, density and velocity fluctuations would always be in phase. However, due to the generation of the non-acoustic modes, the fluctuations of the basic quantities in the system are no longer in phase and increases or decreases in amplitude are not necessarily correlated. This would not happen if only sound waves were present. An example of this is shown in Figure 6, where the amplitude of the density, pressure, and velocity fluctuations for cases with long chemical times compared to the acoutic times are shown as a function of time. In each case the pressure amplitude increases with time, but when $\beta = 0$, $m < 2$ (activation energy zero and less than second order reactions) the amplitudes of the density and velocity fluctuations actually decrease. Note also that the density, pressure and velocity fluctuation amplitudes do not vary in phase with each other. Also shown on this figure is a case where the chemical time is not long compared to the acoustic time. In this case there is a noticeable oscillation in the amplitudes, in contrast to the case of a relatively long chemical times.

Later work by Clarke (1977, 1978a and b, 1979, 1981) and Blythe (1979) considered small amplitude disturbances of finite spatial extent propagating through spatially uniform but explosive mixtures. This is in contrast to Toong's work with an infinite wave-train. For a reversible reaction, amplification at the wavehead occurs if the reaction rate in the ambient medium is fast enough. For an irreversible reaction, amplification is unavoidable. In particular, they noted that when a compression wave propagates through an exothermic homogeneous mixture undergoing an Arrhenius chemical reaction, the wave will steepen, and a shock will form before the homogeneous ignition time. However, local ignition can occur before or after the shock formation. In general the steepening of a compression wave in a reacting material will be faster than in an inert material.

Figure 7 (taken from Clarke, 1979) shows the growth of the shock strength with time for a compression wave travelling through a homogeneous reacting mixture and for an inert mixture, both initially in the same state.

As the length of the disturbance is increased, the shock strength grows much more rapidly, but starts later. Also, the time of the local explosion, which occurs some place within the disturbance, is accelerated over the homogeneous explosion time as the result of the reaction. Whether this explosion takes place at the wave front or at the wave peak depends on the the wavelength the propagating disturbance.

Gilbert et al. (1972, 1973) used linearization and WKB techniques to study the effects of fixed wavelength wavetrains in transient chemical reactions. They focussed on the case of $t_r >> t_s$ for a system far from ambient equilibrium. Specifically, they examined the frequency dispersion and amplitude change for a single-step unimolecular decomposition or conversion, $A \rightarrow B$ or $A \rightarrow B + C$, respectively. Figure 8 shows the results of one of their calculations of pressure fluctuations in a cyclopropane system containing a standing wave. As the system decomposes, the most noticeable effect is the 50% increase in the amplitude of the sound wave while the frequency change is only 14%.

The theoretical predictions described above have been tested experimentally by Patureau et al. (1977) and Abouseif et al. (1979). They considered the effects of nonequilibrium exothermic reactions in the ambient medium on sound wave propagation by comparing the acoustic amplitudes before, during and after chemical reactions for a given initial mixture composition, pressure and sound wave frequency. Specifically, they considered mixtures of H_2-Cl_2-Ar in a pyrex tube, through which they passed either travelling waves, standing waves or N-wave packets. Reactions were started photochemically by shining uv radiation on the tube.

The travelling and standing wave experiments tested the prediction of the high frequency limit, which implies long chemical times compared to acoustic times. In this case the predicted pressure varition with time, p', can be written as a function of time, t,

$$p'(t_o + \Delta t) = p'(t_o)\exp\{\frac{1}{2\gamma\, T_o}\frac{dT_o}{dt}\,(m + \frac{\beta(\gamma-1)}{(T_o/T_{o,i})} + \frac{\gamma}{2})\Delta t\} \tag{29}$$

where t_o is the reference time, Δt is some small time period, m is the order of the reaction as defined in Equation (23). T_o is the lowest order unperturbed value of the temperature, $T_{o,i}$ is its initial value prior to any chemical reactions, and β is E_a/RT. Note that there is no frequency dependence in this equation, implying no frequency dependence on the amplification per unit time in this long chemical time limit. Figure 9 compares the results of the experiments to those predicted by Equation (29). Obtaining this curve involved accounting for the natural damping effects and fitting the chemical reaction to an Arrhenius form. The results of the theory and experiments appear in excellent agreement.

More recent experiments by Bass and Detsch (1983) based on analyses by Ellis and Gilbert (1972) show a similar result in the limit of long chemical time or high frequency. It is important to note in comparing these experiments and theories that while the amplification per unit time is constant in the high frequency limit, the amplification per unit cycle is actually frequency dependent. This observation reconciles the apparent discrepancy between the Toong results and the Bass and Detsch results.

The last set of experiments looked at the limit in which the sound wave period and the chemical times were of the same order of magnitude. In this case weak shocks in the form of wave packets consisting of an N-wave followed by weaker trailing waves of lower frequency were passed through the reaction mixture and then the reaction was initiated. Results from these experiments are shown in Figure 10 (from Abouseif et al. (1979)), and it can be seen that amplification occurred just as in the earlier cases. However, here the frequency depence is notable: amplification factors as high as 500% for the trailing waves in the shock experiments are observed. Abouseif et al. (1983) also noted a frequency dependence if the chemical time and the acoutic time become too close.

The most important conclusion from the work of Toong et al., Clarke, Gilbert et al., and Blythe, is that for large values of the chemical time compared to acoustic times, there can be an amplification of the sound waves due to coupling to the energy release process. Some important consequences of this are discussed in Section V.

IV. INFLUENCE OF SOUND WAVES ON CHEMICAL REACTIONS

The presence of sound waves can affect chemical reactions by changing the temperature and pressure and thus locally altering the chemical reaction rates. The work of Clarke (1979), Toong et al. (1974) and Blythe (1979) showed that sound waves tend to accelerate exothermic reactions. Clarke showed that the explosion time is accelerated by compression waves. It is certainly accelerated in the case of shock formation preceeding explosion, since the increased temperature and pressure behind a shock will decrease reaction times and speed up the ignition process. Toong et al. (1974) showed that the presence of fluctuations always accelerates exothermic chemical reactions. We note that this acceleration is a property of the exponential dependence on temperature of the Arrhenius type of reaction. For example, increasing the temperature by ΔT results in more of an increase in the reaction rate than a decrease by ΔT results in a decrease in the rate. Therefore, even though the mean of the temperature or pressure fluctuation is zero, the net effect on the chemical rate is not zero, but positive. This results in an overall acceleration.

Oran and Boris (1981) studied the effects of sound wave perturbations on the chemical induction times of H_2-O_2-Ar mixtures. They combined time-dependent numerical solutions of the compressible conservation equations and a detailed model of the chemical kinetic pathways containing about 50 elementary reaction rates. The first step in analyzing the results was to tabulate the homogeneous unperturbed chemical induction times predicted by the chemical rate mechanism alone, and then to analyze the sensitivity of these times to small perturbations in temperature or pressure. Figure 11 shows typical results of this analysis, which is essentially an estimate of the sensitivity of the induction time to sound wave perturbations. The quantity contoured in Figure 11 is a sensitivity parameter which is defined as the relative rate of change in the chemical induction time due to the change in temperature. A large value of a contour indicates that the induction time of the mixture could change substantially due to the presence of sound waves.

Figure 12 shows some of the results of full numerical simulations in which standing sound waves of given amplitudes and frequencies were imposed on initially homogeneous systems at temperatures that were high enough for reactions to occur but are in different pressure and temperature regimes of Figure 11. For both Figure 12a and 12b, the imposed sound wave was chosen so that there are about three sound wave periods within a chemical induction time. The amplitude of the imposed sound wave was the same in both cases. Each figure shows the temperature versus time profiles at three locations in the mixture. In the upper figure, there is about a fifteen microsecond difference in the induction time generated by the presence of the sound wave. There is a 150 μs difference in the bottom figure, but this case becomes even more interesting when we note that the static induction time for this mixture is about 1500 μs. Thus there has been a total decrease in the induction time of about a factor of ten. Locating these two test cases on the temperature - pressure diagram in Figure 11 indicates that the value of the sensitivity parameter for the upper mixture in is about 10, and for the lower mixture is about 35.

There are several ways to show the effects of the results of chemical-acoustic coupling on the induction time. In Figures 13 and 14 are shown two ways to sumarize the results of full coupled chemical-acoustic calculations. Figure 13 is a measure of how the induction time varies in space due to acoutic fluctuations. For this problem a standing acoustic wave in a homogeneous reactive mixture is modelled and the variation in induction time throughout the mixture is found. $\Delta\tau_{max}$ is defined as the difference between the minimum and maximum induction times occurring within a system perturbed by a standing wave of wavelength L. For example, using the conditions in Figure 12b, $\Delta\tau_{max}$ would be 150 μs. We note that the largest value of the induction time occurring in such a perturbed system may be considerably less than the homogeneous, unperturbed static induction time. The curves of $\Delta\tau_{max}$ are plotted as a function of L/2, equal to one half of the sound wavelength, for several values of u, the maxiumum velocity amplitude associated with the imposed sound wave. These calculations show that there is a maximum value of $\Delta\tau_{max}$. At very short wavelengths, when $t_s << t_i$, phase information on the fluctuations associated with the sound wave averages out. The remaining RMS quadratic effects cause the different portions of the system to react at slightly different times. This enhanced reactivity is a function of the perturbation amplitude, but not the frequency. Thus the curves go to a constant on the left hand side. The negative slope on the right hand side arises when $t_i >> t_s$. The temperature and pressure perturbations across the system are small and vary inversely with t_s. In this case the induction time difference in the system scales linearly with $\Delta\tau_{max}$. Note that resonances occur whenever t_i is an integral or half integral multiple of t_s. This is shown by the oscillations in Δt_{max} just above L/2 = 10 .

Figure 14 (Kailasanath et al., 1984) is another way of looking at the induction time effect. This graph shows the induction time as a function of the amplitude of the impressed sound wave, measured by maximum pertubation velocity induced by the sound wave. The induction time at u = 0 is the static value. First, at small values of u, we note that longer wavelength (lower frequency) perturbations cause ignition earlier than the shorter wavelength (higher frequency) perturbations. The effect is reversed for higher amplitude pertubations. This trend is reasonable when we consider the number of sound wave periods that occurring before ignition. For high amplitude perturbations, the simulations show that ignition occurs after fewer sound wave periods than would fit into a static t_i. This is reasonable since at higher amplitudes there are higher excursions in pressure and temperature and so the ignition time is shortened. For low amplitude pertubations, however, several oscillations of the sound wave can occur before ignition. Now, however, the residence time for the reactive mixture in a perturbed state is also longer. Therefore, for low amplitude perturbations, when more than one sound wave period occurs before ignition, the lower frequency pertubation causes earlier ignition. Thus there is a competition between a residence time effect and a large amplitude perturbation effect, which causes the intersection of the curves. Note that for the hydrogen-oxygen system studied, the long wavelength (low frequency) perturbations are more critical, since for these frequencies smaller amplitude perturbations are sufficient to substantially reduce the ignition time.

The conclusion of this work is that the presence of sound waves can alter the chemical kinetics significantly even before there is any significant energy release. The chemical induction time can be significantly reduced, and the perturbations can cause different parts of the same system to ignite at different times, thus effectively producing hot spots and making the system look inhomogeneous.

V. CHEMICAL-ACOUSTIC COUPLING IN COMBUSTION ENVIRONMENTS

In this section we have briefly described a number of common scenarios in which the physics can be notably affected by chemical-acoustic coupling. This by no means represent a complete list, nor is each subsection meant to be a complete description of the particular combustion environment. We have merely tried to put the effects described above in practical contexts.

A. Pressure Waves Generated by Heat Release

When heat is added to a small volume of the gas in a chamber, the density in that volume is reduced, the volume increases, and sound waves are generated which propagate into the surrounding medium. As we have seen in the material presented above, the physical and chemical properties of the background medium and the amount and time history of the energy deposition determine whether these waves attenuate, amplify, or form into shocks. The early work by Chu (1955) addressed the problem of a local heat addition to a non-reacting, uniform mixture. This could be caused by deposition of laser energy, by a spark discharge, or a local combustion event due to nonhomogeneities in the medium. Chu concentrated on determining the strength of the pressure waves generated and, if they transform to shocks, the strength of the shocks. By considering small amounts of heat addition, he was able to linearize the equation and show an analogy with the problem of two pistons in a tube moving outward from the location of heat addition. Thus he was able to use standard theories to calculate the pressure, density and temperature fields. Some extensions of these ideas to large energy release were possible, but required an exact solution and so were only obtainable in rather idealized geometries.

Clark et al. (1984a, b) have considered shocks generated in confined gases due to rapid heat addition at the boundary. They consider an inert gas, initially in an equilibrium state, between infinite parallel plates and solve the one-dimensional time-dependent compressible equations using perturbation methods. Energy is added at the boundary during a fixed time period which is short compared to the acoustic time (i.e. the wall spacing divided by the equilibrium sound speed.) Conductive heating at the gas layer near the wall induces gas motion from thermal expansion. For moderate input power densities, the result is a weak shock which appears at some distance from the boundary. The strength of this shock is related directly to the duration of initial boundary heating. For greater input powers, a thick, high-temperature, uniform-pressure expanding layer develops which acts like a piston. This can generate a strong shock.

Perhaps the most uncomplicated observation of the direct effects of pressure waves generated by energy release have been in both incident and reflected shock tube experiments. (See, for example, Meyer and Oppenheim (1970), Strehlow and Cohen (1962), or Edwards et al. (1981)). Here ignition starts at or near the contact discontinuity or the reflecting wall. Pressure waves moving into unignited, combustible material produce fluctuations in the material. These waves have little effect in some mixtures, but in others they can accelerate the shock, initiate flames, and generate turbulence. Figure 15 is a streak schlieren photograph of shock ignition of an oxyacetylene mixture taken from Edwards et al. (1981). This figure is effectively a plot of time versus position of the important waves generated and moving through the sytem. An examination of such photographs provides information on ignition centers, reaction waves, and their effects on shock acceleration. Formation of reactive centers such as shown here is characteristic of the mild ignition regime.

