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DETONATION AND THE HYDRODYNAMICS OF REACTIVE SHOCK WAVES

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INTRODUCTION

The contents of this review may be conveniently separated into two rather distinct parts. On the one hand this review covers our current understanding of detonation as a unique natural phenomena, while on the other hand the review discusses the limitations that hydrodynamics places on the utility of the shock tube as a tool for studying high temperature reaction kinetics. Specific reaction kinetic studies are not covered because they are so extensive that it is difficult to do them justice in a short review. The reader is referred to recent more extensive reviews for this purpose¹⁻⁵.

DETONATION

I. Theories Relating to Structure and Stability

The Zel'dovich⁶, von Neumann⁷, Doring⁸ (ZND) model of a one-dimensional steady detonation wave as a shock discontinuity followed by a zone of homogeneous chemical reaction is probably the most useful concept to evolve prior to the mid 1950's. Even though this model must now be modified to accommodate non-steady and three-dimensional effects, its implication that an element of the flow may be treated as containing an exothermic chemical reaction triggered by a strong shock, which may in itself be handled as a discontinuity in the flow, represents a useful tool for understanding the structure and non-steady behavior of detonation waves.

The Chapman⁹-Jouguet¹⁰, (CJ), concept that a minimum steady one-dimensional mass flow exists for any specific exothermic system undergoing reactive shock transition is at present useful only insofar as it allows a rather exact calculation of the velocity and pressure in a detonation wave. Unfortunately, at the present time, this success must be considered to be either fortuitous or at least empirical because recent work has shown that a one-dimensional detonation traveling at the CJ velocity is dynamically unstable.

The first attempt to look at time-dependent stability in a detonation wave was made by Shchelkin¹¹ in 1958. He used a square wave detonation as his model. In this model the chemical reaction is concentrated at the CJ plane and this plane is separated from the shock wave by an induction zone. In his model, he perturbed the position of the CJ plane with time and discussed the consequences of this perturbation. He found that the detonation will be unstable to this type of perturbation if the induction zone reactions have a sufficiently large activation energy. This model is unrealistic because one cannot truly cause the CJ plane to suffer a perturbation, since it is a steady concept. However, Shchelkin was the first to discuss the effect of a time-dependent perturbation on stability.

Since 1963, there have been a number of papers¹²⁻¹⁹ which discuss the stability of the square wave detonation. However, recent work on detonation stability, using models which allow for an extended heat release

zone, show quite definitely that all the interesting amplification mechanisms which lead to instability occur in the region of heat release. Therefore, the square wave model appears to be unsuitable for discussing stability.

Two approaches have been used to investigate the stability of a ZND detonation wave with an extended reaction zone. Erpenbeck²⁰⁻²¹ has examined stability by following the response of the shock discontinuity to a purely transverse harmonic perturbation in the flow downstream of the shock wave (i.e., in the reaction zone). His analysis is applicable to disturbances of arbitrary wave length but requires a model for the heat release reactions throughout the entire reaction zone. He has not formally examined the CJ detonation case, but has always worked with detonations that have some arbitrary overdrive. His analysis, which is performed in Laplace transform space, is very complex and he finds that stability in the general case can be discussed only by resorting to numerical techniques. His results indicate that the occurrence of instability is dependent on the frequency of the disturbance, the activation energy of the first order heat release reaction that he has used in his model and the amount of the overdrive present in the detonation. In a following paper, Erpenbeck²² has extended his analysis to transverse waves of high frequency. In this case, he finds that there are three types of stability regimes which may be present in the reaction zone of the detonation. The type of regime that is present is dependent on the local value of the quantity $d(a^2-u^2)/dx$ where x is measured from the shock and a and u are the sound velocity and flow velocity at a point in the steady flow behind the shock. He observed that this quantity either increases, goes through a maximum, or decreases, in that order, as one passes from the shock wave to the plane where heat addition is terminated. If the quantity simply decreases in the reaction zone he finds that the detonation will be stable to a high frequency transverse wave. If this quantity goes through a maximum or if it increases, he finds that the detonation behaves unstably to a high frequency transverse perturbation. In the limit behavior which he investigated, the ultimate stability is found to be dependent on the activation energy of the first order chemical reaction which was assumed in the analysis. Specifically, systems with a sufficiently low activation energy were found to be stable.

In the other approach to the stability problem, Strehlow and Fernandes²³ and Barthel and Strehlow²⁴ have used a ray tracing technique to discuss the behavior of a high frequency coherent transverse acoustic wave in the reaction zone of a ZND detonation. They also find that the quantity $d(a^2-u^2)/dx$ is important to the behavior of these coherent transverse waves. For a transverse wave propagating in a flowing gas, the direction of propagation of the energy trapped in the acoustic front (i.e., the ray direction) is the direction which is of interest. They find that, in the region where the quantity $d(a^2-u^2)/dx$ is decreasing, an acoustic wave front of any orientation (except for one very specific orientation) subsequently passes through the reaction zone of the detonation at most only twice (due to reflection from the shock front). However a wave front of the proper orientation was found to asymptotically approach the plane where $d(a^2-u^2)/dx$ has a maximum value. Furthermore, this ray was found to subsequently propagate parallel to the shock front for an infinitely long time. Thus, if the detonation did not have a plane where $d(a^2-u^2)/dx$ was maximum, their theory also predicted that the detonation would be dynamically stable to a transverse wave perturbation. They also noted that since the ray propagating at the plane $d(a^2-u^2)/dx = \text{maximum}$ is propagating in a region where a temperature and

density sensitive exothermic chemical reaction is occurring, its rate of amplitude growth is dependent on the details of the chemical reaction. Briefly, they came to the conclusion that any exothermic reaction that has been observed in nature would produce transverse wave amplification under these conditions.

In the region where the quantity $d(a^2-u^2)/dx$ is increasing they found another interesting behavior. In this region, wave fronts which have a particular range of initial orientations convolute with time in such a manner that an element of the wave front propagates some distance away from the shock front, then turns around and returns to the front to reflect and repeat the process. Since this behavior is occurring in a region which contains a temperature-sensitive exothermic reaction, it is also expected that the amplitude of this wave front will grow with time. Interestingly enough they found that this wave front eventually produces multiple shock contacts which asymptotically approach a regular spacing and that this spacing is dependent on the extent and the detailed structure of the reaction zone of the detonation.

