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A STUDY OF DETONATION STRUCTURE: THE FORMATION OF UNREACTED GAS--ETC(U)
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Schlieren photographs of detonations in low pressure H_2-O_2-Ar mixtures and numerical simulations of propagation detonations have revealed the presence of unreacted pockets of gas behind the shock front — Mach stem structure. These pockets are completely surrounded by burned gas, and they in turn burn more slowly, finally giving their energy to the system. Two-dimensional numerical simulations performed to study the development of unburned pockets

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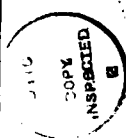1 JAN 73
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20. ABSTRACT (Continued)

show that they are peninsulas of cooler material cut off by the collision of two Mach stems or of a Mach stem and the wall. Their presence is observed when there is sufficient decoupling of the reaction zone from the incident shock. These unreacted gas pockets and the associated long induction distance are discussed in terms of possible mechanisms for cell reinitiation, cell generation, and the behavior of detonations near the detonation limits.

CONTENTS

A STUDY OF DETONATION STRUCTURE:
THE FORMATION OF UNREACTED GAS POCKETS

I. INTRODUCTION

It is currently recognized that the propagation of a detonation is a very complex, three-dimensional process involving interactions between shock fronts, Mach stems, reflected shock waves, and the boundaries of the system through which the detonation is moving.[1,2,3] This knowledge, which showed that more than the steady state Chapman-Jouget (C-J) theory was needed to predict the detailed structure or transient behavior of detonations, came about through both theory and experiment. Theory has shown that even a one-dimensional propagating detonation is unstable for any significant amount of energy release, and that chemical energy release couples nonlinearly with and may amplify acoustic and nonlinear perturbations.[4,5,6,7] The experimental interpretation of the carbon soot tracings and open shutter photographs showed that a propagating detonation leaves a very regular, cell-like pattern on the sidewalls of the confining chamber.[1,8] From studies of these patterns, it was determined that they were etched by the triple point formed at the front of the detonation by the intersection of a Mach stem and an incident shock wave, and thus the cell patterns are histories of the locations of the triple point.

The current picture which has emerged is that the transverse wave, which is the reflected shock intersecting the Mach stem and the incident shock, is crucial in the formation and propagation of a detonation. The incident shock is not steady, but continuously decaying, and the transverse waves shuttle back and forth along the front. The cell, however, is re-initiated when collisions occur between transverse waves moving in the opposite directions.[2] As the cell is formed, the shock wave and the reaction zone are close and the leading shock velocity ranges from 1.2 the C-J value for normal detonations to 1.8 for marginal detonations. As the waves decay, there is more and more distance between the reaction zone and the shock front, so that the velocity of the leading shock may drop to as low as 0.85 the CJ value for normal detonations and 0.6 for marginal detonations. At the end of the cycle, however, the velocity jumps discontinuously as there is a collision of transverse waves, and the reaction zone and shock wave are close again.

To date, the process by which the cell structure is self-sustaining has still not been completely explained. Acoustic amplification through coupling with chemical energy release may be important in the re-initiation process as well as in determining the cell size. We know that the natural cell size is an important basic property of a mixture and related to detonation limits[9] and that current theories can predict this to within a factor of two.[10] It is also known that the transverse waves extend back into the reactive flow regime, although the interaction between the Mach stem and the incident shock right at the front are non-reactive gasdynamic phenomena.[1] The temporal behavior of the Mach stem is determined by a complicated nonlinear interaction among the chemical energy release, the expansion and flow, and the interaction of various waves immediately behind the shock front.[11]

In principle numerical simulations can be used to determine quantitatively the properties of detonation cells. Certainly adequate multidimensional techniques now exist for calculating the complicated interaction of shock waves undergoing either single or double Mach reflections.[12] We also know how to

couple detailed chemical kinetics to models of the fluid dynamics for supersonic flows.[13] For hydrogen mixtures, the complete calculations could be done, given the resources.[14] The problem, however is two-fold. First, it would involve series of extremely expensive calculations which would be near the limits of our computational power. Second, if the calculations were simply done brute-force, we would be inundated with numbers and it might be difficult to unravel the underlying controlling physical processes.

With these problems in mind, we have taken an intermediate approach in this paper, which presents an extension and illumination of the work we alluded to in the 18th Combustion Symposium[15] and also mentioned by Book et al.[12] There we observed the formation of detonation cells in the two-dimensional calculations in hydrogen-air and methane-air mixtures in which we used an induction parameter model to incorporate the effects of chemical kinetics. Here we shall examine in detail the simplest type of transverse wave intersection which occurs in the planar mode: that is, a case which is a purely transient two-dimensional problem simulating a half-cell in a detonation tube. In this case the shock interactions are always single Mach interactions and the path of the intersections is in the detonation propagation direction. Two-dimensional behavior is approximated experimentally when a detonation is passed through a wide, straight channel which has a depth of 1/6 to 1/10 of the preferential wave spacing.[1]

Two-dimensional simulations of the behavior of self-sustaining detonations are also being performed by Taki and Fujiwara[16] whose goal has been to determine the natural transverse wave spacing and thus the detonation cell size. Their calculations, however, must be considered qualitative in the current context for three reasons. First, as they discuss, their numerical method included an artificial viscosity term which adds numerical diffusion, thus smearing shock fronts and losing the resolution crucial to studying complicated Mach stem structures.[12] Second, their induction time phenomenology does not include the anomalous behavior in the region of the H_2-O_2 second extended limit. If, in fact, cell sizes depend on the induction distances, this quantity must be accurately represented. Third, they have chosen to concentrate on the multi-cell problem with a concomitant need to relax local resolution in order to represent 3-5 half detonation cells across the system. The work described in this paper resolves only one half-cell. In turn we can still represent details of the reaction wave-shock interactions when the induction distances are much shorter than average C-J values.