An analogous system system has been studied by Kailasanath and Oran (1983), who solved Equations (1)-(4) numerically in one dimension using a detailed multistep kinetics mechanism for hydrogen-oxygen combustion. The system they studied was also in the mild ignition regime, where ignition times are extremely sensitive to fluctuations. Figure 16 shows the results of these simulations which can be qualitatively compared to Figure 15. Reaction centers are formed, and eventually one of these ignites before the others and forms a reaction wave. These reaction centers form because pressure waves resulting from energy release accelerate the shock front and thereby increase the temperature behind the shock. Thus the temperature behind the shock wave increases non-uniformly, if only by a small amount. In the pressure-temperature regime studied here, ignition times are very sensitive to temperature and pressure fluctuations, and a location in the hotter region, away from the contact surface, can start reacting faster than a cooler location closer to the contact surface. Figure 17 shows three such spots, located in terms of increased OH density in the calculation. The last one to form, the one closest to the shock front, is the one that eventually ignites. Sensitivity of the chemistry to fluctuations has caused this spotty ignition pattern.

B. Sound Wave Interactions with Flames

Sound waves interacting with flames may trigger oscillations in the flames, suppress the flames, or amplify the sound waves themselves. For example, the interaction of pressure waves and a flame can create a system of transmitted and reflected waves which reflect from each other and the various walls or obstacles in the system. These waves interact again with the flame and, when conditions are favorable, feedback effects can lead to oscillations of considerable amplitude. As noted earlier, the departure from equilibrium within the flame front is a prerequisite for instability to occur and interactions with sound waves have been observed to cause flame extinction. Gaydon and Wolfhard (1979), in their review of the early work on premixed flames, have said:

> "It is important to realize that the energy imparted to the flame by the sound is very small compared with the energy release in the combustion, and unless the sound can trigger some instability, such as the vortices in the sensitive flames, we should not expect sound to produce any marked change. However, although the energy contribution is small, the amplitude of the movement due to sound is not so small, and there are some interesting changes in flame shape."

They note that the presence of sound waves narrows the region of conditions in which a flame is stable. However, the mechanisms primarily responsible for these effects are not directly connected to the chemical release; they are interactions between physical properties such as the flow field and flame area. Their contention is that the heat release provides the source of energy and defines the flame front. Acoustic-flame interactions in premixed flames have been studied by Putnam and Dennis (1953 a,b), Kaskan (1953), Blackshear (1953), Bailey (1957) and Chu (1956). Good reviews of these and other work are given by Gaydon and Wolfhard (1979) and Putnam (1964b).

More recent studied of acoustic interactions with diffusion flames have been done by Toong et al. (1965) and Clarke (1973). Clarke has shown that, given the right combination of activation energy and heat of reaction, the instabilities alluded to by Gayden and Wolfhard (1979) can occur due to a chemical-acoustic interaction. Toong and associates have attributed the oscillations they see to Tollmien-Schlichting waves. The interaction of sound wave with diffusion or premixed flames is an important practical problem in combustors.

C. Chemical - Acoustic Effects in Detonations

It has been known for some time that a smooth, planar propagating detonation is unstable. This means that small perturbations can trigger the instability and the system may change to a more stable structure (Erpenbeck (1965), Fickett and Wood (1966)). We also know now that detonations do not propagate as smooth fronts. Their equilibrium structure is composed of a set of interacting, intersecting shock waves called the incident shock, the Mach stem, and transverse waves. The intersection of the incident shock and Mach stem is a triple point whose position in time describes a rhombic shape called a "detonation cell." The incident shock is not steady, but is continuously decaying. The transverse wave, a reflected shock intersecting the Mach stem and the incident shock, shuttles back and forth across the detonation front. A detonation cell is reinitiated when collisions occur between transverse waves moving in opposite directions.

Behind the shock fronts, there is a reaction zone which varies in distance from the front depending on the induction and energy release times of the material, chemical-acoustic coupling effects, and the length of time it has been since cell reinitiation. The velocity of the leading shock varies from above the Chapman-Jouguet value to below it and takes a sudden jump when the cell structure is reinitiated by transverse wave collisions. Detonation cells have been measured by coating the inside of a detonation tube with soot and letting the triple point trace out the pattern. In general the pattern is quite irregular, but a characteristic cell size can be determined for a particular material at a given temperature and pressure. Excellent summaries of what is known about the cell structure have been given by Strehlow (1979), Fickett and Davis (1979).

We see that the factors that determine the detonation cell size are complicated interactions of fluid dynamics and chemical kinetics. The fluid dynamics here involves a number of interacting shock waves, pressure waves, sound waves, and perturbations due to energy release. The chemical reactions are occurring in an environment which is always subjected to fluctuations and pressure perturbations. Thus we have gone up in level of complexity in the flow properties from the shock tube problem described above.

Figure 18 shows a series of schlieren photographs by Edwards for detonations propagating down a tube filled with a mixtures of hydrogen, oxygen and argon. These are marginal detonations: that is, a detonation which is close to the values of pressure, temperature, and stoichiometry for which it will die out. Notable in these figures are the ragged pockets of unburned material cut off behind the detonation front. Figure 19 shows two-dimensional numerical simulations of detonations propagating in a similar mixture (Oran et al., 1982). This also predicts the presence of the unburned pockets. A closer look at the predicted temperature and pressures in the pockets indicate that they are in a weak ignition regime for hydrogen-oxygen, i.e., they are very sensitive to fluctuations.

The formation of the pockets provides an extinction mechanism for a detonation. Large, slow-burning pockets would reduce the energy available to drive the detonation front. However, in the weak ignition regime, they could also have other possible influences. Sound waves interacting with the unburned pockets could provide a new source of perturbations, since they could burn considerably faster than would be predicted by the static induction and energy release times of the mixture. This could initiate an instability and thus provide the initial impetus to allow the material to form a new number of detonation cells characteristic of the chamber size. In an open environment where there are no walls, the pockets could provide the perturbation necessary to allow new cells to generate and the number of cells to increase.

D. Reactive Turbulence and Vorticity Generation

Vorticity is a fundamental property of turbulent and transitional flows. It is defined by

$$\underline{\xi} \equiv \underline{\nabla} \times \underline{v} \tag{30}$$

and its generation occurs according to

$$\frac{d\xi}{dt} = \frac{\underline{\nabla}\rho \times \underline{\nabla}P}{\rho^2} \tag{31}$$

Thus whenever there is a misalignment in the pressure and density gradients, vorticity is generated. Since the presence of density gradients is a characteristic of reacting systems, and pressure waves or fluctuations are generally present, rotations in the flow are generated through the mechanism described by Equation (31).

As an example of the interaction of reacting systems and sound waves in which vorticity generation is a major factor, consider a spherically expanding flame in which the pressure is constant and the density increases from the center outward. The interaction of a plane travelling sound wave with this flame would take place on several levels. First, there is the generation of vorticity by the interation of

$$\underline{\nabla}\rho(\text{flame}) \times \underline{\nabla}P(\text{soundwave}) \tag{32}$$

Through the first half-period of the sound wave, vorticity is generated and rotational velocities are created in the region of the flame. However, this is not cancelled by the second half-period of the sound wave, which now operates on a velocity field slightly altered from the initial conditions.

Thus the cumulative effect of many oscillations is to create vorticity. For wavelengths very much longer than the diameter of the flame, the flame feels time-varying pressure gradients. For short wavelengths, there is still an effect due to the cumulative variations. This timescale analysis is similar to the energy release - sound wave interaction.

Superimposed on the sound wave - density gradient interactions are the interactions of chemical kinetics and energy release with the sound waves. We know that energy release and induction times can be decreased, and sound waves can be amplified. From Equation (31) we see that the sound wave amplification increases the strength of the vorticity generated. Increased energy release rates speed up the flame expansion rate. The overall effect, then, could be to enhance energy release, create vorticity structure, and thus generate a turbulent flow from a laminar system.

The scenario investigated by Markstein (1958) is similar to the one described above: a nearly spherical flame interacts with a weak shock. This could be considered as a situation in which the sound wavelength is infinite. Figure 20 is a schlieren taken in a stoichiometric n-butane-air mixture. As the shock passes through the flame, the upper flame boundary becomes compressed by the shock (0.10 ms). Later (0.40 ms), a vortex ring has formed, and the enhanced flow at the center of the ring pulls unreacted gas through, which then ignites. The new length scale generated here is roughly one-half of the length scale associated with the local density gradients existing before the shock-flame interaction. The local pressures and temperatures, as well as the interfaces, change due to the rotational motion associated with vorticity and the expansion due to combustion. The local reaction rates change and amplify the fluctuations, which in turn amplify the chemical rates. The process continues, progressively generating more sound waves and smaller length scales through vorticity generation. We see finally (0.70 ms) a fine-grained turbulent burning zone developing as as result of the vorticity generation - sound wave - chemical kinetic interactions. The passage of the reflected shock now introduces even more small scale structure in the system.

Two-dimensional numerical simulations have been performed by Picone et al. (1984) of a simplified problem in which a weak shock interacts with a bubble with a temperature and density gradient characteristic of a flame. However, since there are no chemical reactions or energy release here, the hydrodynamic effects can be isolated. Figure 21 shows the structures generated by the interaction of the shock and the bubble. The length scales of the structures is halved again when the reflected shock passes through. The calculations does not include the three dimensional pertubations and effects of ongoing chemical reactions and energy release which would generate smaller scale structure.

E. Combustion Instabilities

Oscillatory pressures have plagued combustors for many years. The difficulty has caused failures in solid rocket motors, liquid rocket engines, turbojet engines, and ramjet engines. The problems include the large vibrations themselves as well as subsequent effects such as flame blow-out. Chemical-acoustic coupling is one of a number of mechanisms that can lead to instabilities in such combustors. The fundamentals of solid-propellant combustion have recently been reviewed in a collection of papers edited by Kuo and Summerfield (1984).

As we have seen above, when reactions occur in a complicated flow field, many processes can interact. However, there are certain effects, such as reduction in ignition times, which can be directly attributable to chemical-acoustic coupling. When a highly reactive and exothermic mixture burns it generates a spectrum of pressure waves moving in all directions. First, there is the possibility that the waves generated could couple directly with and thus amplify the natural frequencies of the chamber. However, those waves moving towards previously reacted material probably decay, whereas those moving towards the unreacted material could heat or perturb unreacted material enough to reduce the ignition time. The tendency of the flame zone to expand could be halted as expansion fans catch up with the front. The coupling mechanism whereby pressure waves enhance reaction rates has been discussed above in some detail.

VI. CONCLUSIONS

Considering the complexity of the interaction between fluid dynamics and chemistry in combustion systems, it is important to establish the fundamental coupling mechanisms that can occur. The chemical-acoustic coupling process has two types of timescales, a chemical time and an acoustic time. However, it is not always easy to determine exactly which chemical time or which acoustic time is the important one. This ambiguity arises because of complex geometries or spatial inhomogeneities such as those occurring in rocket, turbojet or ramjets combustor environments.

A characteristic acoustic time is usually defined as the sound speed divided by a characteristic length. However, in actual combustors the sound speed varies throughout the chamber due to variations in temperature and composition. The choice of length scale is also important: the transverse or radial length of the material or combustor or some combination for unusual geometries. While in general these timescales can be calculated, they will vary for each system configuration. Finally, there are at least two chemical times to consider: the chemical induction time of the fuel and the time for energy release. Both of these can vary if the mixture is inhomogeneous.

In the material presented above, the coupling between chemistry and acoustics has been demonstrated for idealized systems. The importance of this interaction process in more complicated systems is less obvious and can be masked due to complex geometric effects as well as effects which are dispersive in both amplitude and frequency. Thus to understand and isolate the effects of the coupling process, simplified models have been used in the analyses and simulations. The work described in the bulk of this paper has shown the potential significance of the coupling. However, because of the simplifications used in the models, most of the work to date can only be related qualitatively as opposed to quantitatively to combustors.

Extensions of the research described in this paper involve looking at the coupling process in more complicated scenarios. Theoretical efforts must consider sound waves coupled to realistic flame models, sound wave effects on multi-step chemical reaction mechanisms, and chemical-acoustic interaction in complex geometries. This necessarily means making greater use of numerical techniques than has been done in the past, many of which are currently available. For example, using complex chemical kinetic mechanisms, we can predict how the induction and energy release times of hydrocarbon fuels are altered by sound waves and which frequencies are the most effective in causing changes. Currently existing flame models with detailed chemical kinetics can be used to predict coupling effects and these can be directly comparable to experiments.

There have been relatively few experimental efforts in chemical-acoustic coupling: it is difficult to isolate the effects from naturally occurring dissapative and dispersive effects. Efforts to look at these problems have provided useful corroboration of, and some contradictions to, the the theoretical predictions. The possibility now exists of monitoring the details of the chemical reactions during the interaction process with diagnostic techniques such as LIF and CARS. These techniques are particularly applicable to interactions of premixed or diffusion flamefronts with sound waves. It is also important to experimentally test the validity of predictions made for the changes in the induction times of materials, which are subtler than the changes in the energy release time but dominant in some chemical system.

ACKNOWLEDGMENTS

This work has been sponsored by the Office of Naval Research. The authors would like to thank Mrs. F. Rosenberg for her valuable editorial assitance and Professors J.F. Clarke, T.-Y. Toong, and D.H. Edwards, and R.G. Gilbert for providing figures. The authors would like especially to thank Professors J.F. Clarke and T.-Y. Toong for their helpful suggestions.