Relative to the problem of longitudinal instability in detonation waves, Erpenbeck²¹ has performed an analysis for transverse waves with arbitrarily long wave lengths (which effectively become a longitudinal disturbance in the flow). He once again found values of overdrive above which instability occurs. In a recent paper, Fickett and Wood²⁵ have performed a one-dimensional non-steady method of characteristic analysis on a propagating overdriven detonation produced by a piston motion. They carried their analysis out to very long times and observed a continued large oscillation in the shock velocity for the case that Erpenbeck predicted would be unstable. They also observed no sustained oscillations for one of Erpenbeck's stable cases. In a following paper, Erpenbeck²⁶ has been able to predict the magnitude of these large scale oscillations by extending his analysis to include nonlinear terms.

From the above we see that one should expect hydrodynamic instabilities in many of the flow situations in which a shock wave is closely followed by exothermic chemical reaction. Specifically, these results show that the CJ plane is not important to the occurrence of flow instabilities and therefore that the self sustenance of detonation is not a requirement for instability. In other words, the instabilities observed on self-sustaining detonations are quite certainly a restricted example of a general class of hydrodynamic instabilities which occur in flows containing a heat release zone following a shock transition.

II. Structure

A number of investigators have observed that essentially all detonations propagate with a complex non-steady frontal structure. Early work in this area was performed primarily by Russian investigators and has been summarized in a number of recent Russian texts.^{27,28,29,30} The primary findings of these and subsequent investigators is that the main shock front consists of many sections which are locally convex towards the upstream flow and the intersection of these sections are traveling across the front as waves (see for example the early photographs obtained by White³¹). Both the occurrence and structure of these transverse waves are intimately connected to the chemistry of the reactions occurring in the detonation. In the following sections our current understanding of the nature of this transverse structure and its interaction with the chemistry will be discussed.

a. The Nature of Transverse Waves

The transverse waves that propagate across the front of a detonation have been found to occur at the front as Mach stems or triple shock intersections of finite amplitude^{27,32,33}. The single spin detonation that occurs in a round tube is unique because it was the first non-one-dimensional structure to be observed and because it is the only example of a transverse wave structure which is steady in the proper coordinate system. (In this case, the coordinate system which renders the wave motion steady is one which rotates at the spin frequency with its axis of rotation along the tube axis.) Spinning detonation contains a very complex double Mach stem pattern which has been described by Voitsekhovskii, Mitrofanov and Topchian²⁷ and verified by the careful work of Duff³² and Schott³³. In all other observed transverse structures, there are always opposing transverse waves propagating across the front simultaneously and the structure at the front is necessarily two or three-dimensional non-steady.

At the present time, all the details of this complex structure are not understood. However, as mentioned above, it is known that these transverse waves consist of Mach stem interactions at the front. These have been observed experimentally, both on propagating detonations^{34,35,36} and as an artificially produced reactive Mach stem on the "laminar" detonation which White and Cary³⁷ first produced by expanding an overdriven detonation through a cylindrical nozzle. A spark interferogram of such an artificially produced Mach stem and an analysis of its triple point structure is shown in Figure 1. This structure was observed by White and Cary³⁷ and analyzed by Strehlow.³⁸ Strehlow found that even though the shock triggered a highly exothermic reaction a few microseconds after it passes an element of the gas, the triple point structure is still controlled by an unreactive-unreactive slipstream balance. The observations on propagating detonations have not been analyzed as extensively at the present time. There are still serious questions concerning the disposition of the reflected shock and the manner in which it interacts with the reaction zone.

The triple points propagating on the shock front of the detonation have been found to have the property that they will "write" a line on a smoked surface. This behavior was first observed by Antolik³⁹ in 1875 and was used by E. Mach in conjunction with his studies of spark discharges and the interaction of shock waves produced by bullets fired in the laboratory. The technique lay dormant for over 80 years until in 1958, Denisov and Troshin⁴⁰ started to use it to study detonation structure. Oppenheim and Utriew⁴¹ have recently verified that the triple point is writing the pattern by direct laser schlieren photography looking through the smoked film.

This property has been very helpful in the study of transverse wave phenomena in detonations. To perform an experiment, one simply smokes a surface with either wood, acetylene, kerosine, or any other appropriate smoke and exposes it to the detonation. After exposure the film is "fixed" with a clear lacquer spray. The quality is almost as good as that of a photographic negative. A few examples of smoke track records obtained with detonations propagating in a rectangular tube are shown in Figure 2.

In Figure 3, the results of studying a symmetric interaction using the smoke track technique are summarized⁴². In this case, a number of sand grains were placed on the smoke plate before the detonation

passed over the plate and each grain wrote a wake pattern whose axis is normal to the local orientation of the leading shock wave of the detonation. The intersection geometry is therefore completely determined and one may calculate all shock and flow properties in the neighborhood of the intersection. From this calculation one concludes that the normal shock velocity is discontinuously increased by about 30 to 40% at the center line of the intersection and therefore that all portions of the leading shock are attenuating very rapidly in the detonation. This is very disturbing, because even though the shock is locally propagating at a velocity which varies from 10 to 20% below CJ to 10 to 20% above CJ, the detonation as a whole is still observed to propagate at very close to the CJ velocity. The mechanism which makes this possible is not understood at the present time.

b. Acoustic Coupling and Limits

The transverse waves on the detonation front exhibit a degree of regularity which is dependent on the geometry of the detonation tube and on the chemical system which is supporting the detonation. The effect of detonation tube geometry is primarily caused by the fact that the high pressure regions propagating across the front (immediately behind the Mach stem) produce transverse pressure waves in the gas column following the front and that these transverse waves have a tendency to couple with resonant transverse modes of the gas column downstream of the front. Manson⁴³ and Fay⁴⁴ predicted this behavior for near limit detonations approximately 15 to 20 years ago. It now appears, however, that this coupling is incidental to the occurrence of transverse waves on the front. For example, Duff and Finger⁴⁵ have observed transverse structure on a spherical detonation. However, the occurrence of transverse waves on the front is not incidental to the self-sustenance of the detonation wave. It has been noted repeatedly in the literature that as the initial pressure is lowered or as the detonable mixture is diluted with an inert gas, the characteristic spacing of the structure gets larger. It also has been observed that the last transverse frontal structure that occurs in any specific tube corresponds to the lowest transverse acoustic mode of the hot gas column following the front in that tube. In the case of round tubes, this mode is the single spin mode. It is also now quite certain, that detonations propagating just inside these limits propagate at an average velocity lower than the CJ velocity. This is evidently caused because a significant fraction of the chemical energy released in the reaction zone resides in transverse wave energy for an appreciable length of time in these limit detonations.