The case studied in detail in this paper is for a hydrogen-oxygen-argon mixture near marginal conditions in which a detonation is initiated by a shock having just about enough energy to ignite the mixture. That is, initially it is only slightly overdriven and decays quickly to the self-sustaining conditions. The interesting feature which we study in the calculation is the formation of unreacted pockets of gas within the cell which are completely surrounded by reacted material. It is shown how these pockets are formed as they are cut off by the interaction of transverse waves. Such pockets have been observed by Subbotin[17] as well as by Edwards and here we present some of the latter observations. Based on the results of the simulations we discuss one possible mechanism which could contribute to cell re-initiation in marginal detonations. This is based on the effect of the transverse wave on a previously shocked but unreacted medium near the incident shock. Finally, we examine the possibility that the delayed burning of the observed unreacted gas pockets might be an origin of new cells.

II. NUMERICAL MODEL

The numerical model used to perform the calculations described below solves the time-dependent conservation equations for mass, momentum and energy[13] in two dimensions using one variant of the FCT algorithm.[18] This model with various initial and boundary conditions has been used extensively to study single and double Mach reflections,[12] mixing and vortex formation at material interfaces,[19] and the effects of the Rayleigh-Taylor instability on the surface of pellets imploded by lasers.[20] Applications[12] of this model to the study of propagating shocks colliding with wedges and to height-of-burst

problems are particularly relevant to this paper. Extensive comparisions to experimental data have shown that the calculations reproduce experimentally observed quantities to within experimental accuracy. Furthermore, the double or single Mach reflection shock structures are well-resolved.

In addition to the solution of the convection terms, we have included an induction parameter model which is a phenomenology developed to reproduce the correct ignition delays and subsequent energy release due to chemical reactions.[14,15] The convection and energy release algorithms are coupled by methods described by Oran and Boris.[13] The adaptive gridding method used in the calculations[13,15] consists of fine zones in the x direction which surround and move with the detonation or shock front and encompass the transition region behind it. The remaining cells in the x direction are evenly spaced, except that a smooth transition from fine to coarse zones is effected in the x direction. The cells in the y direction are evenly spaced. This is shown schematically in Fig. 1.
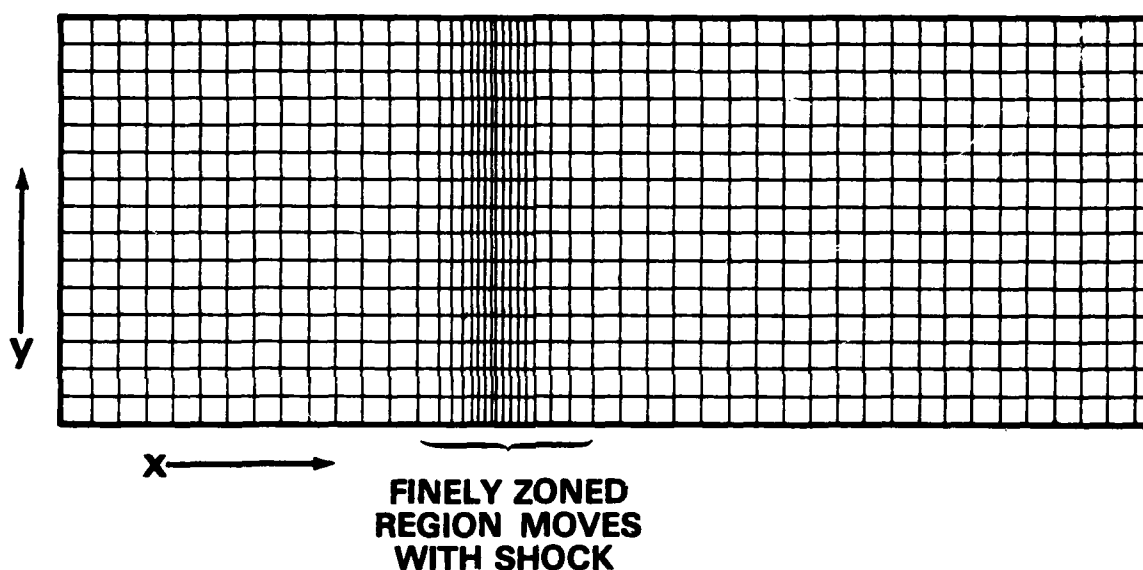

**FINELY ZONED
REGION MOVES
WITH SHOCK**

Fig 1 — Schematic of the two-dimensional computational grid used in the detonation simulations. For the cases described, $\Delta y = \Delta x_{min}$ and Δx_{max} could be as much as an order of magnitude greater than Δx_{min}. Note then the size of the zones transions smoothly in the x-direction.

Calculations of reactive shocks in one-dimension have been done using a detailed model of the hydrogen-oxygen chemistry. These have proved very useful and accurate in predicting phenomena which are intrinsically one-dimensional in character and also for studying the interaction of gas dynamics and chemical energy release with realistic chemical models.[7,21] However as has been noted,[15] it is currently possible but exorbitantly expensive to include a detailed chemistry in a two-dimensional model. Thus in order to be able to at least approximate the effects of gas dynamics and energy release in multidimensional models, we require simplified or phenomenological models to represent the chemical kinetics.[14,15,22,23]

The simplest model we have considered to date is one in which three quantities must be specified: the time before any energy is released τ (the chemical induction time), the time it takes to release the energy, and the final amount of energy released. As shown by Strehlow[1], we can then define a quantity, $I(t)$.

$$I(t) = \int_0^t \frac{dt'}{\tau(T,P)}$$

3

which is a measure of how long the material has remained at a given temperature and pressure. In the calculations this quantity is convected with the fluid and it is used to indicate when the available chemical energy should be released. That is, when $I(t) = 1$, energy release begins by specifying how much fuel is converted to product. No matter how fast the energy release rate is, the reaction is *not* assumed to go immediately to completion.[15] That is, the energy is released over several timesteps determined by the ratio of the sonic transit time to the timestep itself. The purpose for doing this is to keep a whole computational cell from burning at once and thus eliminate numerical "surging."