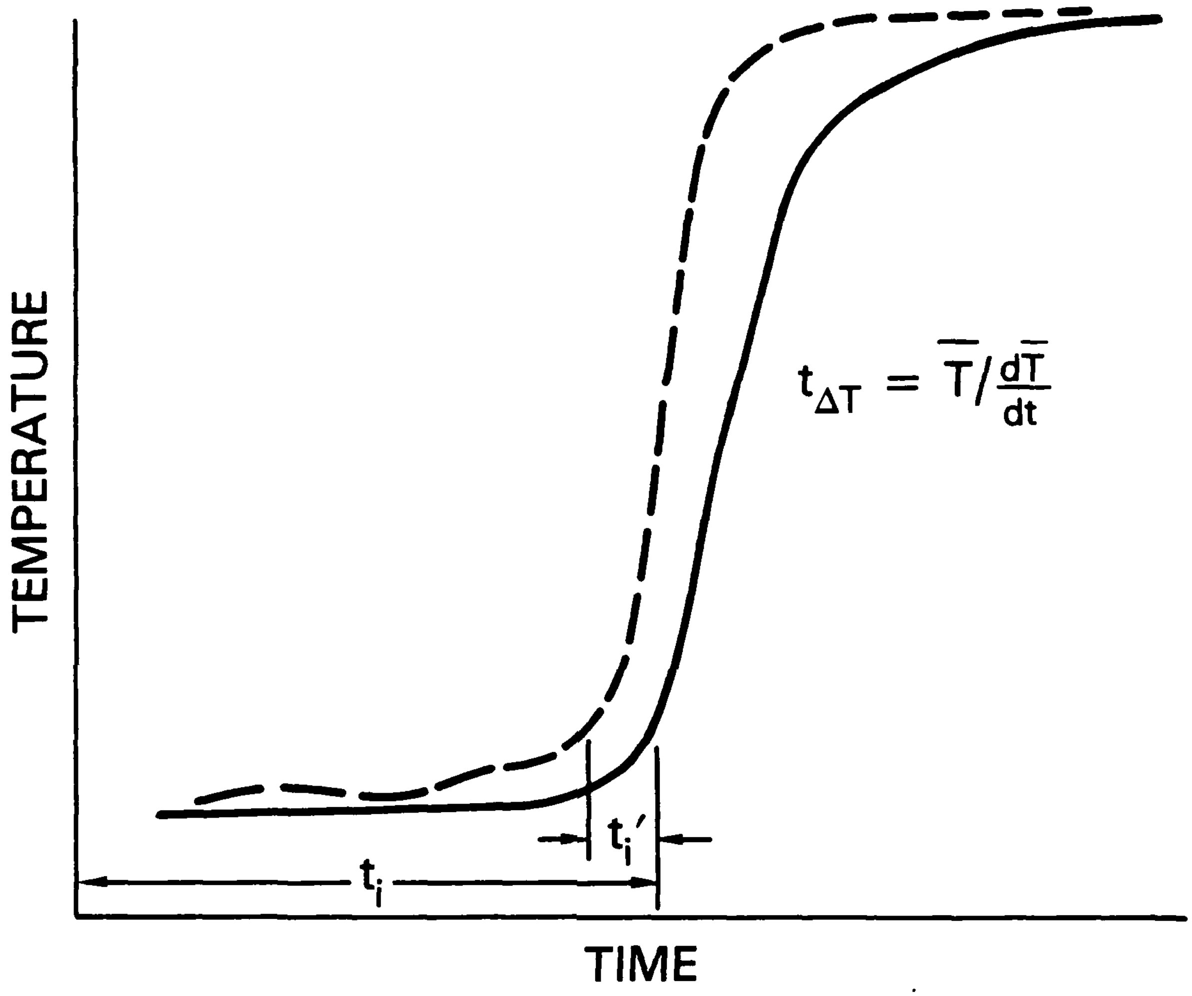


Figure 1. Qualitative illustration of temperature as a function of time for a premixed igniting system. The t_i is the chemical induction time, t_i' is the change in the induction time due to an acoustic pulse, and $t_{\Delta T}$ is the energy release time. Note that the acoustic pulse can change both the induction time and the chemical release time.

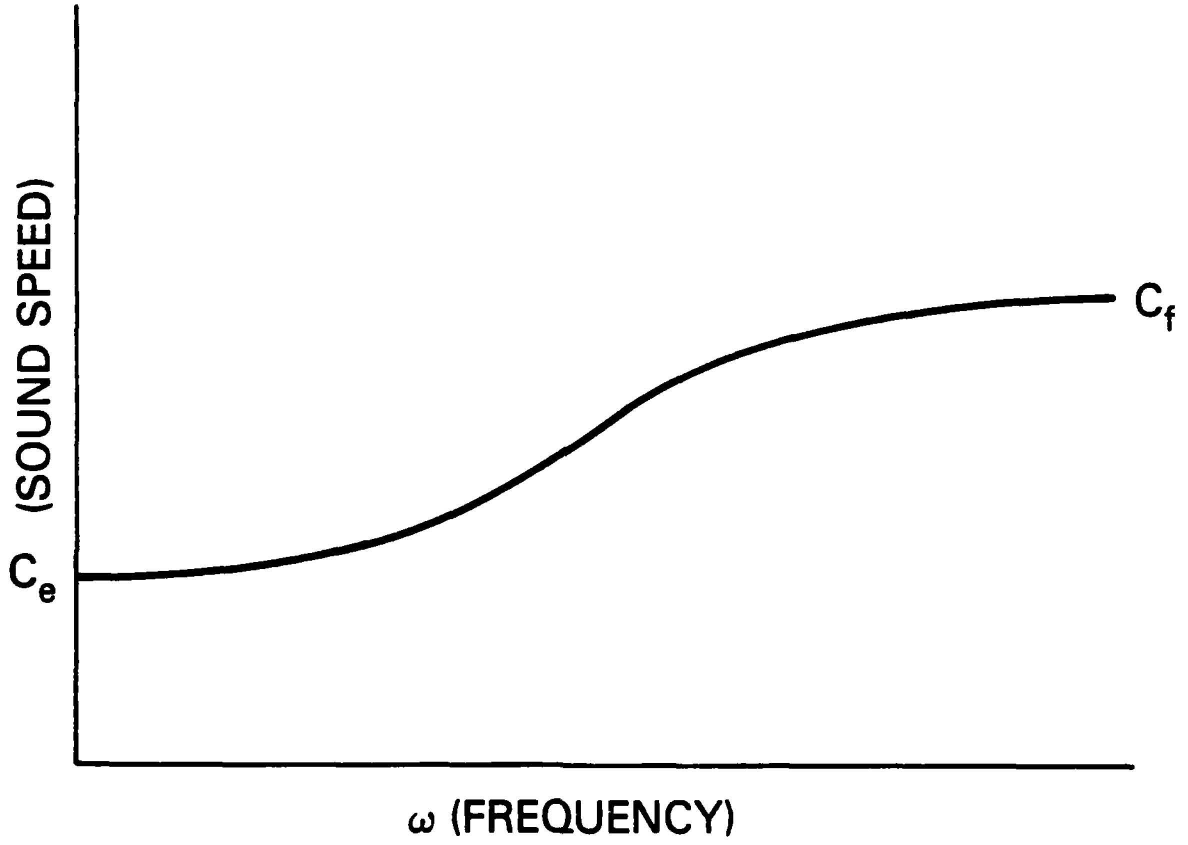
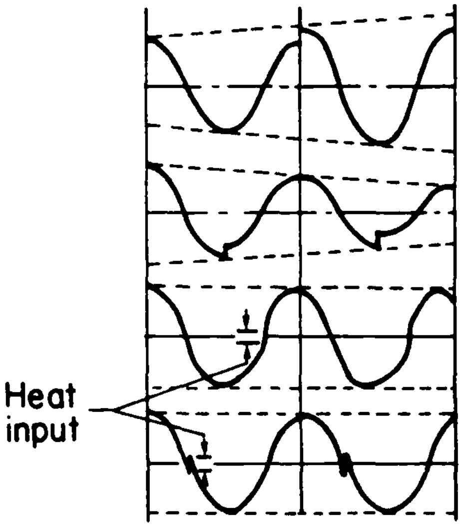


Figure 2. Qualitative illustration of the sound wave phase velocity as a function of frequency showing variations from the equilibrium sound speed, c_e, to the frozen sound speed c_f as the frequency ω increases.

Rayleigh's Criterion



	Phase of heat supply relative to phase of pressure	Effect on amplitude	Effect on frequency
1	In phase	Increase	None
2	Opposite phase	Decrease	None
3	Quarter-period before	None	Increase
4	Quarter-period after	None	Decrease

(A. Wood, Acoustics p. 93, Interscience, 1941)

Figure 3. Illustration of Rayleigh's criterion showing how heat input can change the amplitude and frequency of a sound wave (from Wood, 1966)

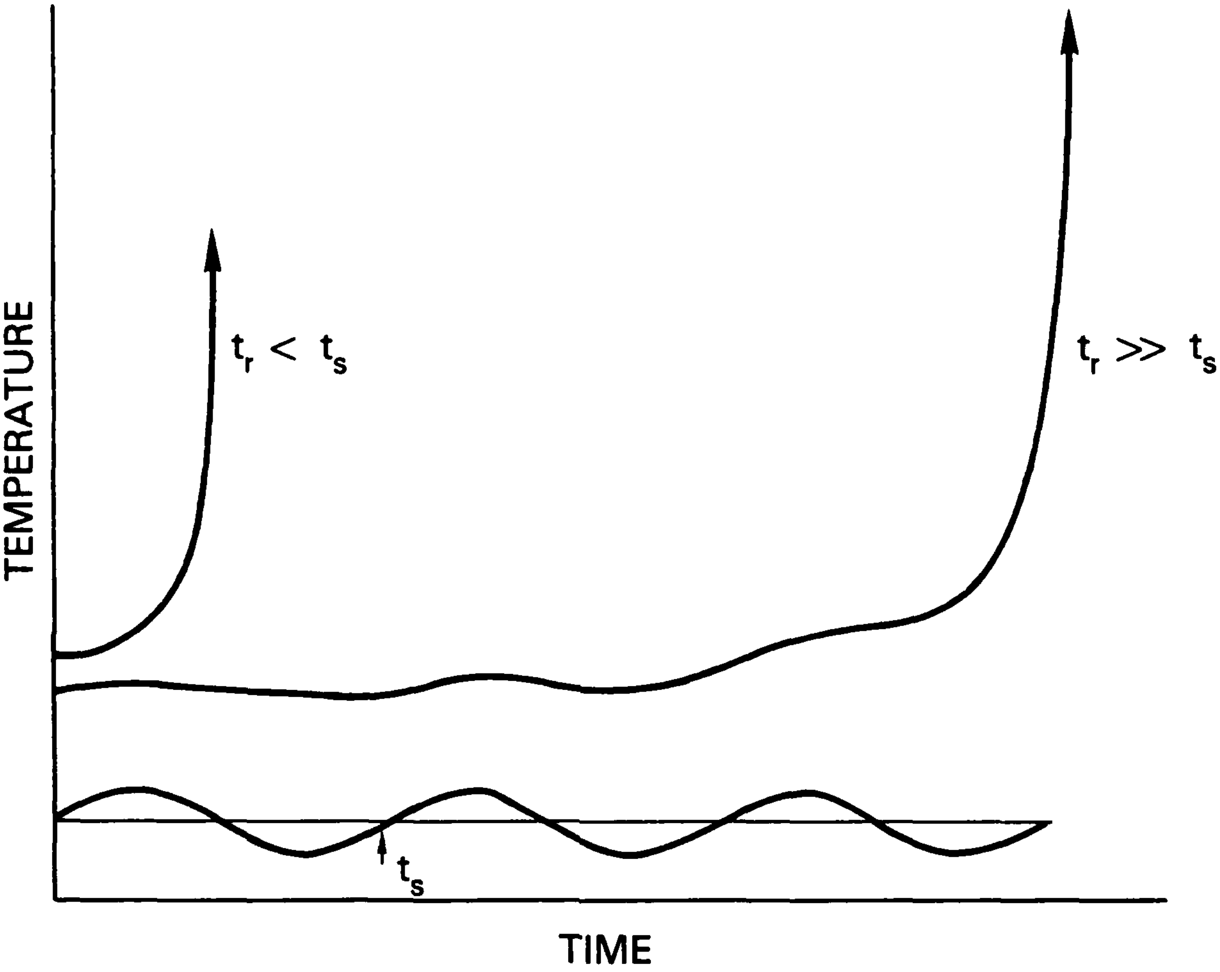


Figure 4. Qualitative illustration of the important timescales in a chemical-acoustic problem and how they may be related. The t_r is the chemical reaction time and t_s is the sound period. Two cases are shown: one for short reaction time and another for a reaction time long compared to the sound transit time. The sinusoidal curve is an amplified graph of the sound pulse.

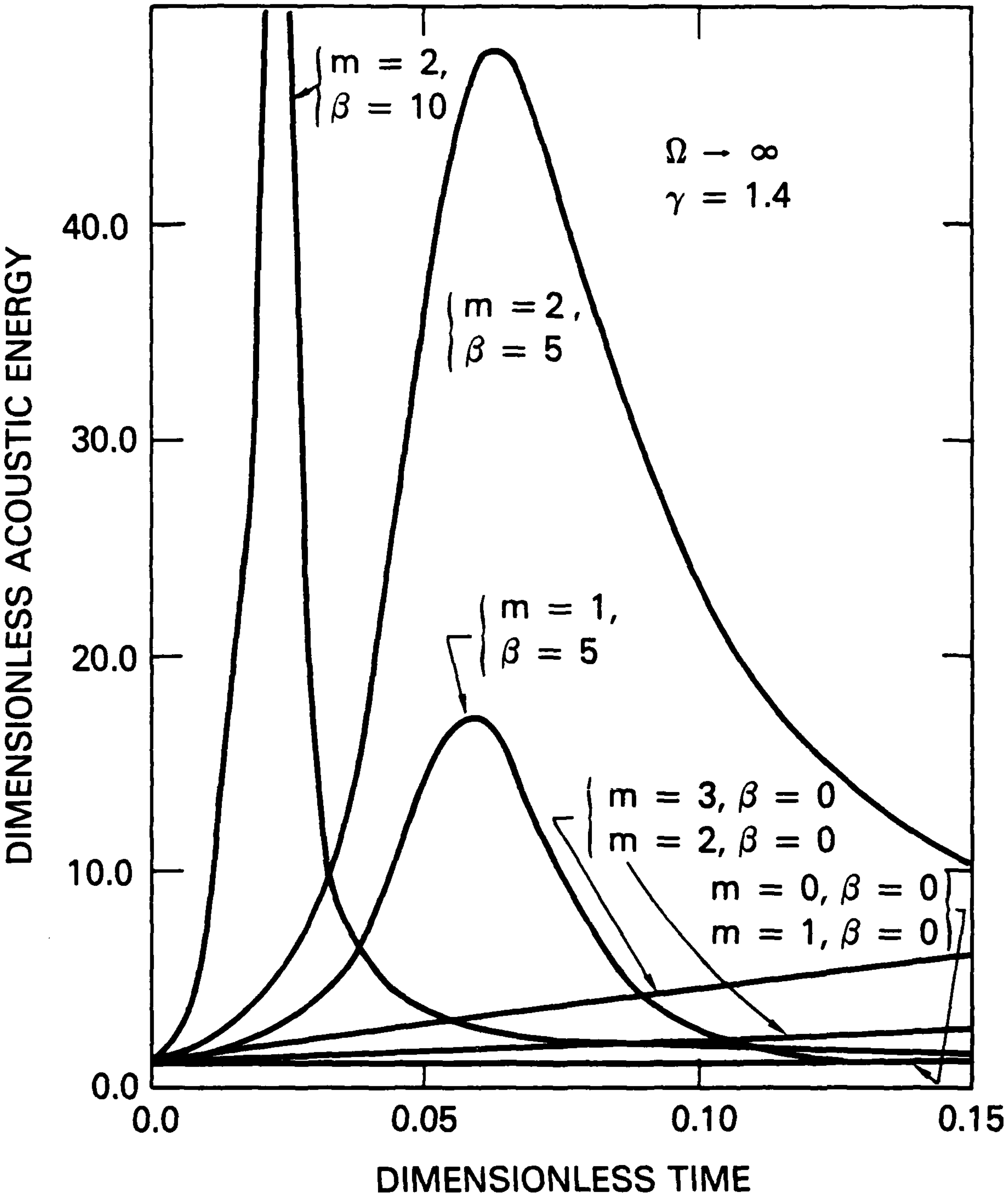


Figure 5. Acoustic energy as a function of time for the limit of long chemical reaction time for a chemical reaction of order m and activation energy β (from Toong et al., 1974).