In certain chemical systems, the tube geometry has a very marked effect on the regularity of the writing pattern. Figure 4 illustrates that extremely regular patterns can be observed in rectangular and planar (very narrow rectangular) geometries. Incidentally, the planar mode occurs when the preferred transverse spacing of the detonation is at least five times the width of a narrow channel. It is the simplest self-sustaining detonation mode which may be studied, since it is two-dimensional non-steady. In other chemical systems however, it has been observed that the regularity of the transverse structure is poor in all tube geometries. Typical smoke track records are shown in Figure 2 and results for a number of fuel-oxidizer mixtures are summarized in Table 1. The reason for this behavior has not been discovered at the present time. It is, however, quite obviously related to the chemistry of the system which is detonating.

c. Spacing of Transverse Waves and Chemistry

In systems which show a reasonably regular writing pattern in rectangular tubes, it has been observed that the characteristic size of the pattern is dictated by the initial pressure level and the dilution⁴². The transverse wave spacing of regular smoke track patterns is usually defined as the average distance between two successive waves propagating in the same direction measured along a line parallel to the average orientation of the leading shock wave, i.e., along the diameter of the tube.

The most extensive data on spacing has been taken in the hydrogen-oxygen system containing argon as an inert diluent. This data is summarized and compared with the acoustic predictions for the spacing and with average induction zone and recombination zone lengths in Figure 5. Here the recombination zone length has been approximated by normalizing the reciprocal of the maximum rate of recombination and using the flow velocity at the start of recombination. We notice from this figure that the spacing is always larger than the sum of the induction plus recombination zone lengths in this system and therefore that each element of the detonatable mixture is never traversed by more than one transverse wave of the same family before it completes its reaction. We also note from Figure 5 that the spacing predicted from the acoustic theory is considerably less than any of the reaction lengths or than the spacing of the transverse wave at the front of the detonation. Thus it appears that the acoustic theory cannot be used to predict the spacing of the finite amplitude transverse waves that are observed on propagating detonations. This is not surprising, however, since the interaction of opposing waves appears to be the regulating mechanism in a propagating detonation⁴². However, it is interesting that the spacings which are measured and the spacings which are calculated using the acoustic theory are roughly proportional to each other.

d. Spontaneous Growth of Transverse Waves

The acoustic theory of Strehlow and Fernandes²³ predicts that coherent transverse waves should grow in amplitude as they propagate through the reaction zone of the detonation and the Barthel and Strehlow²⁴ theory predicts that a single acoustic front should convolute to produce a number of evenly spaced shock contacts after some time. This contention has been verified experimentally for at least one case, during the homogeneous initiation of detonation behind a reflected shock wave in a hydrogen-oxygen-argon mixture. Figure 6 contains an (x, t) initiation photograph and a smoke track record obtained simultaneously on a side wall of the tube. It is interesting to note that during this nicely one-dimensional initiation experiment, very weak transverse waves appear on the front and then grow in amplitude (as indicated by an increase in the refraction of opposing waves at their intersection). In a recent paper, Strehlow, Liaugminas, Watson and Eyman⁴² have shown that the appearance location on the smoke track records may be used to predict a reasonably constant exponential coefficient for the linear theory of acoustic growth. From these experimental results we may deduce that there is little doubt but that the acoustic theory of amplitude growth offers a viable mechanism for the appearance of the finite amplitude transverse waves that occur on propagating detonations.

III. Initiation and Failure

a. Flame to Detonation Initiation

This subject has been studied extensively in tubes of varying geometry and in many chemical systems. By far the most quantitative study of the subject has been performed by Oppenheim, Urtiew and co-workers⁴⁶⁻⁵² at Berkeley. The phenomena is complex and the process of flame acceleration (eventually leading to detonation) is very dependent on the condition of the tube walls and the presence of obstacles in the flow.

In general the process develops in the following manner. A flame, generated at the closed end of tube produces a compression wave in the gas which steepens into a shock wave some distance from the end of the tube. This flow develops a boundary layer which becomes turbulent and causes the flame to accelerate. The acceleration of the flame then subsequently reinforces the original shock wave. Eventually, the shock preheats the gas sufficiently to cause the onset of detonation. However, the actual occurrence of detonation is sometimes obscured by the proximity of a very turbulent flame in the neighborhood of the initiation point and in general the process is not one-dimensional. The acceleration process depends very strongly on the ratio of the burning velocity to the sound velocity in the combustible mixture. High values of this ratio cause more rapid acceleration in the system and therefore cause a shorter detonation "induction" distance. If the tube has a finite length and is closed at both ends, shock reflection from the far end may occur before ignition. The reflection process will always preferentially trigger detonation and it has the added disadvantage that it produces very high pressures before the detonation occurs. This type of initiation is extremely important to the prediction of possible hazardous detonations in large industrial installations. It appears, from the information now available that one may make quite realistic estimates of the possibility of detonation for any specific apparatus and combustible mixture.

b. Homogeneous Initiation

Homogeneous one-dimensional initiation has been observed behind a reflected shock wave in a conventional shock tube^{54,55,56}. Figure 6 shows an x,t schlieren inferogram of such an initiation process. The gas dynamics of this process have been modeled by Gilbert and Strehlow⁵⁶ using an (x,t) method of characteristics analysis. They assumed that each element of the gas reacts with kinetics which are dependent only on the previous time-temperature pressure history and that the gas dynamics may be modeled in a conventional manner. The results of their analysis agreed with experimental observations of high temperature reflected initiation in every detail and showed that this type of initiation process is directly caused by the interaction of reaction kinetics and inviscid gas dynamics. In short, they found that because shock heating is a wave process it generates a reaction wave which interacts with and accelerates the shock wave in a manner which is completely predictable if the kinetics are well understood. Furthermore they found that for this process thermal conduction and diffusion are unimportant to the wave development process.