Initially we attempted to find an analytic form for $\tau(T,P)$, the chemical induction time. Since then we have determined that this involves a great deal of needless work and is not nearly as accurate as using the extremely fast, vectorized table look-up algorithms recently developed by T. R. Young at the Naval Research Laboratory. Induction times for homogeneous mixtures of $H_2:O_2:X/2:1:4$ over a range of temperatures and pressures have been calculated[7] by integrating the set of ordinary differential equations based on the chemical rates given and calibrated by Burks and Oran[24]. Here X is the diluent and may be either argon or nitrogen. These rates can be converted conveniently to other dilutions through the formula

$$\tau(2:1:X) = R(2:1:X)\,\tau(2:1:4),$$

where

$$R(2:1:X) = \frac{3 + X}{7}.$$

This formula has been tested extensively against detailed integration of the chemical rate equations and works well for the cases considered below. A generalized form for variable stoichiometry has also been derived and some testing has been done.[7]

III. EXPERIMENTAL RESULTS AND NUMERICAL SIMULATIONS

Figure 2 consists of excerpts taken from a series of schlieren photographs of detonations propagating in stoichiometric mixtures of hydrogen and oxygen with 60% argon in a pressure range from 50-80 Torr. The tube in which the detonations propagated was 3" high and 1/4" wide. This narrow width was chosen purposefully so that any transverse disturbances orthogonal to the main mode were suppressed and thus the detonations were essentially two-dimensional. The appearance of the pockets of unburned gases behind the detonation front were noted to be a common feature in these relatively lean mixtures in which we see one or two transverse waves in the tube. This effect has also been observed by Subbotin[17] in experiments in mixtures of $H_2:O_2:Ar/2:1:3$ at pressures under 100 Torr and he has speculated that such unreacted gas pockets occur only in marginal detonations. The experiments performed at Aberystwyth were used as a guide for the calculations presented below.

A number of calculations were performed in which we varied physical parameters such as system size, method of initiation and energy input, and calculational parameters such as maximum and minimum zone size. The calculations were performed on a 150×50 Cartesian grid. To permit spatially extensive calculations, a region of 50 fine zones of size Δx_{min} (Fig. 1), centered on the shock and extending back through the reaction zone, were embedded in a grid of 100 coarse cells of Δx_{max}. The size of the zones in the y-direction was the same as those of the fine zones in the x-direction. Thus the total system length could, for example, be 40 cm whereas the fine zones permit resolutions of 0.05 cm.

The calculations described below take advantage of the fact that there is a line of symmetry through the center of a detonation cell. Thus, (as shown in Fig. 7) we can make an analogy between a triple point collision with a wall and a triple point collision with another triple point. This is certainly
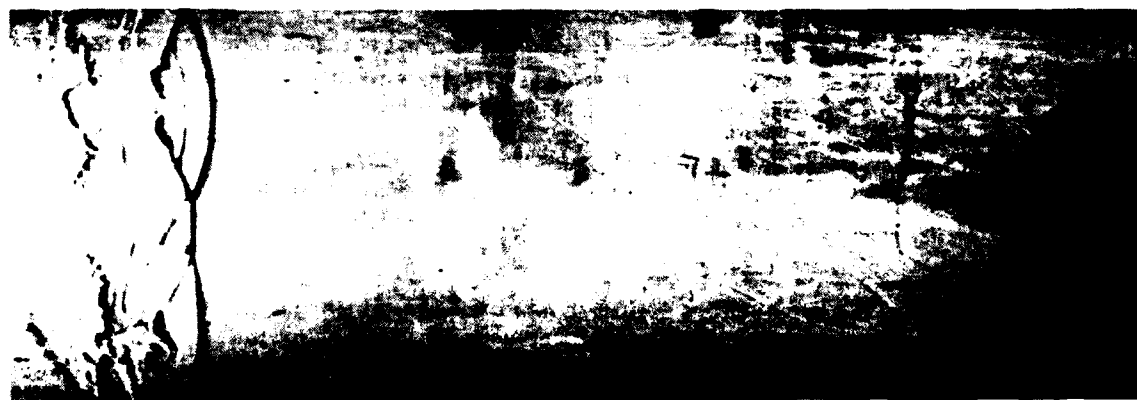
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Fig. 2 — Three schlieren photographs taken from a series of experiments in a tube 3" high and 1/4" wide filled with stoichiometric H_2-O_2 mixtures in 60% Ar at pressures from 50-80 Torr. Unburned pockets of gas are observed behind the detonation fronts.

true in the ideal, two-dimensional cases studied in this paper though it may not hold for more complicated three-dimensional structures. Studying only a half cell in detail allows us to double the computational resolution and focus on details of the structure.

Two methods of detonation initiation were used: the first assumed that an oblique shock oriented at a given angle and with a given Mach number exists initially at the left side of the tube.[15] The second method assumed there was a spherical shock of a given radius and Mach number in the bottom left hand corner to the tube. In both cases the induction parameter behind the shock was set to 1.1, which means that energy was quickly released behind the shock front, and to 0.0 for unreacted material. When the chosen Mach number was too low, the shock propagated forward and a slow reaction zone was left far behind. When the Mach number was too high, a highly overdriven detonation was formed that took some time to decay. In these latter cases, the reaction zone was compressed tightly against the shock front and there was not enough resolution to resolve the unburned pockets, if they occur, until the detonation had decayed substantially. Thus the optimum Mach numbers for the purposes of the present paper were those critical values which provided just enough energy to permit propagating detonation. As expected, the spherical initiations required considerably higher initial Mach numbers than the oblique shock initiations, although quantifying this difference was not the goal of this paper. However, when the initial Mach numbers caused detonation, the resulting downstream structure of the shock front — reaction zone complex were the same. Thus we now feel confident that the results discussed below are not an artifact of the initial conditions. Figures 3, 4, and 5 are a sequence of computer-generated contours of the pressure, temperature and induction parameter for a calculation initialized with a Mach 4 shock propagating at a 35° angle in a 65 Torr, 298K, stoichiometric hydrogen-oxygen mixture diluted with 60% argon. The tube was 2.5 cm high and 41.25 cm long, giving a $\Delta y = \Delta x_{min} = 0.05$ and $\Delta x_{max} = 0.5$ cm. The figures shown were taken after the transverse shock had bounced a few times so the shock — reaction zone complex had become essentially independent of initial conditions.