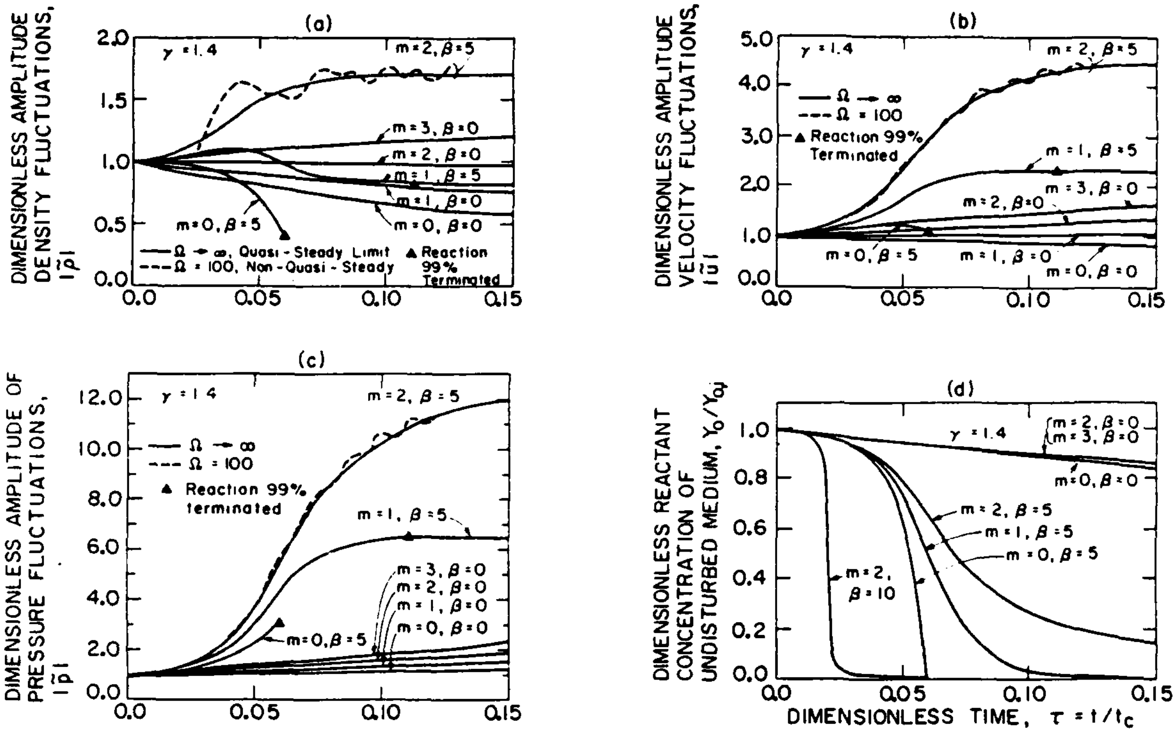


Figure 6. Calculated amplitudes of (a) density, (b) velocity, and (c) pressure fluctuations and (d) undisturbed reactant concentration for the long (solid lines) and short (dashed lines) chemical time limit as a function of dimensionless time (from Toong et al., 1974).

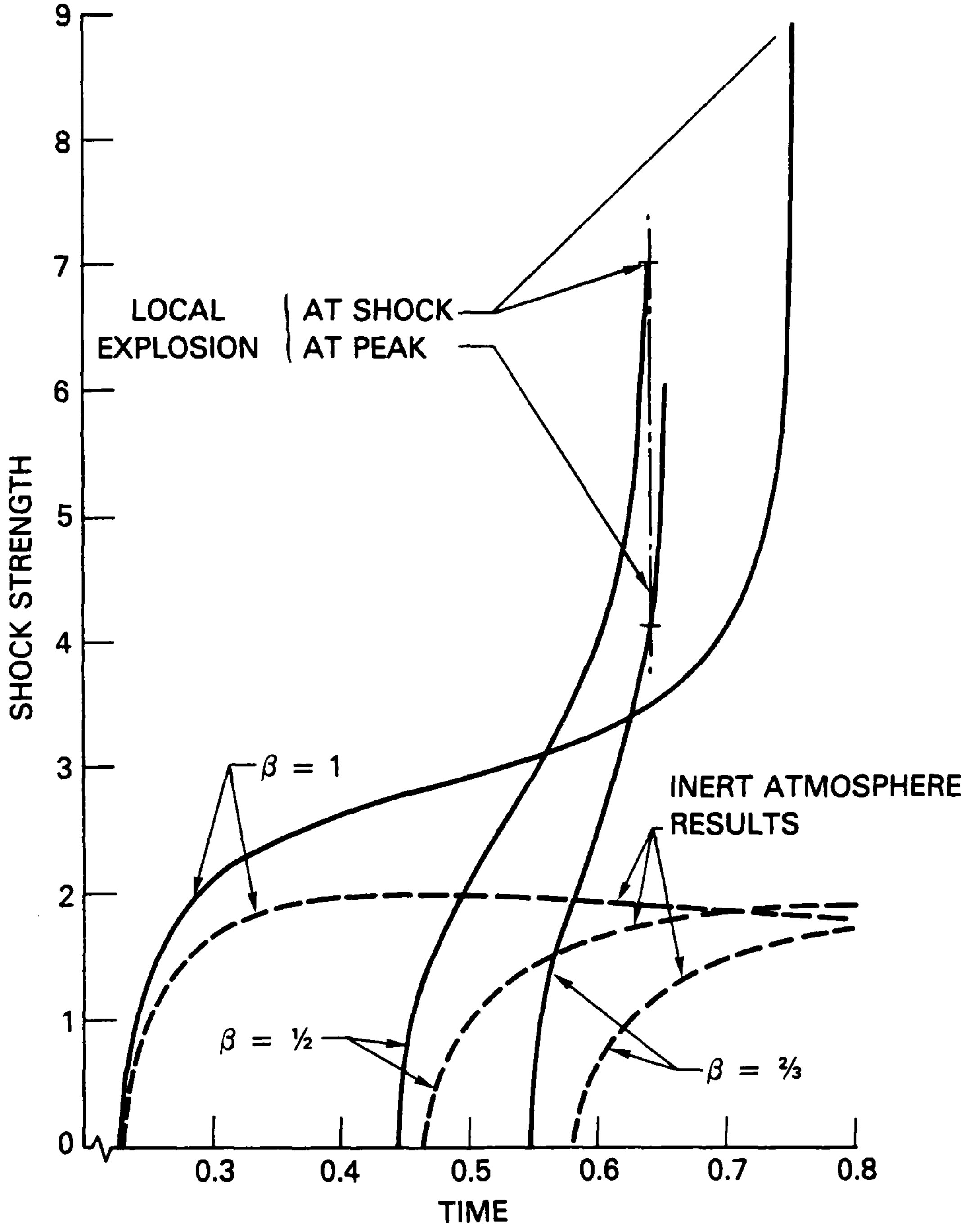


Figure 7. Calculated shock strength as a function of time in a reactive atmosphere compared with the behavior from the same initial state in an inert atmosphere. The shock strength grows rapidly as the disturbance length increases (from Clarke, 1979). The parameter b is the inverse of the disturbance length.

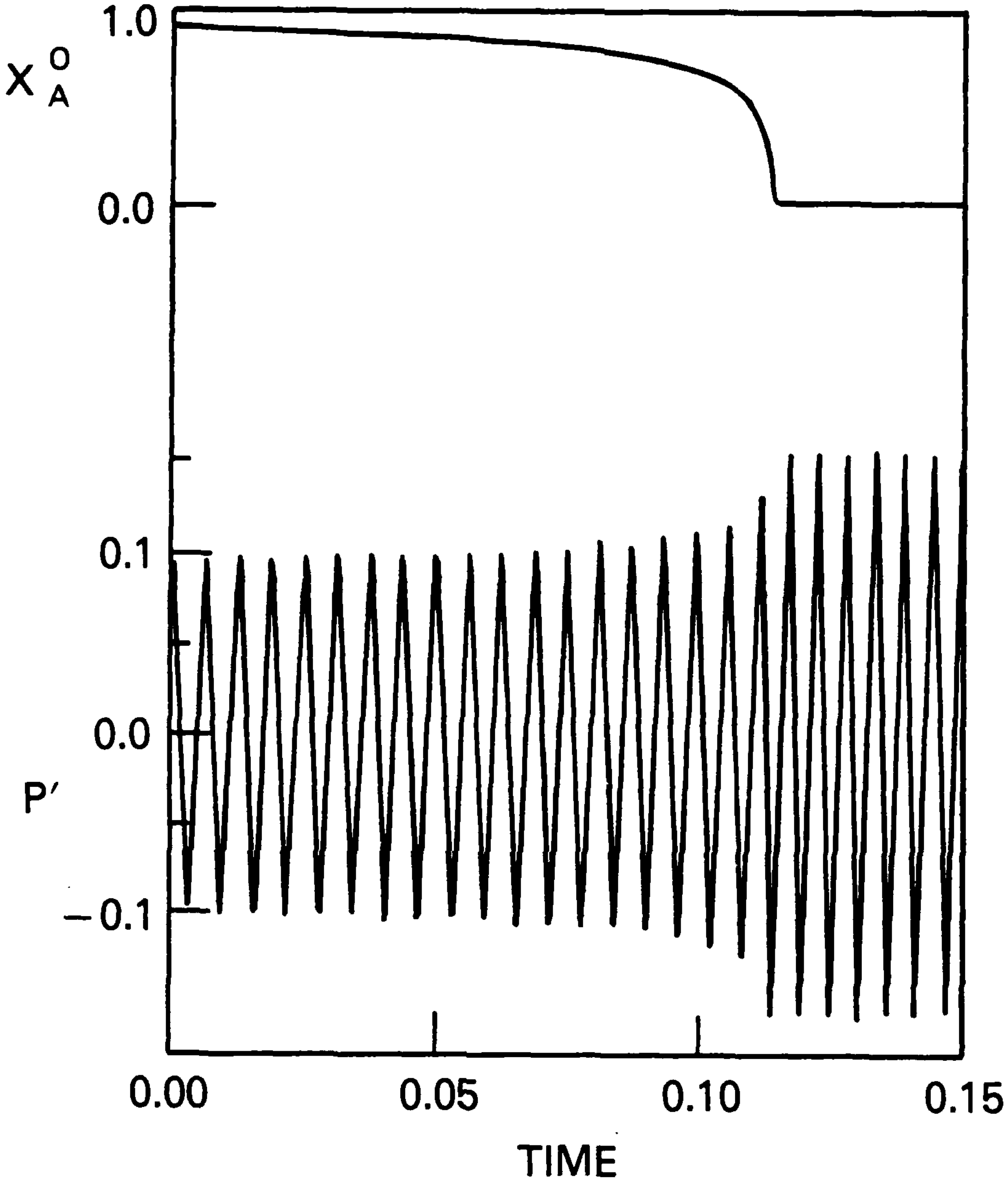


Figure 8. Calculated sound propagation in a unimolecular reaction A → B in the high-pressure limit: dimensionless Fourier transform of the inhomogeneous part of the pressure p' as a function of reduced time corresponding to the decomposition of cyclopropone at 953 K, sound frequency 500 Hz. The time dependence of the homogeneous mass fraction of A, x_A^0, is also shown. The amplitude and frequency increase, respectively to about 150% and 114% of their zero time values.

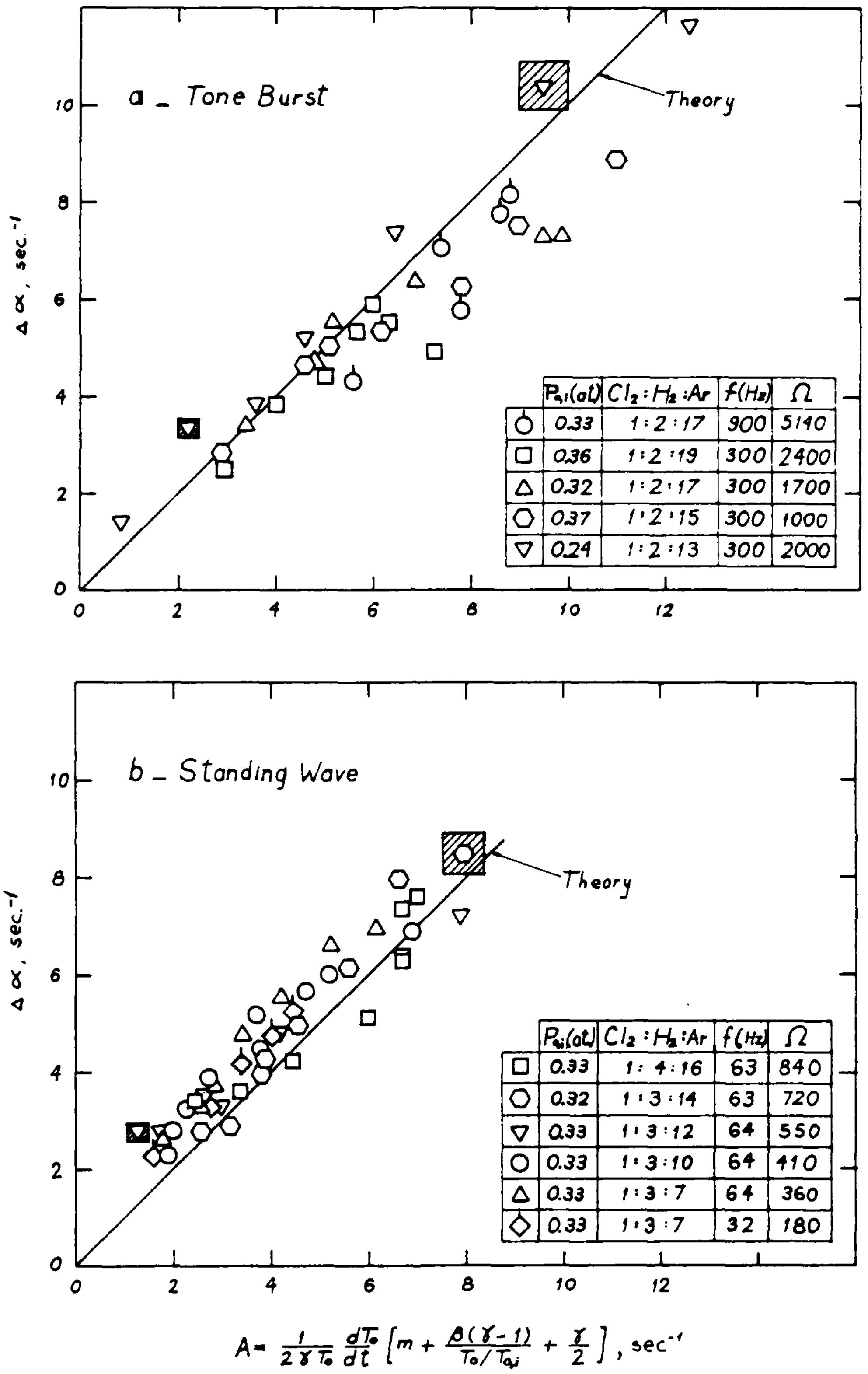


Figure 9. Observed sound amplification rate, Δα, versus predicted rate, A, for tone-burst and standing-wave experiments (from Abousief et al., 1979).