In a following work, Strehlow, Crooker, and Cusey⁵⁷ have experimentally and theoretically studied the process of detonation initiation when a step shock wave passes into a slowly converging channel. Once again they observed that the first point of initiation, in real time, in the converging channel could be predicted by simple application of non-steady gas dynamics and the proper reaction kinetics.

c. Hot Spot Initiation

The most extensive studies of initiation have been performed in the hydrogen-oxygen system^{56, 58, 59}. At high temperature and low pressure this system exhibits a strictly homogeneous initiation behavior in simple flow geometries. However at low temperature (900°-1100°K) and high pressure (i.e., at high reactant concentrations) the system shows a very unusual type of initiation which is difficult to categorize. At its first appearance, the initiation occurs as a series of "hot spots" and these propagate in a number of spherically growing flames until the coalescence of these flames leads to the initiation of detonation. An example is shown in Figure 7. This behavior appears to occur in the pressure and temperature region which is roughly above and to the left of a line on a (P, T) explosion limit diagram obtained by extrapolating the second limit line into the "explosion" region. It appears that this behavior occurs under conditions where appreciable quantities of the species HO₂ are able to accumulate in the mixture⁵⁹. More quantitative work on this type of initiation is needed.

d. Detonation Failure

In this reviewer's estimation the problem of detonation failure has not adequately been studied at the present time. It is quite apparent that each frontal element of a self-sustaining detonation is failing at a rate which is quite rapid when compared to the rate of heat release in the reaction zone. It is also true that if the detonation is allowed to propagate into an enlargement of the tube, sections of the detonation are observed to "fail" because opposing transverse waves are not reflected from the wall and momentarily fail to intersect⁶⁰. However there has never been a truly quantitative study of this process. To date there has been only one theoretical study of this problem and this study is relatively incomplete. Strehlow and Hartung⁶¹ constructed a "steady" one-dimensional overdriven detonation and then allowed it to interact with a strong rarefaction fan approaching it from the rear. The kinetics were assumed to be zero order with a constant activation energy. Rapid failure of the detonation was found to occur for activation energies of 18 and 50 kcal. For a third case, when the activation energy was assumed to be zero, the results were inconclusive because the detonation did not decay to below the CJ velocity. As mentioned above, more work is needed on failure of detonation waves.

IV. The Use of Detonations in Reaction Kinetic Studies

There have been two major techniques developed to study reaction kinetics in actual detonations. White and Cary³⁷ have produced "laminar" detonations by passing an ordinary detonation through a convergent-divergent nozzle. While this flow is non-steady it is still useful for rates studies. They have applied it to the study of vibrational relaxation in exothermic systems and to the study of the effect of vibrational relaxation processes on induction zone kinetics. This technique has also been used to study induction zone kinetics by Mullaney and co-workers^{62, 63}.

In another technique, a one-dimensional detonation has been stabilized in a wind tunnel and the reaction profile studied by observing the pressure profile downstream of the normal shock⁶⁴. This technique has interesting engineering implications because it measures overall rates of heat release directly in a nicely controlled one-dimensional geometry.

Unfortunately, it appears that all attempts to resolve the reaction zone in ordinary self-sustaining detonations are doomed to failure, because of the presence of transverse waves. Soloukhin in his discussion of what are, in his terminology, "multi-fronted detonations"⁶⁰, has pointed out that either averaging across the transverse structure with optical techniques or using pressure transducers at the wall will always yield results which are incorrect in terms of a one-dimensional theory. Thus the utility of ordinary propagating detonations for kinetic studies is at best marginal.

SHOCK TUBE HYDRODYNAMICS

Since the inception of its use in the early 1950's the chemical shock tube has greatly expanded the temperature range available for quantitative experimental studies of the physico-chemical properties of gases. The success of its application as a research tool may be gauged by the fact that throughout the world the extant chemical shock tube literature consists of over a thousand references^{65,1} and is presently being increased at the rate of approximately 150 references per year.

In the field of reaction kinetics studies, there are two fundamental reasons for this success. In the first place the step shock wave produced during the operation of an ideal shock tube yields an extremely precise and reproducible heating cycle with excellent initial conditions. In the second place the shock tube is particularly suited for studies in systems that are highly diluted with an inert gas and studies in such dilute systems have consistently yielded the best quantitative kinetic data. Therefore, one might say that gas dynamics is the servant which allows one to make relatively precise rate measurements at high temperatures in a chemical shock tube. However, it is also the master of the situation in that, (1) the geometry of the chemical shock tube is dictated primarily by gas dynamics considerations, (2) specific gas dynamics non-idealities limit the utility of the apparatus and (3) the coupling of the chemical reaction and the gas dynamics is always important to the study of reaction kinetics; particularly in exothermic systems.

At the present time sufficient information has been compiled concerning the behavior of shock tubes, so that the design of a shock tube for precise chemical rate studies is possible. This portion of the paper will present a critical discussion of the relative merit of various experimental techniques and in addition will review recent work on non-ideal shock tube behavior and recently acquired knowledge concerning the occurrence of those flow idealities caused by the presence of exothermic reactions.

I. Studies Behind "Steady" Incident Shock Waves

The most precise measurement of a chemical reaction rate constant may be obtained by observing the reaction immediately behind the incident shock wave produced in a conventional shock tube. For the

highest precision it is imperative that these measurements be made at high dilution in a monatomic gas and that both the rate measurement and the shock velocity measurement be extrapolated to the instant the shock uncovered the observation station. In all other cases flow non-idealities will cause difficulties in the interpretation of the data. This type of extrapolation is possible in only certain specific cases, for example, in the study of the initial dissociation rate of a diatomic or more complex molecule. Typical examples of investigations which have yielded results of high precision using this technique are the measurements of the dissociation rate of triatomic molecules by Olszewski, Troe and Wagner^{66,67,68,69}, the measurement of hydrogen dissociation by Myerson, Thompson and Joseph⁷⁰ and the measurement of oxygen atom recombination by Kiefer and Lutz⁷¹.

In cases where the reaction process must be followed for a long time after shock passage the measurement of a correct rate is not really very straightforward. The raw data after being reduced with the help of ideal shock tube theory must still be corrected for the non-ideal flow behavior in the particular tube that is being used in the experiment. Since combustion drivers, heated gas drivers, double diaphragm tubes, detonation drivers, etc., produce notoriously non-ideal flow situations in the test gas, we will discuss correction procedures which must be used for only the simplest driving process: a cold gas driver operating with a simple pressure burst in a conventional constant area shock tube. Even in this case there are three overlapping but relatively independent corrections which must concern the investigator if he is to obtain reasonable rate constants. The first of this concerns the diaphragm material used in the study. Diaphragm opening time is a function of both the inertia and the tearing properties of the diaphragm material^{72,73}. Probably the best material in this respect is cellulose acetate because it disintegrates so thoroughly on bursting. Mylar is quite slow and irregular and has been shown to produce irreproducible late pressure pulses⁷⁴ (probably due to flapping or late tearing) which could lead to scatter in the rate measurements. Metal diaphragms in general appear to open rather slowly but reasonably reproducible, if scored uniformly.