These figures show a number of interesting and perhaps surprising features. Most obvious is the formation of the unburned pocket, shown by the sequences in Fig. 5. As the triple point and Mach stem approach the upper wall, the reaction zone falls further and further behind the incident shock front. This can be seen by comparing the position of the incident shock at cycle 1300 from Fig. 3 to the corresponding induction parameter contours in Fig. 5. Eventually the collision of the wall and the triple point cuts off the more slowly reacting gas pocket which burns later.

The calculated range of shock velocities at the upper and lower wall is from 1200 to 2600 m/s, which is 0.7 to 1.5 of the C-J value. The transverse cell spacing estimated from the work of Strehlow and Engel[25] is about 3.8 cm. Thus we see that the system modelled is slightly larger than a half cell, but not large enough for a full cell to form. It is encouraging, however to see effects in the calculations which are probably indications of the system's efforts to develop the full cell pattern. These appear as both an occasional extra kink in the shock front — reaction zone structure and by anomalous, elongated behavior in the horizontal cell length. Detailed studies of these properties, however, will be left for future work.

In order to study the unreacted gas pockets in more detail, a more highly resolved calculation was performed on a smaller system with the same gas conditions. If the height of the system is less than a half-cell, the half-cell structure is forced. Thus Fig. 6 shows a sequence of computer-generated contours of the temperature and induction parameter for a system of the same mixture as discussed above but in a tube of height 0.5 cm. Here $\Delta y = \Delta x_{min} = 0.01$ cm and $\Delta x_{max} = 0.1$ cm. The formation of the unreacted gas pocket as it is cut off by the reflection of the triple point is clear in the last frames, at cycle 1100. The temperatures in the pocket are lower than the surrounding temperatures and the pressure is in the 1-2 atm range. Such pockets provide a source of energy that is released behind the shock front.
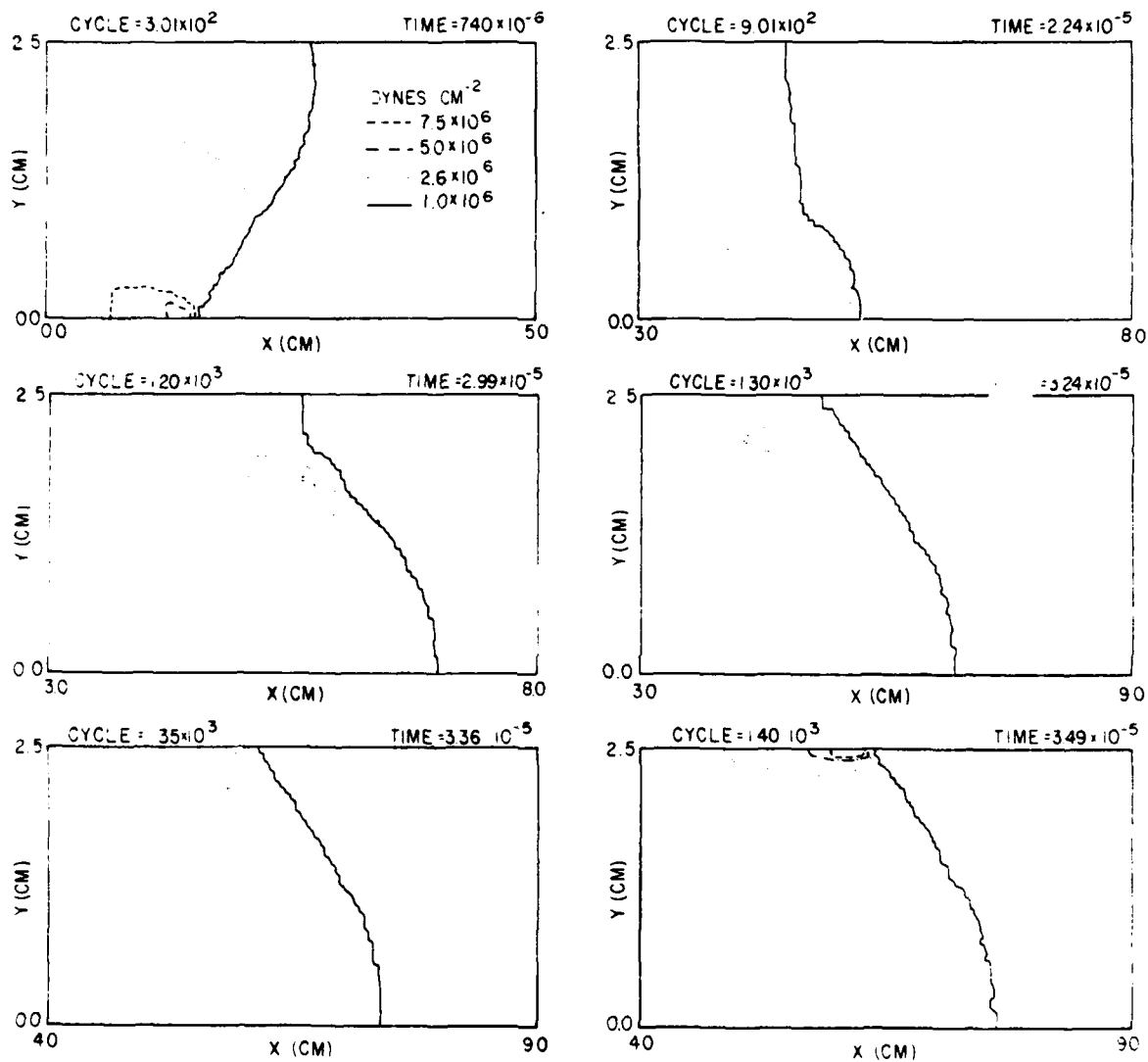
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PRESSURE CONTOURS