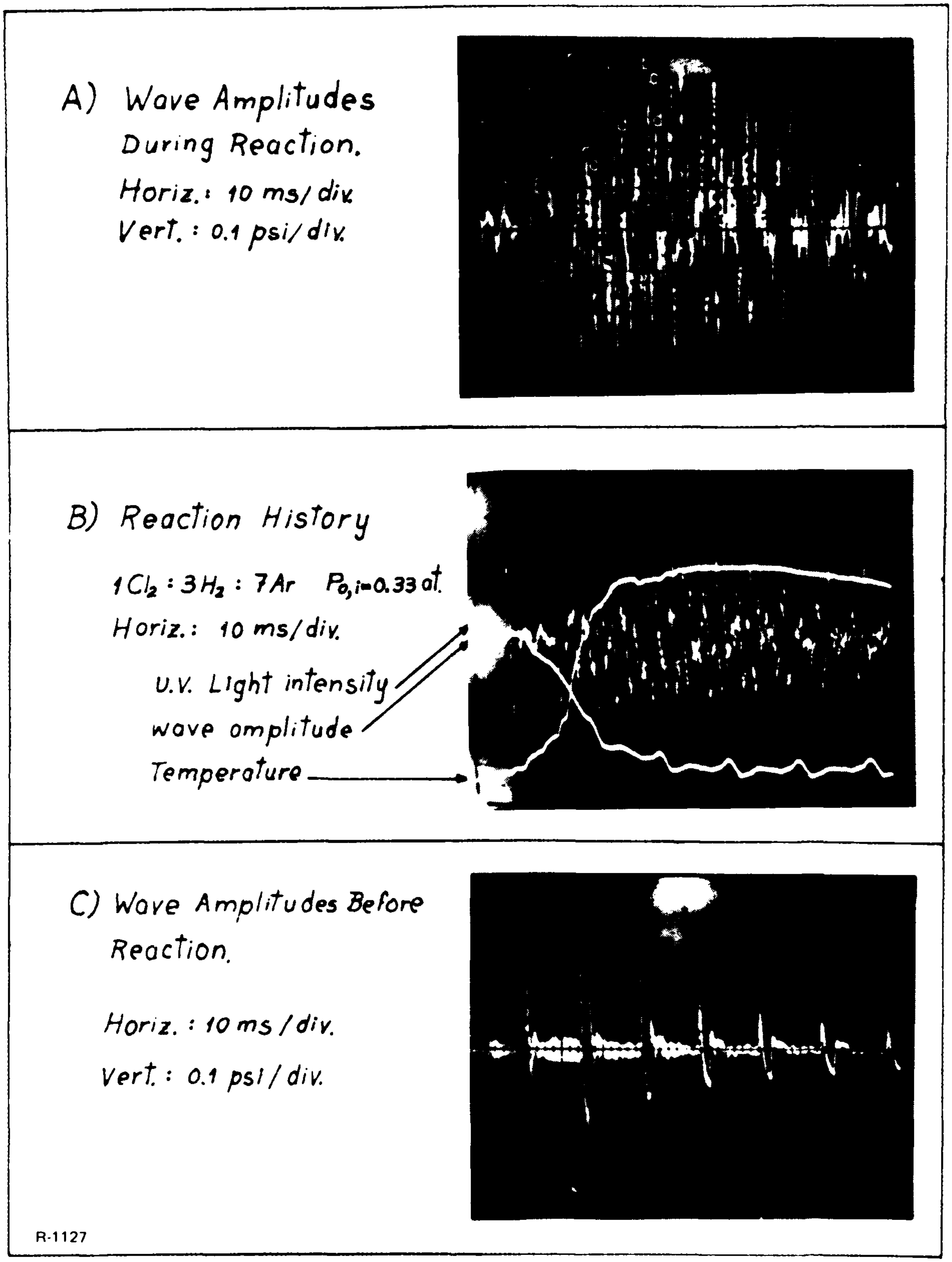


Figure 10. Experimental data from a typical weak shock experiment showing showing shock wave amplification due to chemical reactions (from Abousief et al., 1979).

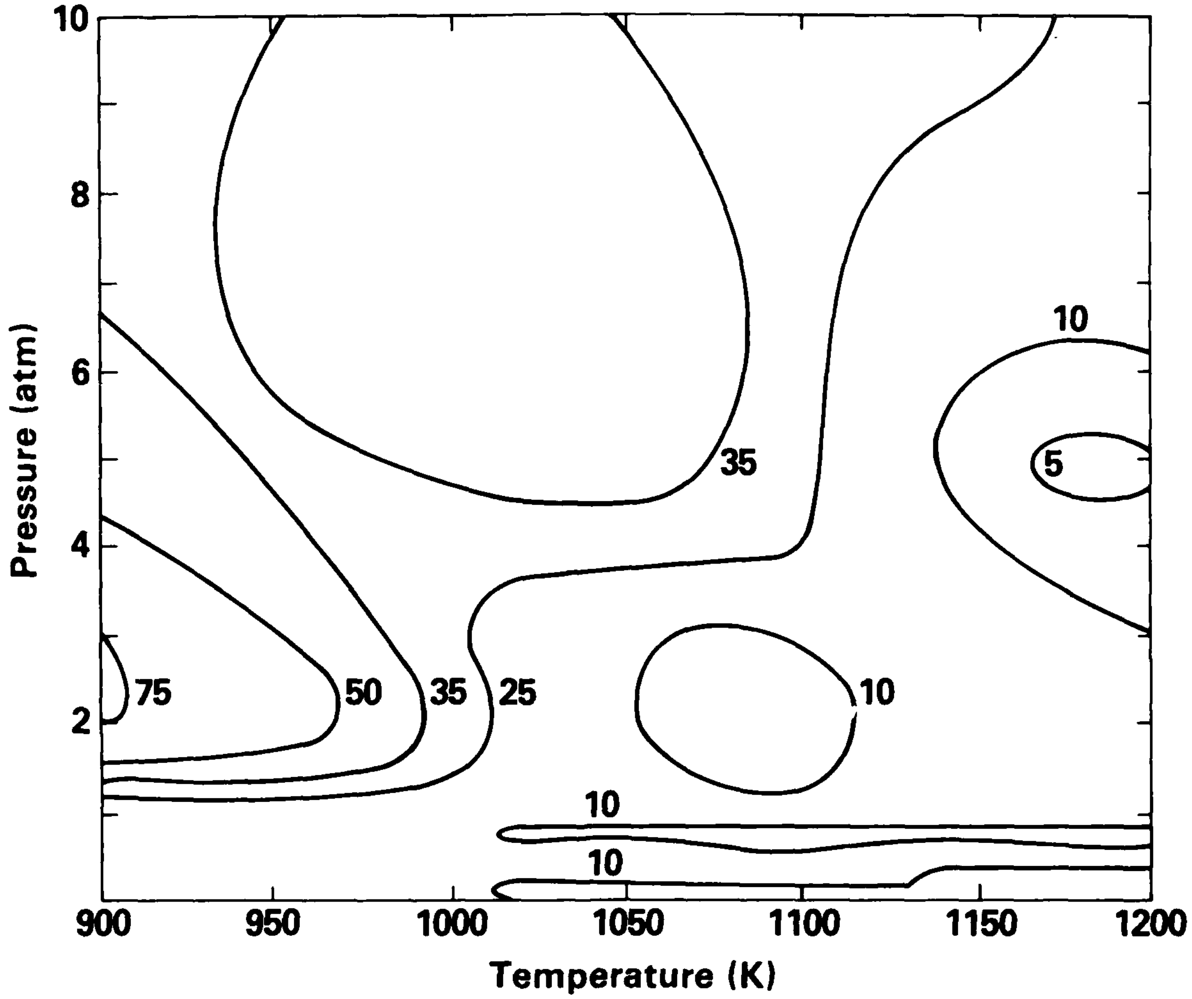


Figure 11. Contours of the quantity $(-T/\tau)*(\Delta\tau/\Delta T)$ shown on the temperature-pressure plane for a mixture of hydrogen-oxygen-nitrogen in the ratio 2:1:4. Large values of the quantity indicate sensitivity of the induction time τ to sound wave perturbations.

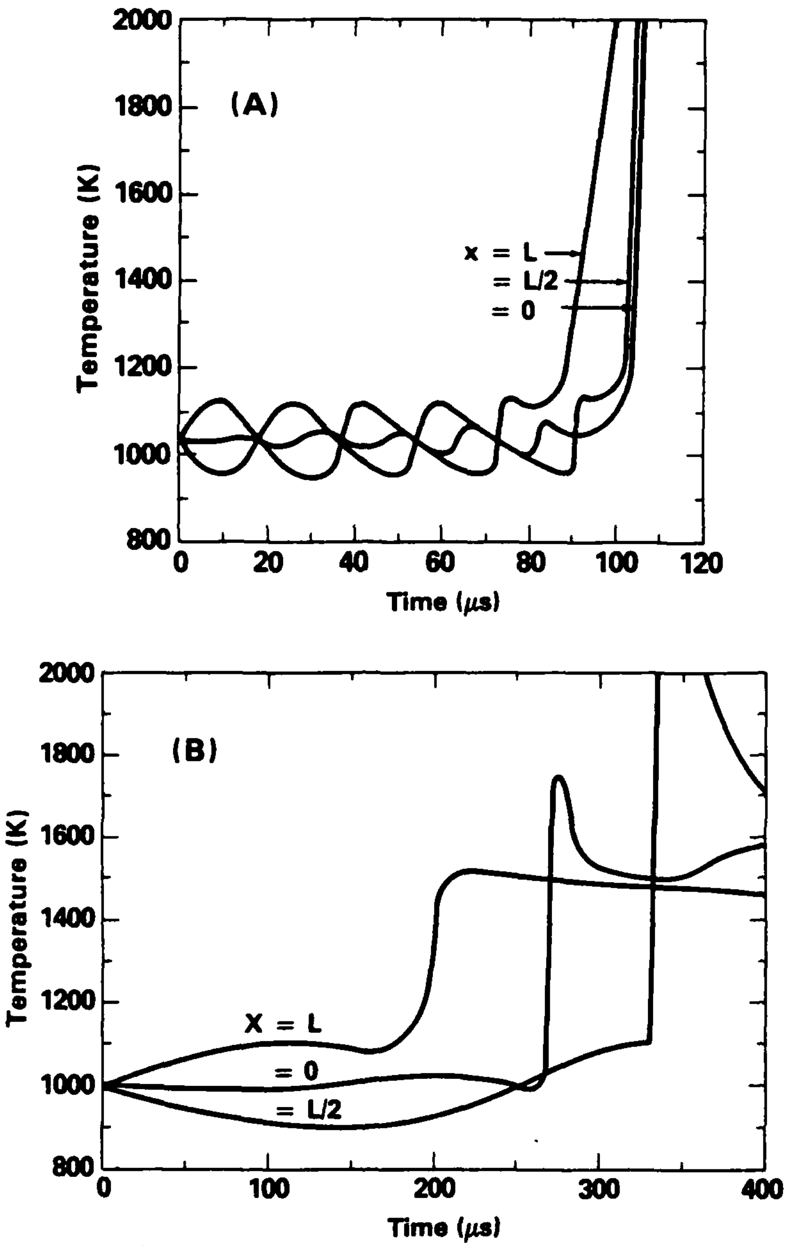


Figure 12. Calculated variation of temperature as a function of time in an initially uniform reacting mixture of hydrogen-oxygen-argon at three locations in the standing sound wave pulse of half-wavelength L, for given initial pressure, P_o, temperature, T_o, and sound wave amplitude, u. (a) H_2:O_2:Ar/2:1:7, $P_o = 1.3$ atm, $T_o = 1036$ ^oK, $u = 1.0 \times 10^4$ cm/s, L = 1.17 cm. (b) H_2:O_2:Ar/8:2:90, $P_o = 3.72$ atm, $T_o = 1000$ K, $u = 1.0 \times 10^4$ cm/s, L = 15.0 cm (from Oran and Boris, 1981).