It has been observed that even when reproducible, the rate of the diaphragm opening process is extremely important in determining the early behavior of the shock tube flow. White⁷² and Alpher and White⁷³ have shown that for a slow diaphragm opening process, the shock velocity increases for a considerable distance and only then exhibits the decay that one would expect from boundary layer growth. It has been shown theoretically that this slow acceleration may easily yield velocities which are slightly above the theoretical velocity for that bursting pressure ratio. Since this is the formative period for the shock wave, it is best to make kinetic measurements outside of this region. This region extends for a distance from the diaphragm clamp which is a function of the diaphragm material, the bursting pressure ratio and the tube diameter. White⁷² reports an accelerating shock 40' from the clamp with a metal diaphragm in a 3 1/4" square tube at $M = 15$ in argon. However at a shock Mach number of 8 he found that the maximum velocity occurred only 12 feet from the diaphragm. If one remains clear of this accelerating region the major problem which may arise from the diaphragm bursting process is the problem of late spurious pressure signals from materials such as mylar.

Other flow non-idealities that occur in a shock tube during the late flow are all connected to the development of the boundary layer behind the incident shock. These effects have been reviewed by Spence and Woods⁷⁵ and by Holbeche and Spence⁷⁶. In addition, Mirels^{77,78,79,80,81}, in a number of papers has discussed the seriousness of this problem. If one assumes an ideal diaphragm burst it has been found that the flow essentially divides itself into two regimes. The first of these is a period when the shock is decaying due to the boundary layer growth. During this period, the length of the hot column of test gas is growing at a reasonably constant rate that is somewhat below the theoretical rate predicted by ideal theory. After this initial period the boundary layer has grown to such an extent that it is accumulating test gas from the main sample at the same rate as the incident shock is accumulating new test gas. In this situation, the rate of decay in the incident shock velocity becomes less than before and the length of the test gas column becomes relatively constant. This effect was first reported by Duff⁸². Recently Fox, McLaren and Hobson⁸³ have shown that Mirel's theory quite adequately predicts the observed behavior.

There are two distinctly different types of corrections necessary because of these flow non-idealities. In this paper the effects of non-ideal flow will be discussed for only the state property temperature because reaction rates are primarily temperature sensitive. In the first place the shock wave is continually decelerating -- at first rapidly and later slowly -- and this introduces a temperature gradient in the gas sample which can be large. This temperature gradient may be calculated if one assumes that the shock attenuates at a specific rate and that there are two possible limit behaviors for the pressure decay behind the front. These are: 1. Each fluid element retains its shock transition pressure. In this case the gas contains a relatively step pressure gradient due to attenuation and 2. Each element of gas after shock compression, is isentropically expanded to the instantaneous shock transition pressure. The real behavior behind the decaying shock lies somewhere between these two limit behaviors. These two assumptions yield the following equations for the temperature gradient in the gas following the shock wave.

- With a residual pressure gradient (no pressure change after shock transition)

$$\frac{dT_2}{T_1} = \frac{4(\gamma-1)}{(\gamma+1)^2} \left(\frac{\gamma M_1^4 + 1}{M_1^2} \right) \frac{dM_1}{M_1}$$

- With isentropic expansion

$$\frac{dT_2}{T_1} = \frac{4(\gamma-1)}{(\gamma+1)^2} \frac{(M_1^2 - 1)^2}{M_1^2} \frac{dM_1}{M}$$

Figure 8 is a plot of $T_2/\text{millisecond}$ in the flow behind a shock wave in argon with an attenuation rate of 1% per meter. As this figure shows, the gradients are sizable and very dependent on the testing temperature T_2 .

The second correction has been estimated by Mirels⁸¹ on the basis of a boundary layer development theory. In his theory for the late flow he assumes a constant velocity shock wave propagating down the tube and he calculates the changes in flow properties along the tube axis due to

the boundary layer growth. His results are plotted as a function of the dimensionless distance from the shock wave l/l_m where l_m is the equilibrium length of the hot gas column. In general he finds that all state properties increase in value as one travels away from the shock. Specifically for a $\gamma = 5/3$ gas at $M = 3$ he finds that the temperature rises to about $1.07 T_2$ at a distance $l = 0.2 l_m$ and then remains relatively constant for the remainder of the transit time. However he finds that an even more serious correction than this is the correction to the testing time that must be included for strong shocks. Specifically, the real testing time for a strong shock increases rapidly over the ideal testing time and at a position $l = 0.2 l_m$ the real testing time is $3/2$ of the ideal testing time. This has been verified quantitatively by Fox, McLaren and Hobson⁸³.

The non-ideal temperature drifts caused by the effects of shock velocity attenuation and boundary layer growth are in the same direction (each yielding a temperature increase as one travels away from the shock) and they may be added to a good first approximation. The testing time correction augments both of these temperature gradients and can in itself cause large errors if ideal flow is assumed. It appears as though some of the extant reaction kinetic data obtained in shock tubes should be re-evaluated in the light of these developments. It should be pointed out that these corrections can change the apparent activation energy of the reaction as well as the absolute rate because in all cases the correction becomes more severe as the run temperature increases.

In the previous discussion it was implied that dilution with an inert monatomic gas is desirable for accurate kinetic rate measurements. Modica and La Graff⁸⁴ have recently studied the decomposition of N_2F_4 using both argon and nitrogen as a diluent and their comparative results reinforce the contention that monatomic gas dilution is necessary to reduce the ambiguity of results. In another interesting experiment along these same lines Seery and Britton⁸⁵ have shown that xenon is apparently not a completely inert diluent during the dissociation of fluroine.

II. Reflected-Shock Time-Resolved Techniques

Rate measurements made behind the reflected shock offer certain advantages over measurements made using the incident technique. In particular, sampling with a time-of-flight or quadrapole mass spectrometer is possible and laboratory time is real time because the gas is relatively quiescent in the neighborhood of the end-wall. Also, for the same initial conditions the reflected technique produces considerably high temperatures than an incident technique.

In this section we will discuss some difficulties exhibited by only the simplest time-resolved reflected techniques, those in which a sample is drawn from the end wall for mass spectrometer analysis or in which an emission or absorption spectroscopic technique is used to observe a gas sample trapped near the back wall. In these cases the initial temperature behind a reflected shock may be calculated quite accurately from the incident shock velocity measured at the moment of impact, assuming that the gas is entirely inert. The shock velocity at impact must be determined by extrapolating upstream measurements and the calculation is only valid for the case where the reactant is highly diluted with a monatomic carrier gas. As in the case of incident measurements, if it is not possible to extrapolate

the rate to the shock, corrections for the flow non-idealities must be included to accurately determine the rates.