Fig. 3 — Pressure contours at selected times from a calculation of a detonation propagating in a stoichiometric H_2-O_2 mixture at 65 Torr and 298K in a tube 2.5 cm high. The legend is in the upper left hand box. The ragged edges of the surfaces are an artifact of the computer software.
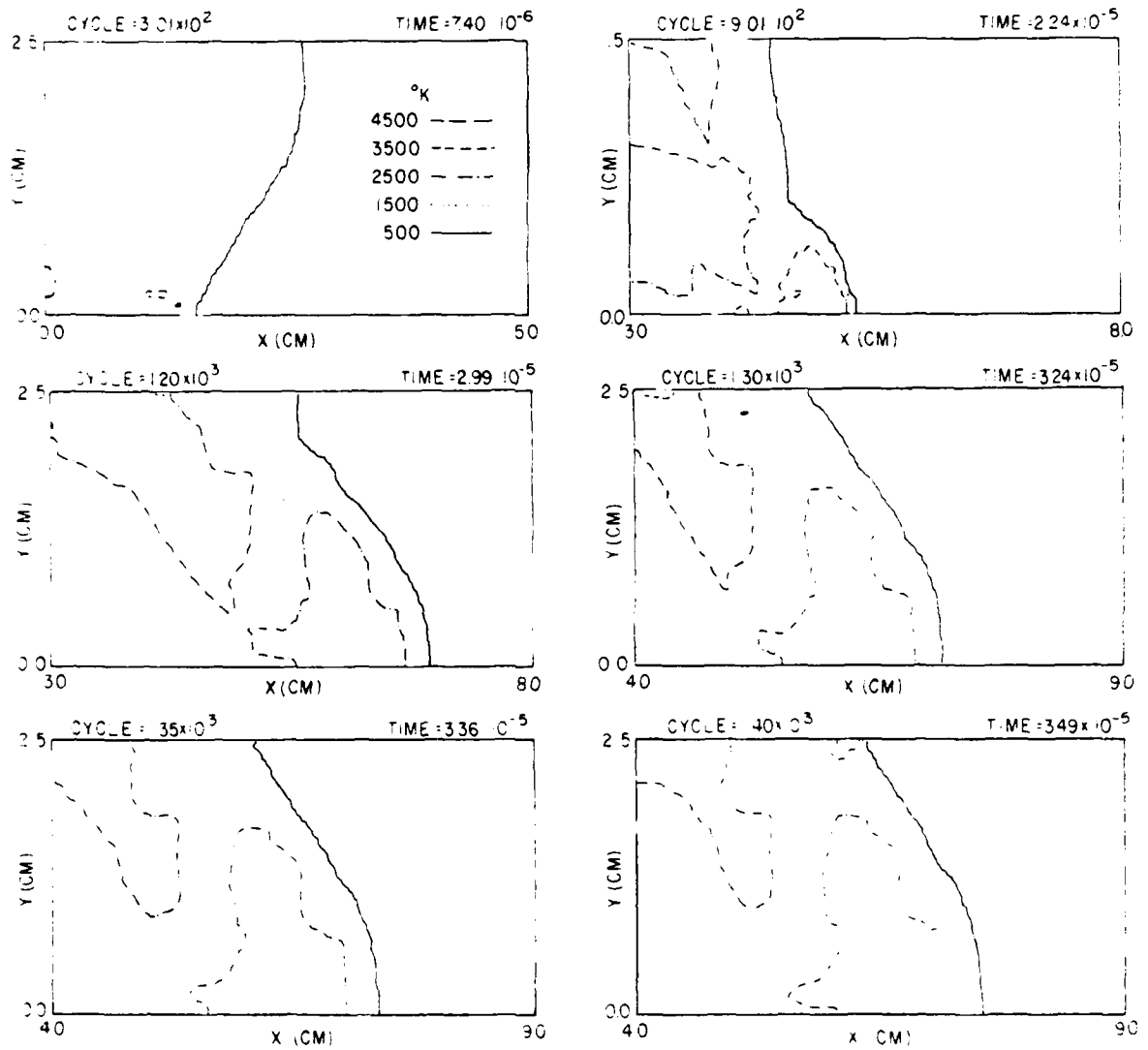
TEMPERATURE CONTOURS