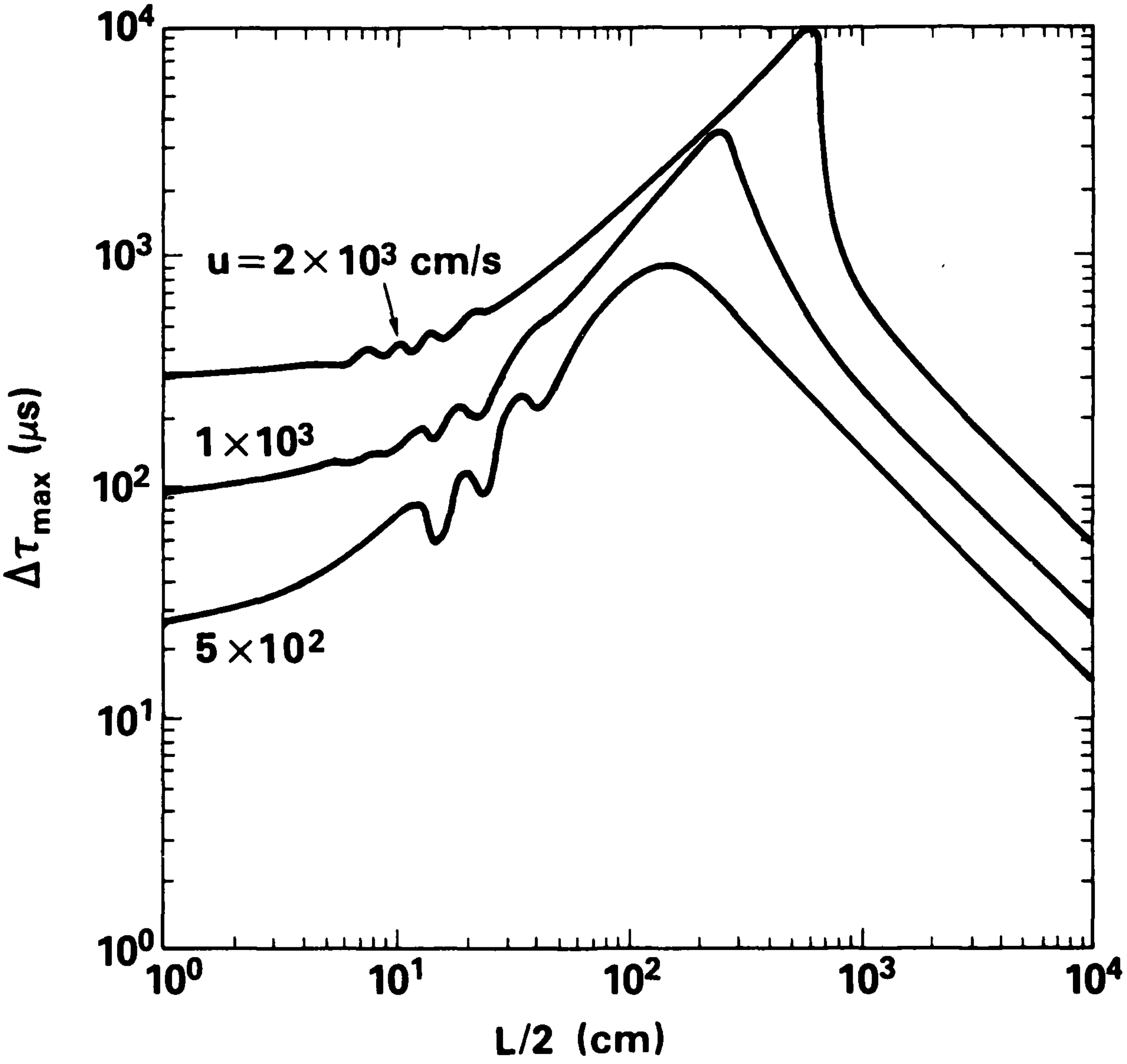


Figure 13. Calculated maximum decrease in the induction time as a function of the half-wavelengh L of a sound wave in a system of H_2:O_2:Ar/8:2:90, T_o = 1000 K, P_o = 3.72 atm, for three initial values of the sound wave amplitude u (from Oran and Boris, 1981).

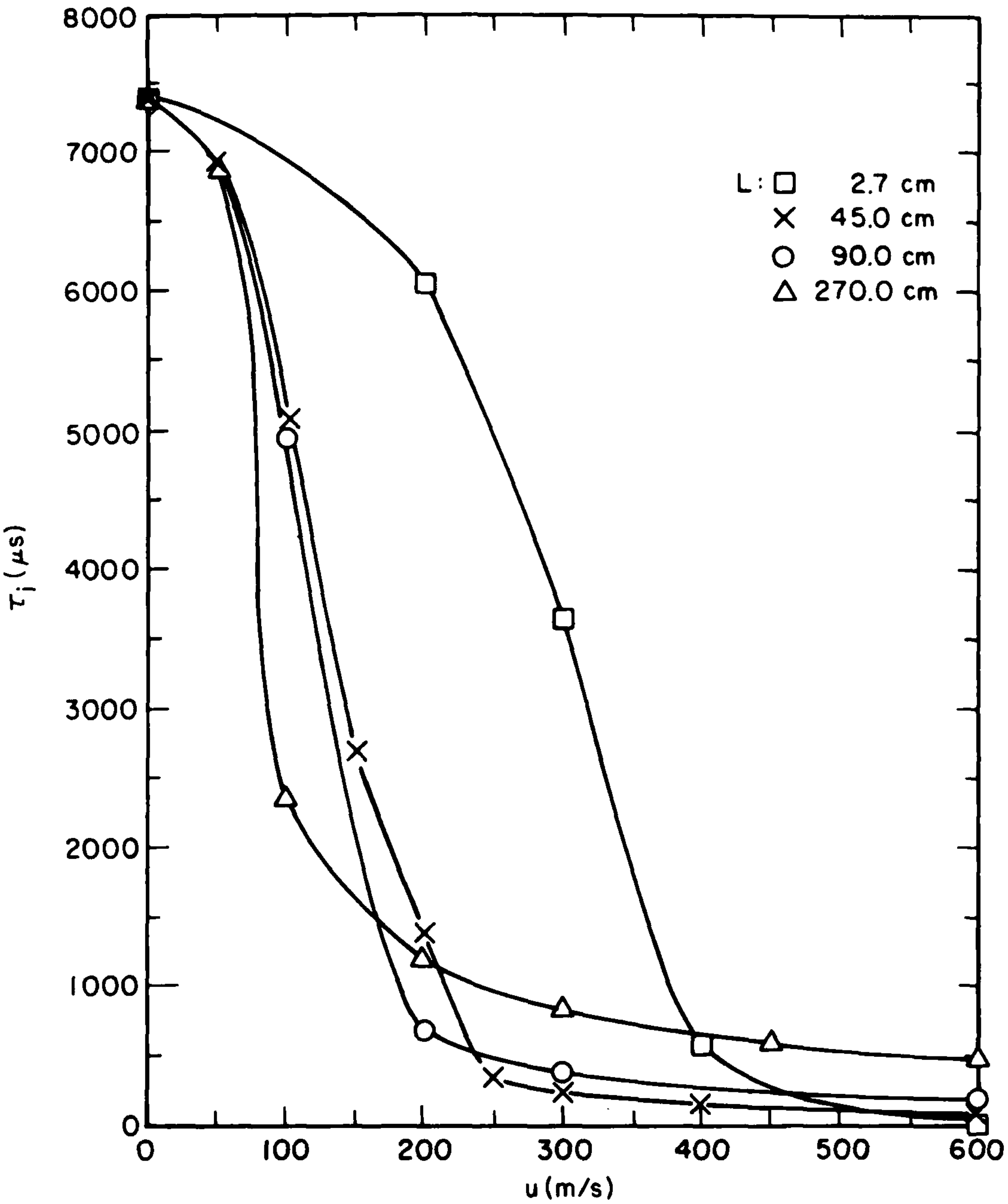


Figure 14. Calculated variation in the induction time as a function of initial amplitude of a sound wave velocity perturbation for a mixture of $H_2:O_2:N_2/2:1:4$, $P_o = 7.9$ atm, $T_o = 975$ °K, for four values of the half-wavelength L (from Kailasanath and Oran, 1984).

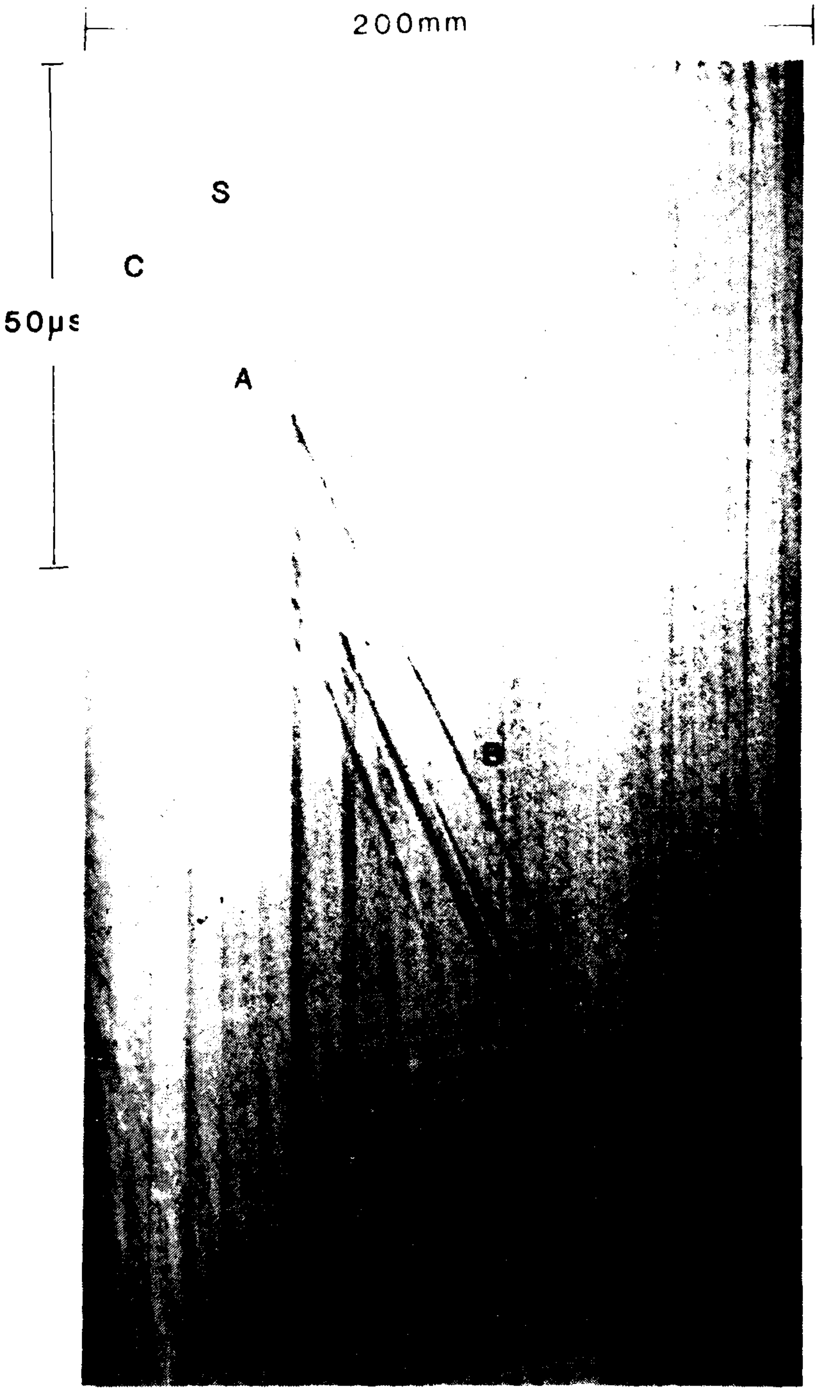


Figure 15. Streak schlieren photograph of shock ignition of $C_2H_2 + 2.5O_2 + 6N_2$ at $P_o = 0.013$ MPa; S, incident shock; C, contact surface; A, beginning of exothermic reaction; B, onset of shock front acceleration; D, transition to detonation wave E; R, retonation wave; F, flame fronts originating from reaction centers. (from Edwards et al., 1981).

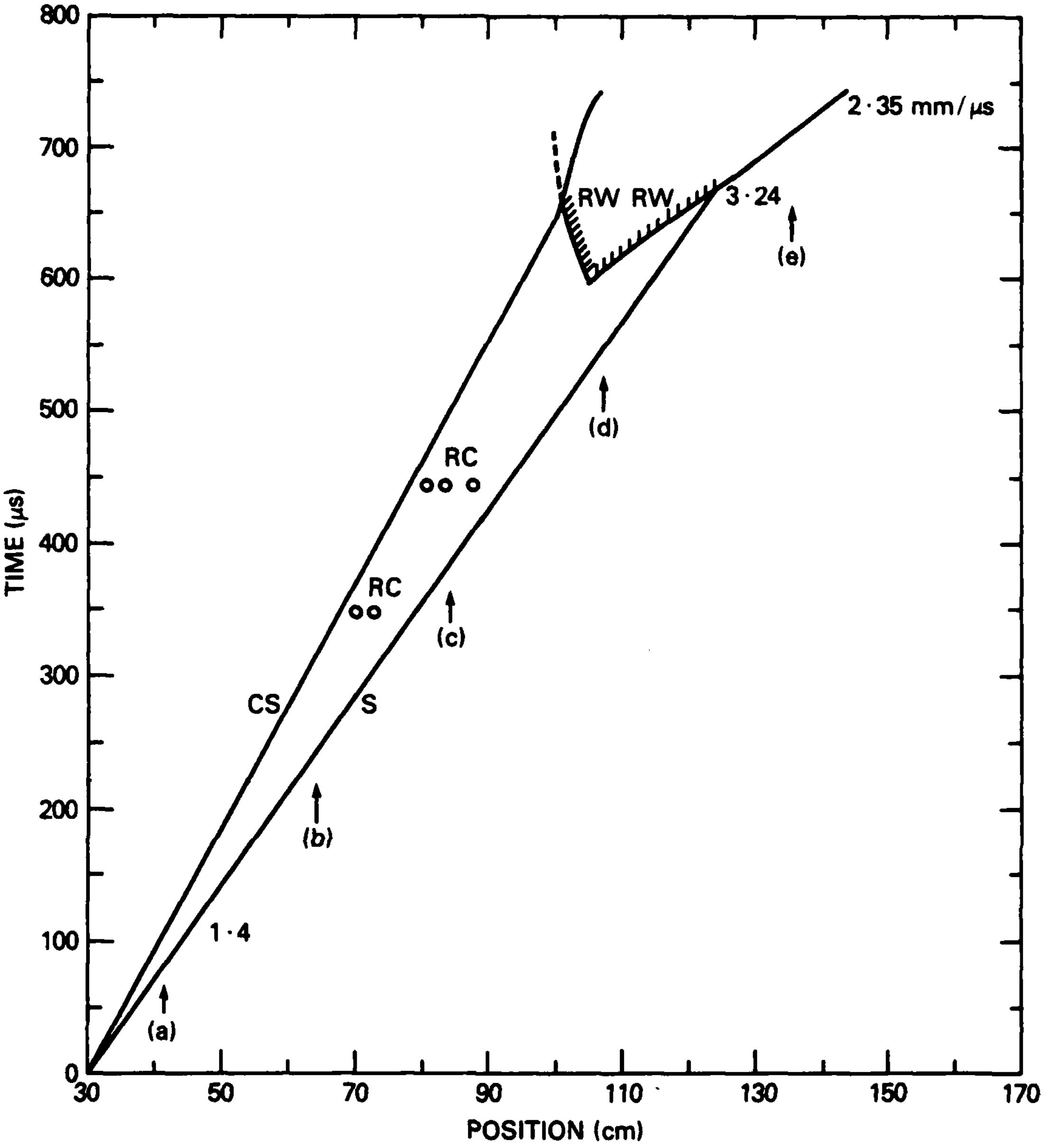


Figure 16. Calculated time versus position for the main events for a $H_2:O_2:N_2$ mixture with the incident shock geometry as in the streak schlieren in Figure 15: S, incident shock; CS, contact surface; RC, reactive centers; RW, reaction wave; (a) pre-ignition regime, (b) quasi-steady shock-reaction complex, (c) formation of reactive centers, (d) hot spot formation leading to overdriven detonation and (e) detonation relaxation. (from Kailasanath and Oran, 1983)