Boundary layer corrections are still important in this case. If one operates near the back wall (within approximately 5mm of the back wall) the testing time correction ceases to be important and if one operates with a monatomic carrier gas, bifurcation problems are not serious⁸⁶. However the pressure rise in the gas traveling behind the incident shock does cause a serious perturbation to the pressure-time and temperature-time history of the gas sample trapped near the back wall. As the reflected shock propagates away from the back wall, it encounters this pressure increase and transmits signals to the gas at the back wall. Fortunately the sample near the back wall is compressed isentropically during this process and for a monatomic gas the temperature increase may be estimated rather accurately using the equation:

$$\frac{\Delta T}{T_2} = \frac{\gamma - 1}{\gamma} \frac{\Delta P}{P_2} \approx 0.4 \frac{\Delta P}{P_2}$$

Thus a pressure gauge may be used to monitor the rate of pressure change at the back wall and this rate of change may be used to estimate the equivalent temperature drift near the back wall. Alternately, Mirels' theory may be used in conjunction with Rudinger's simplified method of characteristics analysis⁸⁷ to calculate a pressure and temperature change at the back wall for the particular shock tube and initial conditions of the experiment.

In the case of mass spectrometer analyses sampling techniques are important and the correction for heat transfer to the wall should be considered in any calculation⁸⁸. These corrections are reasonably straightforward, however, since in this time scale convection does not occur. Therefore, only a conduction equation need be solved to determine the necessary correction. Heat transfer can also be important in spectroscopic techniques because absorption in the cold boundary layer gases at the wall can completely dominate the measurement in certain cases⁸⁹. Allen, Textoris and Wilson⁹⁹ have developed an ingenious apparatus to circumvent this difficulty.

In addition to the above, reflected-shock, time-resolved techniques suffer from two unique difficulties related to the chemistry. In the first place, the occurrence of chemical reaction behind the incident shock cannot always be dismissed in an *a priori* manner but should be examined for each experimental system. This problem is particularly serious for a first order reaction with a relatively low activation energy and could also be serious for a second order reaction with a very low activation. In the second place, the occurrence of a reaction behind the reflected shock leads to a non-steady reflected shock behavior. There are two reasons for this; either the molecular weight or the enthalpy of the gas may change due to the occurrence of the chemical reaction. By far the most important of these two changes under ordinary conditions is the enthalpy change associated with the reaction process. If an endothermic reaction occurs behind the reflected shock, the shock wave will decelerate and this will cause a pressure decrease and an attendant isentropic temperature decrease at the back wall. Johnson and Britton⁹⁰ have discussed the main features of this flow and a detailed method of characteristic calculation of the interaction for the vibrational relaxation of oxygen has been reported by Nafzinger⁹². He found that the shock relaxation process

took approximately 7τ , where τ is the characteristic relaxation time behind the reflected shock at the back wall. He also observed that this non-steady process led to a substantial residual entropy gradient along the tube axis in the neighborhood of the back wall. The effect of exothermic reactions will be discussed in Section IVb.

III. Single Pulse Techniques

Single pulse techniques⁹³ suffer from a number of relatively distinct problems. In this reviewer's opinion, the sum total of these deficiencies are serious enough to make the technique very difficult for quantitative kinetic studies. It is, however, still a useful technique for qualitative and comparative studies for new or complex systems.

Let us first consider those difficulties that are inherent in a single pulse experiment performed with an ideal shock tube. Even in this case, there are five major problems which limit the technique's utility. These are: 1. Analyses are performed after the experiment is completed. Thus only stable products may be identified and, at best, only the systems overall behavior may be studied. Therefore the deduction of responsible individual reactions with a concordant determination of their rates is very difficult. 2. The heating cycle is not the same for the entire gas sample. Therefore, an average processing time must be estimated for the run. 3. Contact surface tailoring is necessary to obtain long test times and thereby reduce the errors caused by the averaging process mentioned in Part 2 above. However off-design tailoring will cause substantial temperature changes during the last (and usually major) part of the testing time and this is serious if the reaction has a high temperature sensitivity. 4. Cooling must necessarily be produced by a rarefaction fan and this introduces yet another uncertainty into the calculated testing time. 5. If the reaction is not thermally neutral, effects of self-heating must be included. These last two problems have been discussed by Palmer, Knox and McHale⁹⁴.

The non-ideality of a real shock tube flow compounds these problems. By far the most serious non-ideality is the growth of the boundary layer behind the incident shock. This leads to three distinct difficulties which cause problems. In the first place the temperature cannot be held constant for the desired testing time because the upward drift is always appreciable on a time scale of 1 to 4 milliseconds. Secondly, tailoring cannot be exact because of the shift in properties caused by the boundary layer growth. Thirdly, the growing boundary layer can entrain a sizable quantity of the test gas. Therefore the entire reactive sample is not treated by the test pulse. This last difficulty is particularly serious if the entire contents of the tube are being analyzed after the run and may be alleviated to some extent by analyzing only that portion of the test gas that was in the neighborhood of the back wall during the pulse cycle.

The majority of the difficulties mentioned above cause errors in both the activation energy and the actual magnitude of the rate measured with a single pulse tube. This is because the severity of most of these corrections increase as the temperature of the test run increases.

There have been many variations of the single pulse technique introduced in the past few years. Tsang,^{95,96,97,98,99,100} for example, has applied the technique to the study of hydrocarbon decomposition and rearrangement reactions using a mixture that contains a small quantity of "propyl chloride" as a trace reactant. This species is used to independently

obtain information on the temperature-time pulse so as to increase the accuracy of the experimental decomposition measurement. This type of "trace reaction" technique has utility only if it can be proved in an unambiguous manner that the trace reaction is occurring independently of the unknown reaction. It should have the highest accuracy when the activation energies of the two reactions are approximately equal.

Tschuchikow-Roux^{101,102,103} has proposed a ball valve technique which may be used to isolate a sample either between the ball valve and the end plate of the tube or just in the bore of the ball valve. However, his heating cycle analysis is based on ideal shock tube flow theory and since he proposes to calculate heating time by subtracting two large times which are themselves calculated by using ideal theory, the accuracy of his calculation will be low. However, there is the possibility of improving the accuracy by including the necessary boundary layer growth corrections.