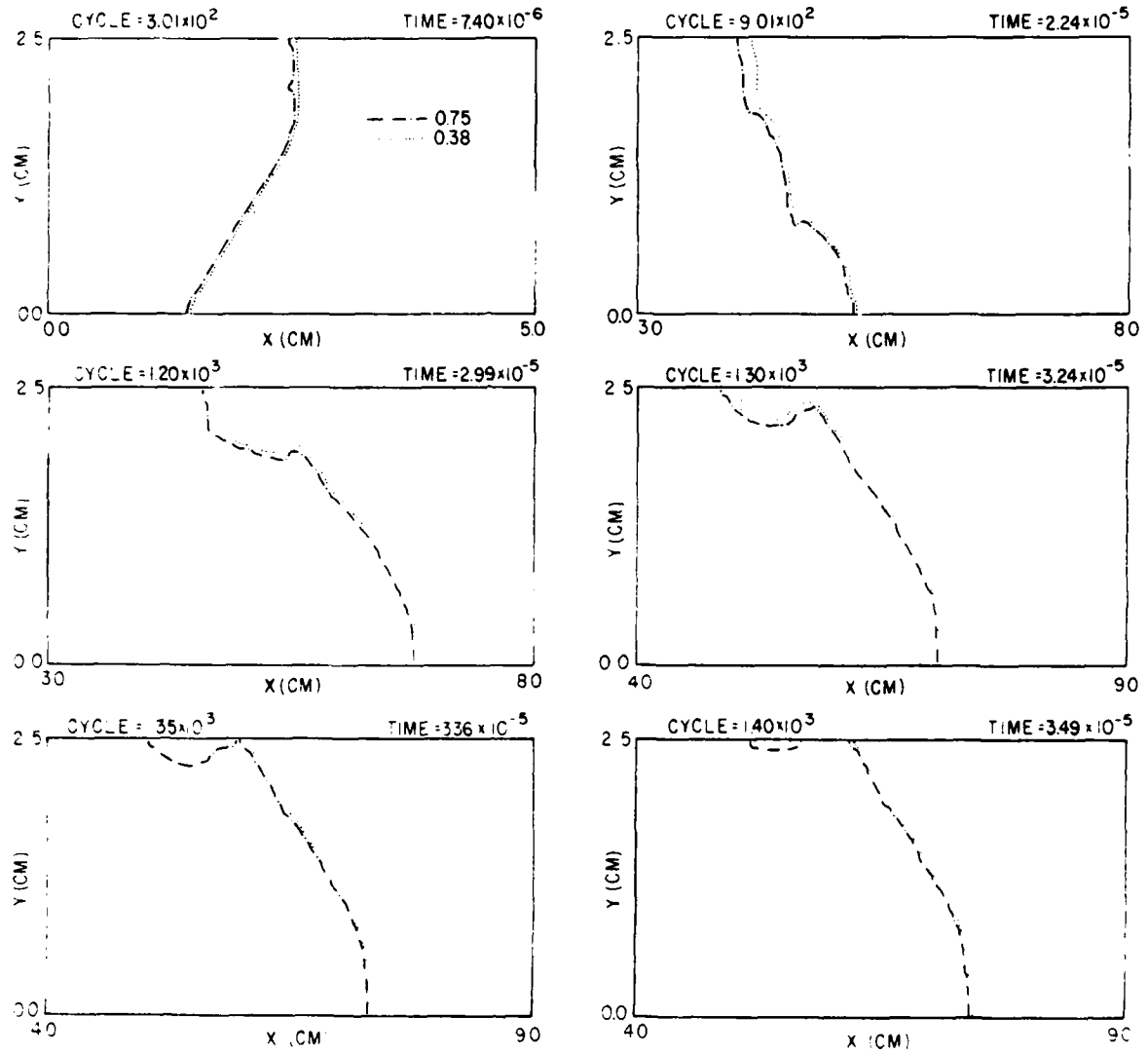Fig. 4 — Temperature contour corresponding to Fig. 3

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INDUCTION PARAMETER CONTOURS

Fig 5 — Induction parameter contours corresponding to Fig. 3.

9

INDUCTION PARAMETER CONTOURS

TEMPERATURE CONTOURS

CYCLE = 9.01×10^2 TIME = 1.37×10^{-5}

0.01 ————
0.26 ————
0.51 —·—·—
0.76 ········
1.01 ————

CYCLE = 9.01×10^2 TIME = 1.37×10^{-5}

°K
4500 ————
3500 ————
2500 —·—·—
1500 ········
500 ————

CYCLE = 1.00×10^3 TIME = 1.56×10^{-5}

CYCLE = 1.00×10^3 TIME = 1.56×10^{-5}

CYCLE = 1.10×10^3 TIME = 1.73×10^{-5}
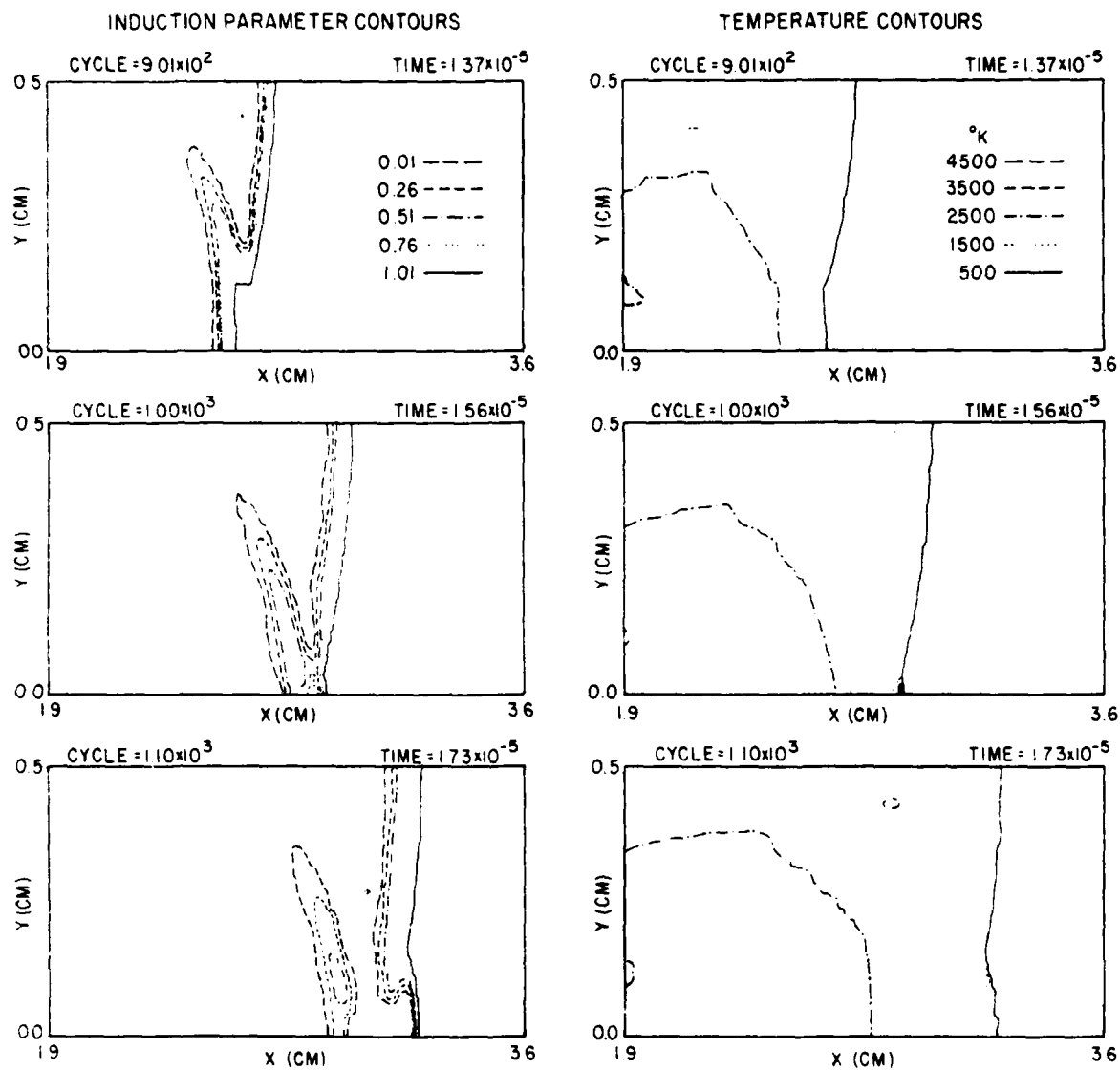
CYCLE = 1.10×10^3 TIME = 1.73×10^{-5}