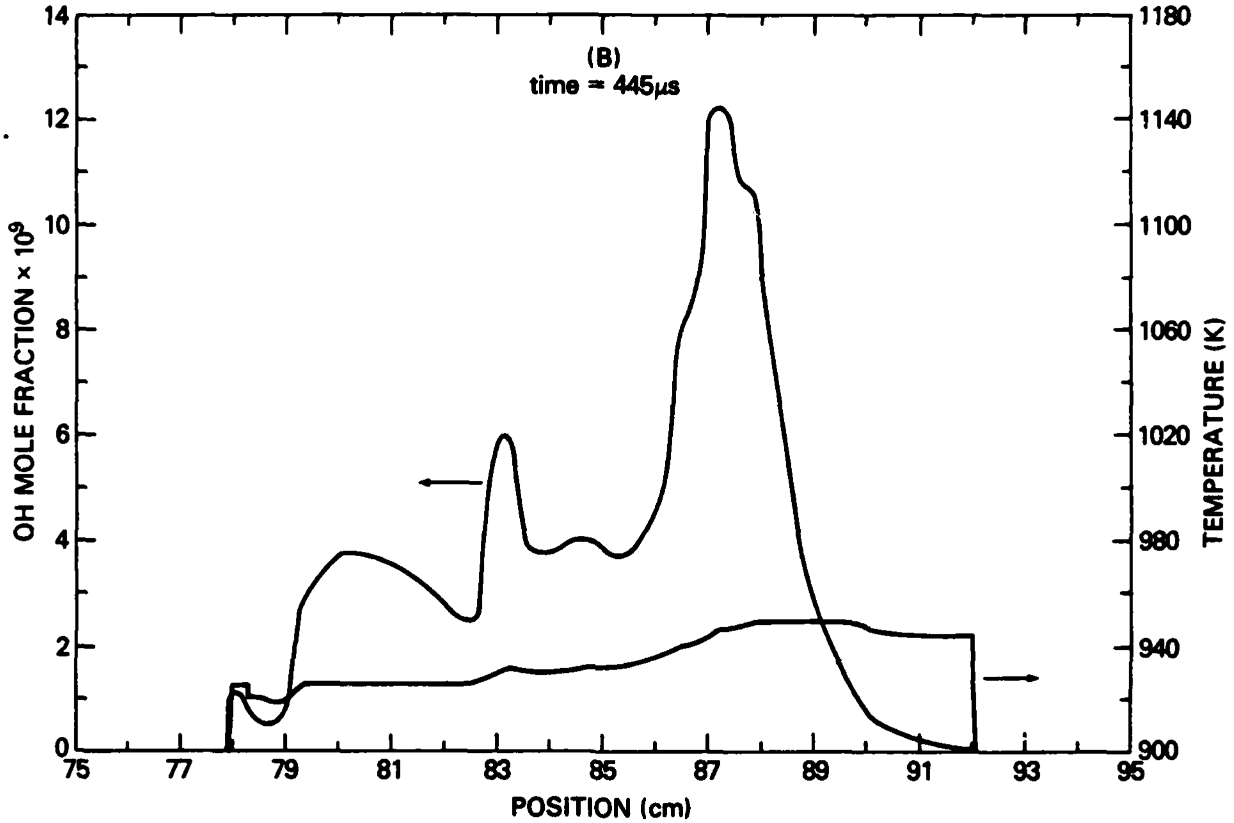


Figure 17. Calculated spatial variation of the temperature and the OH mole fraction between the contact surface and the shock wave at 445 μs corresponding to Figure 16. (from Kailasanath and Oran, 1983)

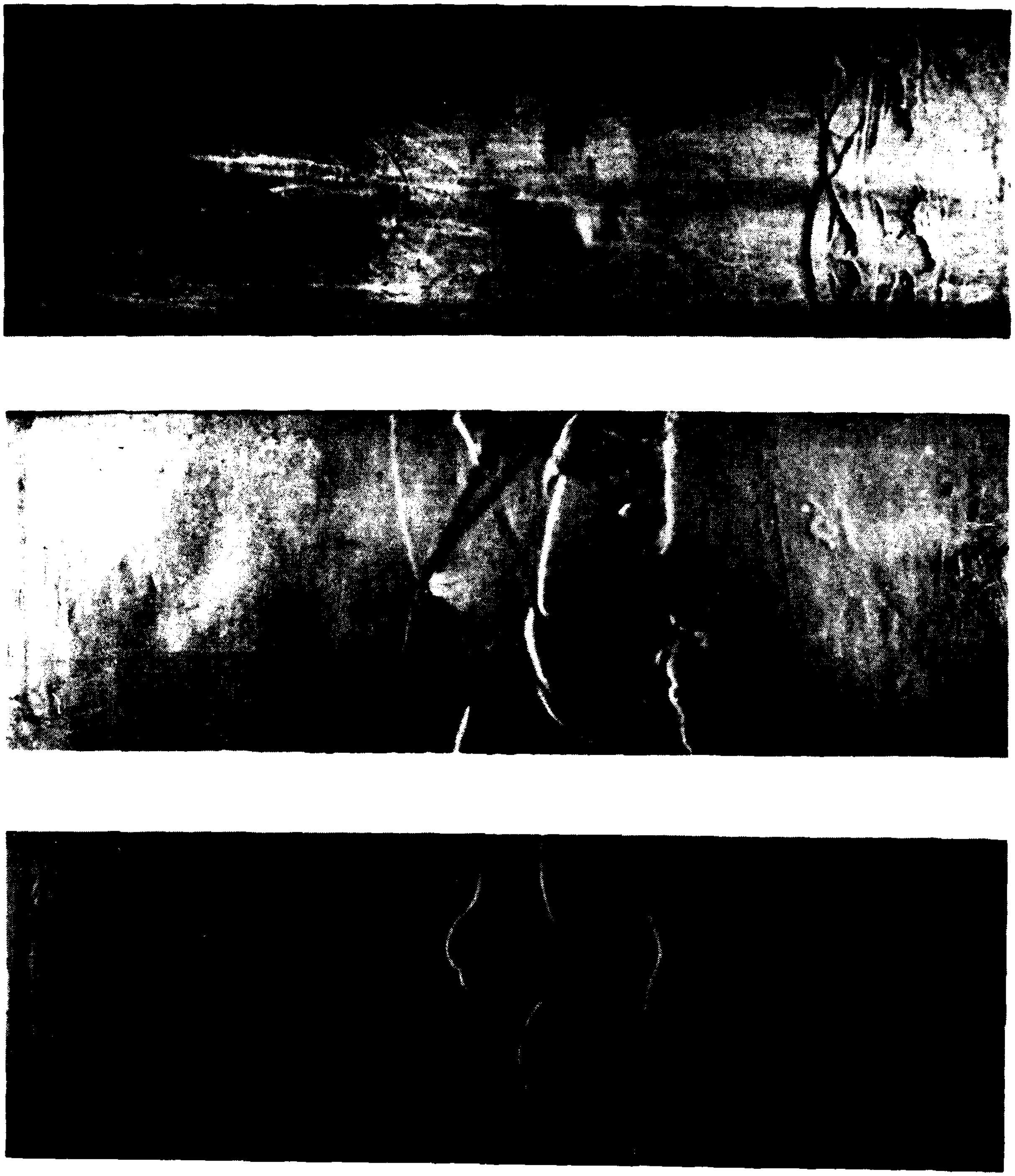


Figure 18. Three schlieren photographs of propagating detonations taken from a series of experiments in a tube 3" high and 1/4" wide filled with stoichiometric H_2-O_2 mixtures in 60% Ar at pressures from 50-80 Torr. Unburned pockets of gas are observed behind the detonation fronts (from Oran et al., 1982)

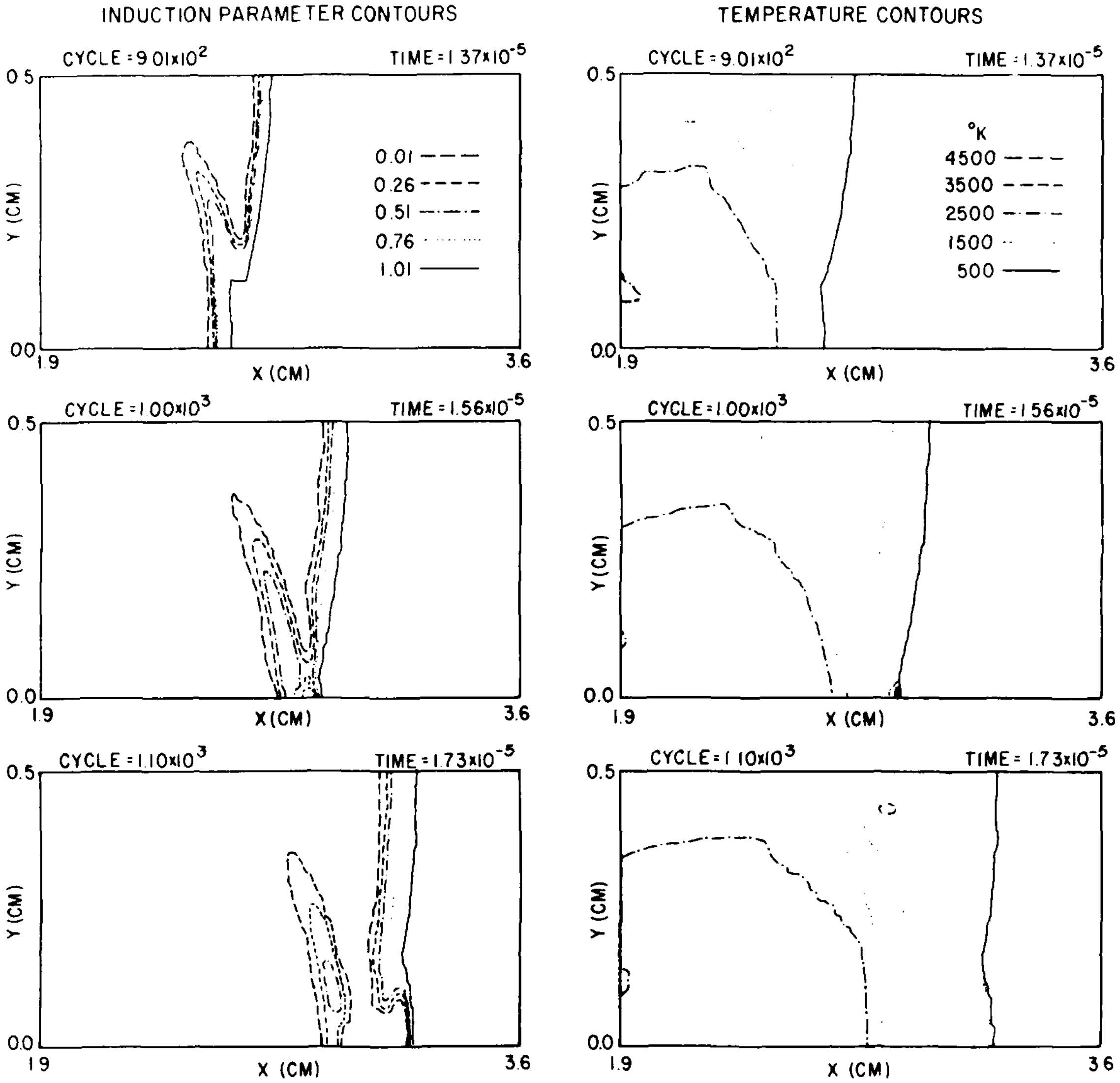
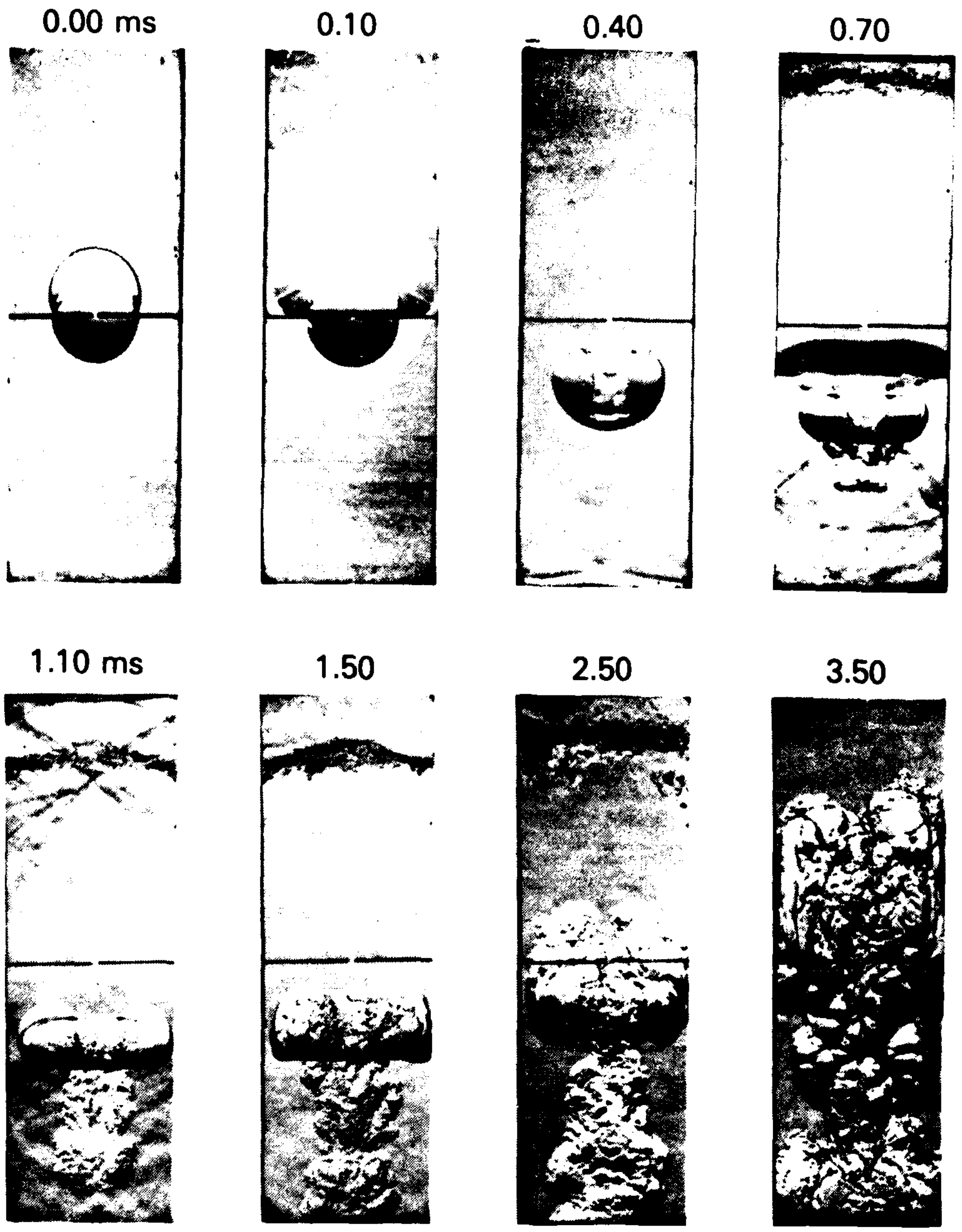