Lifshitz, Bauer and Resler¹⁰⁴ have introduced and used a tube with a side dump tank on the test section leg. In conjunction with this they operate the tube without a tailored interface. Instead they "tune" the length of the driver section by inserting plugs at the end wall so that the strong rarefaction fan in the driver gas, after reflecting from the end of the driver section, will overtake the contact surface at the instant that it is met by the reflected shock wave. Thus, testing time in the sample theoretically ranges from zero to approximately 2 t_{ave} . The technique suffers, however, from the fact that non-ideal boundary layer growth will always cause the contact surface to intersect the reflected shock earlier than the theoretical time. If this effect is not included in the "tuning" operation the test gas sample will suffer a complex time-temperature heating cycle and the interpretation of the data will be difficult. The presence of a side dump tank modifies the downstream flow in the test section but a reasonable constant shock velocity is still attained near the end plate. The side dump tank is used primarily to prevent multiple high pressure reflections in the gas sample. It is probably adequate for this purpose.

IV. Exothermic System Instabilities

In addition to all the difficulties mentioned above, in exothermic systems the application of the shock tube to reaction kinetic studies is limited by the occurrence of non-steady flow processes which may be triggered by the presence of the exothermic reaction. Specifically, two distinct types of flow instabilities may occur and it has been found that two independent criteria may be used to predict the occurrence of these instabilities on the basis of known flow properties of the particular system which is under investigation. The two criteria will be called the Chapman-Jouguet criterion and the tube diameter criterion.

a. The Chapman-Jouguet Criterion

Consider a gas mixture capable of sustaining a reaction which drives the system toward a state of full chemical equilibrium. With this restriction we define the mixture to be "exothermic" if, in the (P, V) plane, the Rayleigh line through the point (P_1, V_1) which just tangents the equilibrium Hugoniot represents a shock transition whose Mach number, M_{CJ} , is greater than unity. We call the value of this minimum Mach number for steady shock propagation the Chapman-Jouguet criterion for instability. It

may, of course, be calculated quite accurately if thermodynamic properties are available for the system in question.

An example of the application of this criterion may be discussed with the help of Figure 9. The system considered in this figure is a stoichiometric hydrogen-oxygen mixture diluted with various amounts of argon at the initial conditions $T_1 = 300^{\circ}\text{K}$ and $P_1 = 100$ torr. The unreactive shock temperature for a shock of Mach number $M = M_{\text{CJ}}$ is plotted versus the mixture composition in this figure. The vertical lines approximate regions in which studies have been reported, admittedly for different stoichiometries and pressure levels (the numbers on the lines are reference numbers). The approximation is reasonably good because, however, pressure shifts the curve in Figure 9 only slightly and stoichiometry changes are effectively dilution changes on the basis of a CJ calculation. Region I, above and to the left of the stability line $M = M_{\text{CJ}}$, corresponds to the experimental conditions under which steady incident shock waves may be produced and studied in this system, while region II, below and to the right of the stability curve represents initial shock conditions for which a steady shock wave with chemical reaction cannot exist. Note that Belles and Lauver¹⁰⁷ worked with incident shocks just at the edge of region I and that all other extant incident shock studies were performed well inside region I. It is interesting that Belles and Lauver report instability difficulties at low temperatures.

In region II of Figure 1, kinetic studies must necessarily be performed in conjunction with a flow which is longitudinally non-steady. For this reason well controlled and reproducible initial conditions are important to the design of these experiments. For example, the simple fact that the initial flow produced by the diaphragm bursting process is neither one-dimensional nor reproducible obviates the use of a conventional incident shock technique for region II studies. There are, however, four shock tube techniques which either have been used or have a potential use for performing kinetic studies under region II conditions. These will be discussed individually.

The first of these techniques uses the reflected shock region. Reflected shock studies, when performed in a gas mixture with a high heat capacity ratio, yield a reasonably one-dimensional heating cycle with well defined initial conditions⁵⁴. Even though the flow following reflection remains non-steady for an extended period of time (and indeed may yield a detonation wave propagating away from the end wall) the gas near the end wall may be used for kinetic studies since it is quiescent, it suffers no additional shock transitions after reflected shock passage and its pressure-time history during the reaction process may be predicted reasonably well⁵⁵. This technique has been used successfully by numerous investigators^{54,105,106} in situations where the steady incident technique would not be applicable in terms of the CJ criterion.

The second technique consists of passing a weak, but well established, step shock wave produced in a conventional constant area shock tube into and through a section of tubing whose cross sectional area is slowly decreasing with distance. If the rate of decrease of area is sufficiently slow (a maximum wall angle to the center line of approximately 10° for example) the flow will become a quasi one-dimensional non-steady flow in which the leading shock wave is

continually accelerating. In this situation a non-steady flow calculation is required to sort the reaction kinetics from the gas dynamics effects. This experiment has been performed and such a calculation has been made for a tube in which $d(\ln A)/dx = -k$ during convergence⁵⁷. Figure 10 contains iso-temperature and iso-pressure plots in an (x, t) coordinate system for a typical theoretical run assuming that the flow is entirely inert. The region which is mapped is bounded by the leading shock wave, the reflected shock wave and the "explosion line". The locus of the "explosion line" in this figure was determined by assuming that the fraction of the delay time to explosion which has been used at each instant along each separate particle path may be calculated using the relation.

$$d\phi = \frac{dt}{\tau(T, P)}$$

where

$$\tau = \frac{A}{[O_2]} \exp(-E/RT)$$

With this model the points at which

$$\int_0^t \frac{dt}{\tau(T, P)} = 1$$

on each particle path describe the locus of the explosion line. This approach assumes that the reaction is thermally neutral during the induction period and therefore that the reaction does not interact with the flow. Furthermore, as presently constructed, the model only allows one to predict the location of the minimum time in the explosion line locus on an (x, t) plot since the effect of heat release during the explosion is not included in the model. Reference 57 contains a more complete discussion of this technique.

To this author's knowledge the third possible non-steady technique has not yet been applied to the study of reaction kinetics in exothermic systems. It is an incident technique which involves the use of a ball valve whose internal bore is exactly equal to the diameter of the shock tube. This type of ball valve shock tube has been used in the study of endothermic systems¹⁰¹ and to prevent premature explosions in reflected shock studies⁵⁴. If the initial pressure were adjusted externally before opening the valve and if the shock tube were operated vertically, density separation of an inert gas near the diaphragm and the explosion mixture downstream could be maintained until a well formed incident shock wave passes the location of the ball valve. Observation at a number of window stations downstream of the valve would then allow reaction kinetic studies to be studied even though the flow was non-steady. This technique has two advantages relative to the converging channel technique in that both the construction of the apparatus and the calculation of the flow would be simpler. A simpler calculation of the flow may be made because one may assume the flow in the reactive mixture to be pseudo steady until an

appreciable amount of chemical reaction has occurred. This technique also has an advantage over the reflected shock technique for those cases where the system to be studied cannot be diluted to repress bifurcation. This is because, in the ball valve technique only simple boundary layer growth can occur to perturb the incident flow.