Fig. 6 — Temperature and induction parameter contours from a calculation for the same mixture
as in Figs. 3, 4, and 5 except in a tube 0.5 cm high.

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IV. DISCUSSION

The formation of the unburned pockets was first reported by Subbotin,[17] who explained that collisions between waves in which the reaction zone has fallen sufficiently behind results in the formation of pockets of unburned gas and an increase in the extent of the combustion zone. As we have seen from Figs. 3-5, these pockets are in a region of relatively high pressure (1-2 atm) and relatively low temperature (900-1000K). Thus they are in an extremely "sensitive" region, meaning that their ignition is not controlled so much by the induction time of the material as by the fluctuations and perturbations occuring in it. In such a dynamic medium as that around the pocket, we have shown that sound wave fluctuations will play an important part in determining their disappearance rate.[7]

Consider cycle 901 in Fig. 6. From this diagram we can see that there are two ways in which the pockets could be formed. First, let v_α and v_β be the velocities at which burning occurs horizontally and vertically across the unburned peninsula. Then if $v_\beta/v_\alpha > H(1 + \delta)/\lambda$, where H is the height of the system, δ is a small positive quantity which accounts for curvature and λ is the induction distance from the incident shock, a pocket could be cut off. Considering the temperature and pressures in the material near v_β, it is likely that it will be higher than v_α. Thus, when λ becomes large enough in the simple formula above, the condition for forming gas pockets will eventually be satisfied. Since marginal detonation is generally characterized by very long induction lengths during at least some of the cycle, we expect gas pockets to become prominant as detonation becomes marginal. However we can also imagine situations in which the temperatures and pressures in the pockets are so low that reaction in them is substantially delayed. This would be a case very close to or just below the detonation limits, where the detonation could propagate raggedly and eventually die out.

We can understand a second way in which an unburned pocket could be cut off by considering a situation in which the triple point is close to the lower wall and the reaction zone has fallen way behind. Thus the area associated with λ is quite large. If the transverse wave first intersects the unreacted, pre-shocked material below it closer to the triple point than to the reaction wave, it could ignite a portion near the triple point first. Then a pocket of unburned gas could be cut off before or when the triple point collides with the wall. This process is illustrated schematically in Fig. 7 in which the top portion is a schematic of what happens when transverse waves collide, and the lower is a schematic of the interaction of the transverse wave with a wall. Just before the transverse wave collides with either another transverse wave or with a wall, there is a triangular region of unreacted gas, OAB. After collision, a reaction occurs behind the reflected transverse wave which removes a semicircular portion, O_1O_2, out of the triangle. The unburned pocket detaches from the front and burns downstream. However, both mechanisms are quite similar in that they depend on a substantial induction zone implying substantial decoupling of the shock front and reaction zone.

Ignition of an induction zone by a transverse wave could, perhaps, account for the observations of Liboutin, Dormal, and Van Tiggelen.[26] Their recent work has addressed this basic problem of the reinitiation process in detonation cells by focussing on the chemistry occurring within a cell in a low-pressure, stoichiometric hydrogen-oxygen-argon mixture. Their measurements of OH emission intensity shows that there is a re-initiation mechanism starting about three-quarters of the way down the cell, corresponding to a measured increase in pressure of the transverse shock at the cell center. Based on calculations of the incident shock decay alone, however, the induction times would increase so greatly that chemical reactions would not occur in the material after this distance within the cell. Thus they postulate that some unspecified mechanism is generating pressure pulses at about a distance of 0.7 of the cell length which eventually form a shock and reignite the mixture at the end of the cell.