Figure 19. Calculated temperature contours and induction parameter contours (which indicated the amount of material reacted) for a mixture of H_2:O_2:Ar (from Oran et al., 1981)



MARKSTEIN (1964)

Figure 20. Interaction between a shock wave and a flame of initially roughly spherical shape. Pressure ratio of incident shock to background pressure is 1.3, stoichiometric n-butane-air mixture ignited at center of combustion chamber 8.70 ms before origin of timescale. (from Rudinger, 1958 and Markstein, 1964)

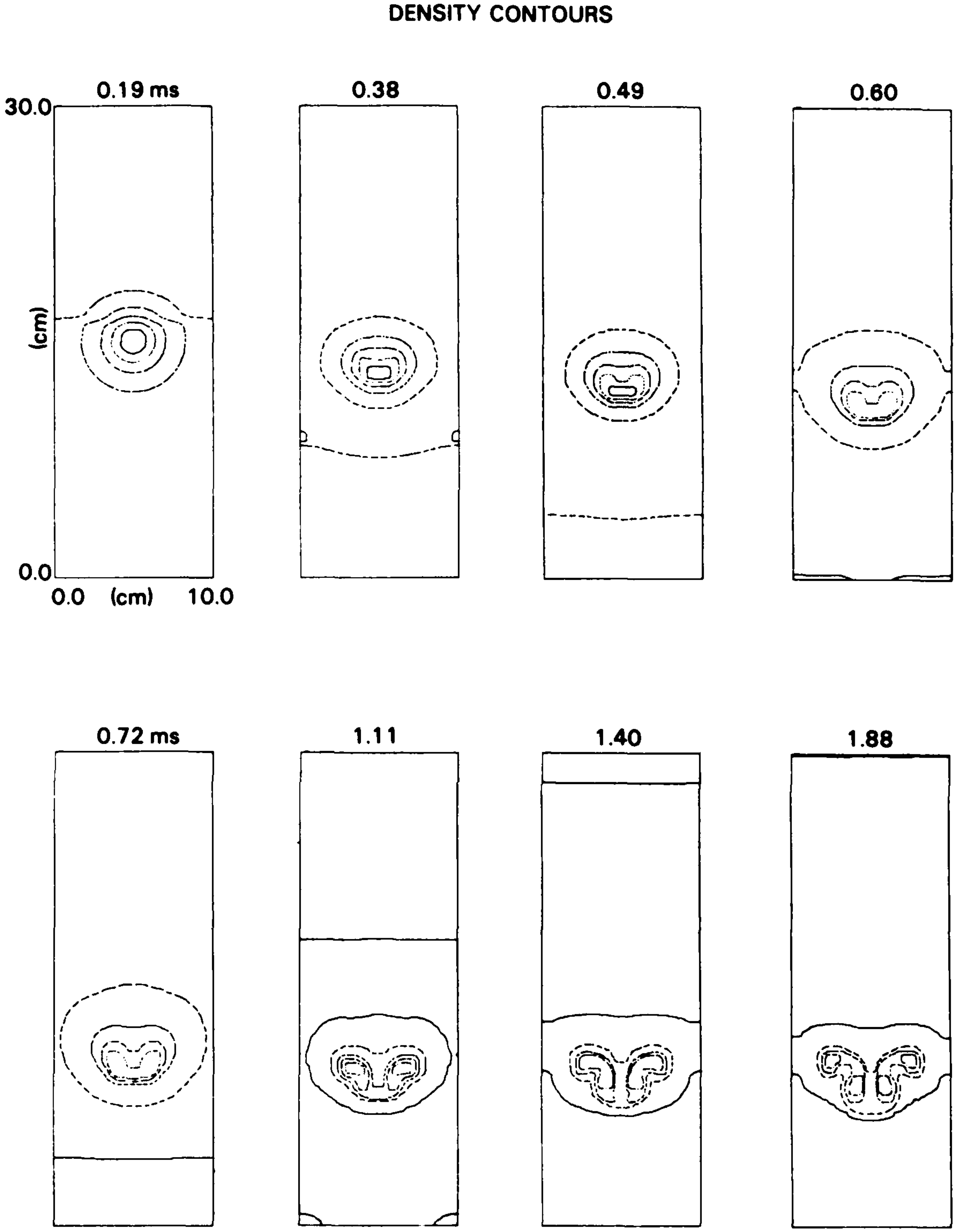


Figure 21. Calculated density contours for a two-dimensional simulation of the experiment shown in Figure 20 (from Picone et al., 1984).

REFERENCES

Abouseif, G.E., Toong, T.-Y., and Converti, J., Seventeenth Symposium (International) on Combustion, p. 1341, The Combustion Institute, Pittsburgh, 1979.

Bailey, J.J., J. Appl. Mech. 24, 1957, p. 333.

Bass, H.E. and Detsch, R.M., Amplification of Sound by Gas Phase Reactions, PARGUM Report 83-02, University of Mississippi, University, MS, 38677, 1983. (AD-A133747)

Bauer, H.-J., and Bass, H.E., Phys. Fluids, 16, 988, 1973.

Blackshear, P.L., Jr., Fourth Symposium (International) on Combustion, p. 553, Williams and Wilkins, 1953.

Blythe, P.A., J. Fluid Mech. 37, 31, 1969.

Blythe, P.A., Seventeenth Symposium (International) on Combustion, p. 909, The Combustion Institute, Pittsburgh, Pa., 1979.

Brinkley, Jr., S.R. and Richardson, J.M., Fourth Symposium (International) on Combustion, Williams and Wilkins Co., p. 450, 1953.

Chu, B.T., Pressure Waves Generated by Addition of Heat in a Gaseous Medium, NACA Tech. Note 3411, 1955. (AD-063030)

Chu, B.T., Stability of Systems Containing a Heat Source - The Rayleigh Criterion, NACA RM 56 D27, 1956. (AD-099348)

Chu, B.-T., Acta Mechanica 1, 215, 1965.

Chu, B.T., Wave Propagation in a Reacting Mixture, Proc. of Heat Transfer and Fluid Mechanics Institute, p. 80, Stanford University Press, Stanford, Ca., 1958.

Chu, B.T., Weak Nonlinear Waves in Nonequilibrium Flow, in: Nonequilibrium Flows, II, p. 33, ed. P.P. Wegener, Dekker, New York, 1970.

Clarke, J.F., Comb. Sci. and Tech., 7, 241, 1973.

Clarke, J.F., Quart. J. Mech. Appl. Math. 27, 161, 1974.

Clarke, J.F., J. Fluid Mech. 81, 257, 1977.

Clarke, J.F., Acta Astro. 5, 543, 1978a.

Clarke, J.F., J. Fluid Mech. 89, 343, 1978b.

Clarke, J.F., J. Fluid Mech. 94, 195, 1979.

Clarke, J.F., Combustion in reactive systems, J.R. Bowen, et al., eds., Prog. Astro. Aero. 76, 383, 1981.

Clarke, J.F., Ann. Phys. 9, 211, 1984.

Clarke, J.F., and McChesney, M., The Dynamics of Real Gases, p. 182, Butterworths, Wash., D.C., 1964.

Clarke, J.F. Kassoy, D.R., and Riley, N., Proc. R. Soc. Lond, A393, 309, 1984.

Clarke, J.F. Kassoy, D.R., and Riley, N., Proc. R. Soc. Lond, A393, 331, 1984.

Edwards, D.H., Thomas, G.O., and Williams, T.L., Comb. Flame 43, 187, 1981.

Einstein, A., Sitzber. Deut. Akad. Wiss. Berlin, Kl.-Math.-naturn., 380, 1920.

Ellis, R.J., and Gilbert, R.G., J. Acoust. Soc. Am. 62, 245, 1977.

Erpenbeck, J.J., Phys. Fluids 8, 1192, 1965.

Fickett, W., Wood, W.W., Phys. Fluids 9, 903, 1966.

Fickett, W., Davis, W.C., Detonation, University of California Press, Berkeley, 1979.

Garris, C.A., Toong, T.-Y., and Patureau, J.-P., Acta Astro. 2, 981, 1975.

Gaydon, A.G., and Wolfhard, H.G., Flames: Their Structure, Radiation and Temperature, 4th edition, pp. 177-194, John Wiley and Sons, N.Y., 1979.

Gilbert, R.G., Hahn, H.-S., Ortoleva, P.J. and Ross, J., J. Chem. Phys. 57, 2672, 1972.

Gilbert, R., Ortoleva, P., and Ross, J., J. Chem. Phys. 58, 3625, 1973.

Kailasanath, K., Oran, E. and Boris, J., Combust. Flame 47, 173, 1982.

Kailasanath, K., and Oran, E.S., Comb. Sci. Tech. 34, 345, 1983.

Kailasanath, K., and Oran, E.S., and Young, T.R., The Relation between Power and Energy in the Shock Initiation of Detonations. II. Application to Hydrogen-Air Mixtures, in preparation, 1984.

Kaskan, W.E., Fourth Symposium (International) on Combustion, p. 575, Williams and Wilkins, 1953.

Kuo, K.K., and Summerfield, M., eds., Fundamentals of Solid-Propellant Combustion, Prog. Astro. and Aero. 90, AIAA, 1984.

Landau, L.D., and Lifshitz, E.M., Fluid Mechanics, p. 245, Pergamon Press, London, 1959.

Lick, W., Adv. Appl. Mech. 10, 1, 1967.

Melvin, A., Moss, J.B., and Clarke, J.F., Combust. Sci. Tech. 4, 17, 1971.

Markstein, G., in Rudinger, G., Combustion and Propulsion, Third AGARD Colloquium, p. 153, Pergamon Press, New York, 1958.

Oran, E.S., and Boris, J.P., Prog. Ener. Comb. Science, 7, 1, 1981.

Oran, E.S., and Boris, J.P., Combust. Flame 48, 149, 1982.

Oran, E.S., Young, T.R., Boris, J.P. and Cohen A., Combust. Flame 48, 135, 1982.

Oran, E.S., Young, T.R., Boris, J.P., Picone, J.M., Edwards, D.H., 19th Symposium (International) on Combustion, p.573, The Combustion Institute, Pittsburgh, PA, 1982.

Patureau, J.-P., Toong, T.-Y., and Garris, C.A., Sixteenth Symposium International) on Combustion, p. 929, The Combustion Institute, Pittsburgh, Pa., 1977.

Picone, J.M., Oran, E.S., Boris, J.P., and Young, T.R., to appear in Prog. Astro. Aero., 1984.

Putnam, A.A., General Considerations of Autonomous Combustion Oscillations, in Nonsteady Flame Propagation, G.H. Markstein, MacMillian, N.Y., 1964a.

Putnam, A.A., Experimental and Theoretical Studies of Combustion Oscillation, in Nonsteady Flame Propagation, ed. G.H. Markstein, MacMillian, N.Y., 1964b.

Putnam, A.A., and Dennis, W.R., Trans ASME 75, 15, 1953a.

Putnam, A.A., and Dennis, W.R., Fourth Symposium (International) on Combustion, p. 566, Williams and Wilkins, 1953b.

Rayleigh, J.W.S., The Explanation of Certain Accoustical Phenomena, Nature 18, 319, 1878; also, Theory of Sound, Vol. II, , p. 226, reprinted by Dover, 1945.

Srinivasan, J., and Vincenti, W.G., Phys. Fluids, 18, 1670, 1975.

Strehlow, R.A., and Cohen, A., Phys. Fluids 5, 97, 1962.

Strehlow, R.A., Fundamentals of Combustion, Chapter 9, Krieger, New York, 1979.

Toong, T.-Y., Combust. Flame 18, 207, 1972.

Toong, T.-Y., Salant, R.F., Stopford. J.M., and Anderson, G.Y., Tenth Symposium (International) on Combustion, p.1301, The Combustion Institute, Pittsburgh, Pa., 1965.

Toong, T.-Y., Arbeau, P., Garris, C.A., and Patureau, J.-P., Fifteenth Symposium (International) on Combustion, p. 87, The Combustion Institute, Pittsburgh, Pa., 1974.

Williams, F.A., Combustion Theory, Addison Wesley, Reading, Mass., 1965.

Wood, A., Acoustics, Dover, New York, 1966.

DEPARTMENT OF THE NAVY

NAVAL RESEARCH LABORATORY
Washington, D C 20375-5000

OFFICIAL BUSINESS
PENALTY FOR PRIVATE USE, $300

POSTAGE AND FEES PAID
DEPARTMENT OF THE NAVY
DoD-316
THIRD CLASS MAIL

U.S.MAIL ®

NRL

THE NAVAL RESEARCH LABORATORY IS AN
ACTIVITY OF THE OFFICE OF NAVAL RESEARCH