This effect, in which a transverse wave ignites pre-shocked, essentially prepared material has an interesting implication for cell reinitiation. When the transverse wave reaches the preshocked material, it will ignite it and send a reaction wave in every direction. Thus if we are along the centerline of a detonation cell and a reaction wave is ignited somewhere in the induction zone, we would observe a
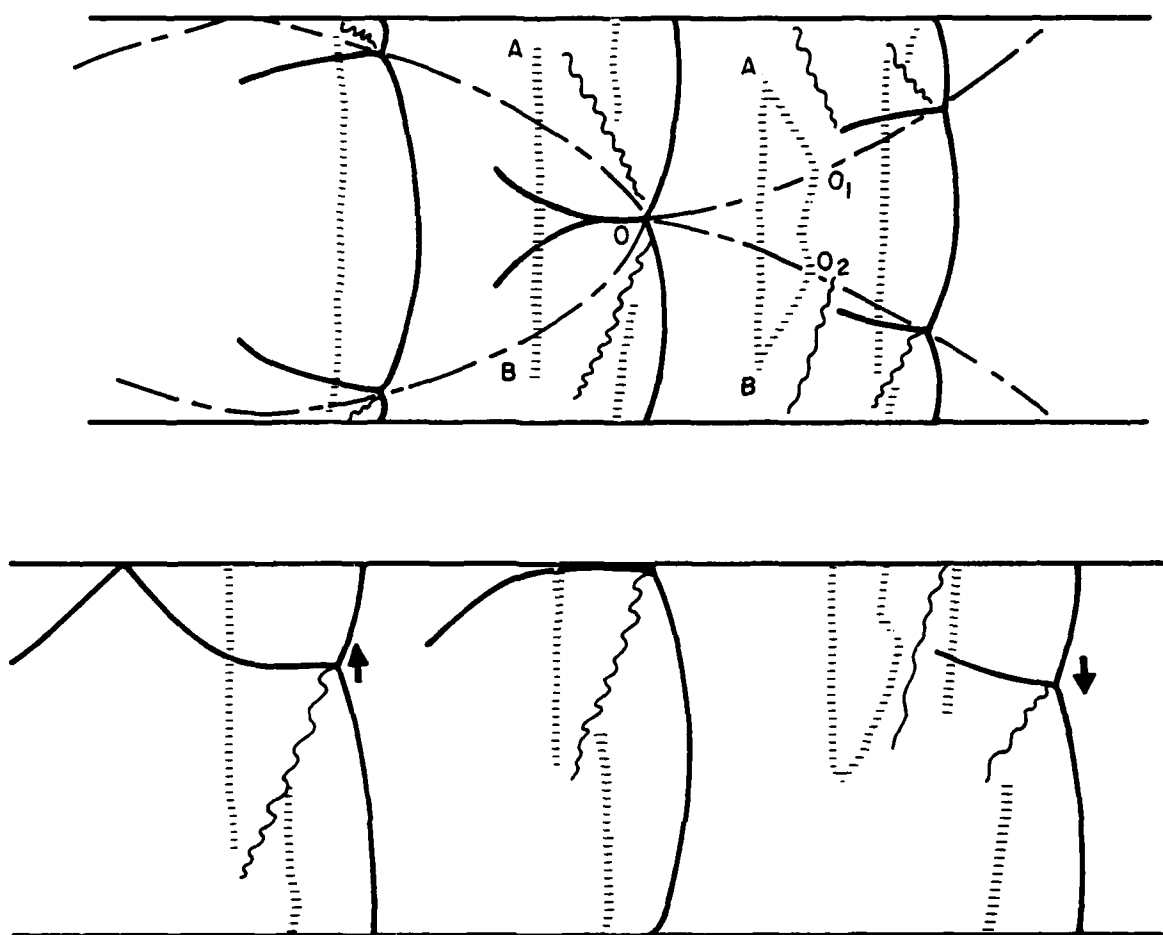
Fig. 7 — Schematic showing how the interaction of transverse waves (upper figure) or a
transverse wave and a wall (lower figure) can cut off an unreacted gas pocket.

sudden rise in pressure followed by relatively sudden energy release which would propagate like a mini-detonation structure through the induction zone. This looks very much like a reinitiation mechanism because it could bring the reaction zone and the shock front very much closer together to form kinks in the incident shock before or during collision with the next transverse wave. Such an effect is more likely in marginal detonations in which the shock velocity may fall well below the C-J value.

Another interesting aspect is how the unburned gas pockets are actually consumed. From both the experiments and the simulations discussed above, we see that they eventually react and their energy is not lost to the cell. However, there are several ways in which they could burn: either as a relatively slow energy release or as a fast reaction wave. Thus as they release energy, they could send pressure pulses, perhaps even a shock, forward to the shock front. Such a perturbation, as noted above, is a mechanism for generating new cells.

V. CONCLUSIONS

Numerical simulations have been extremely useful in delineating features of detonation structure. However, there are two requirements if the simulations are to be quantitatively as well as qualitatively

correct. First, adequate resolution around the detonation front is required, or many potentially important phenomena such as these unreacted gas pockets will not be seen and the results will be invalidated by numerical diffusion. Second, relatively realistic models of the chemical kinetics seem to be required in order to model faithfully the reaction zone-shock front structure. The induction parameter model used here currently seems adequate, but it is not clear yet if such a simplified model can always give quantitatively correct answers.

There are a number of interesting observations from the calculations presented above which require further study. A more detailed study of the shock front — Mach stem — transverse wave structure should delineate the role played by the transverse wave in the induction zone before the triple point is reflected. Modelling a larger system whose height is a cell size or greater would provide insight into how the natural transverse cell size develops. Studying the isolated, idealized problem of an unreacted pocket of material behird a two-dimensional planar detonation would allow us to see how and if its delayed burning effects the detonation front. By varying the composition of the mixture downstream, we could see how the structure changes and how the pockets grow or decrease in size with changes in dilution. Thus we feel that a systematic study of these problems would allow us to determine detonation cell sizes and better understand detonation cell structure.

ACKNOWLEDGMENTS

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