The fourth possible technique involves direct expansion of an incident shock wave through a suitably instrumented two-dimensional nozzle. This technique has been described by White and Cary³⁷ and has been discussed in Section IVa.

b. The Tube Diameter Criterion

Even in circumstances where incident shock wave studies are allowable on the basis of the CJ criterion, transverse instabilities may appear during an experiment. This type of behavior has been reported by Schott¹¹⁰, Schott and Bird¹¹¹ and Hawthorne¹¹² and is important to the kineticist because it can produce state variable fluctuations across the tube which may be misinterpreted if the usual one-dimensional assumptions are used in reducing the data.

Schott¹¹⁰ has shown that in acetylene-oxygen mixtures diluted with 97.5 percent argon, this type of instability behavior produces soot track writings which are equivalent to those obtained when a self-sustaining detonation propagates over a smoked foil. This experimental evidence implies that the transverse instabilities observed in reaction kinetic studies in the shock tube are of the same type as those observed⁴² on propagating detonations. This is really not very surprising since the theoretical results discussed in Section a show that detonation per se is not necessary for the appearance of transverse instabilities.

It is also known that all self-sustaining detonations exhibit a transverse wave spacing which is roughly proportional to the "thickness" of the detonations reaction zone. Thus, theoretical predictions and experimental observations both lead to the conclusion that the transverse structure associated with an exothermic reaction zone triggered by shock passage should exhibit a characteristic spacing which is roughly proportional to the reaction zone thickness.

One more experimental observation must be mentioned before we may present a criterion for the occurrence of transverse wave instability in a reactive shock wave propagating in a specific channel. Both Manson⁴³ and Fay⁴⁴ have shown that transverse structure fails to couple with transverse acoustic modes of the tube when the preferred transverse spacing at the front becomes somewhat larger than the major transverse dimension of the tube. Thus it appears that this type of resonance must be possible before transverse instabilities will appear.

Unfortunately, at the present time, the relation between the reaction zone thickness and the characteristic cell size of the structure is not known except for a few systems and then only in an empirical manner. Therefore, at the present time, the "tube diameter" criterion for the occurrence of transverse instabilities in a steady incident shock wave experiment must be stated as follows: Transverse instability may occur in a steady shock wave followed by exothermic reaction only if the bulk of the exothermic process occurs as close to the shock wave as the major transverse dimension of the tube.

This criterion correctly predicts the occurrence of the currently observed transverse instabilities during incident shock reaction kinetic studies in a shock tube. More quantitative information on this subject is available in Schott's report in this symposium volume.¹¹³

SUMMARY

Detonations in premixed gases contain a very complex internal wave structure which is related to the exothermic chemical reactions occurring behind the leading waves. This structure negates many of the classical one-dimensional arguments concerning detonation and the full reasons for its existence and form are not understood at the present time. The mechanism of self-sustenance of a propagating detonation is also open to question at the present time. The situation is not hopeless, however, because it appears that the application of the principles of non-steady reactive gas dynamics (in which all shocks are considered to be infinitely thin non-reactive discontinuities in the flow) will eventually allow a complete description of the structure from first principles.

The discussion of shock tube hydrodynamics indicates that while the shock tube is an extremely useful apparatus for high temperature gas phase reaction kinetic studies, it has its limitations. These are primarily caused by gas dynamic non-idealities due to boundary layer growth and to the interaction of the chemistry and the flow. The implication of this section is that a good kinetic rate measurement may be obtained from a shock tube experiment only if the full contribution of these non-idealities are considered.

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Table I Structure Regularities of Transverse Waves in Detonations

MIXTURE	DILUENT	% DILUTION	REGULARITY
H ₂ — O ₂	1	—	POOR
	1	N ₂	POOR
	1	CO ₂	POOR
	1	He	EXCELLENT
	1	Ar	EXCELLENT
	1	Ar & N ₂	EXCELLENT
	1.5	—	POOR
	.5	Ar	GOOD
	1.5	Ar	GOOD
	.75	2.06	GOOD
C ₂ H ₂ — O ₂	—	—	POOR
	1	Ar	EXCELLENT
C ₂ H ₄ — O ₂	1	—	POOR
	1	Ar	GOOD
	1	Ar	EXCELLENT
CH ₄ — O ₂	1	Ar	IRREGULAR
	1	2.0	IRREGULAR
C ₂ H ₆ — O ₂	1	Ar	POOR
C ₃ H ₈ — O ₂	1.25	—	POOR
	1.25	Ar	POOR
CO — O ₂	1	Ar	GOOD
NH ₃ — O ₂	1	—	IRREGULAR
	1	Ar	IRREGULAR

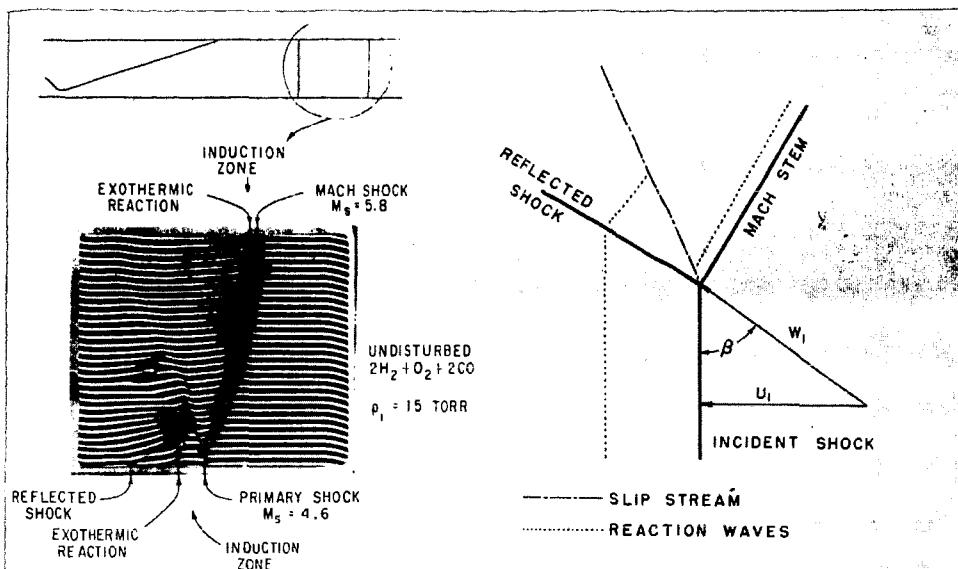
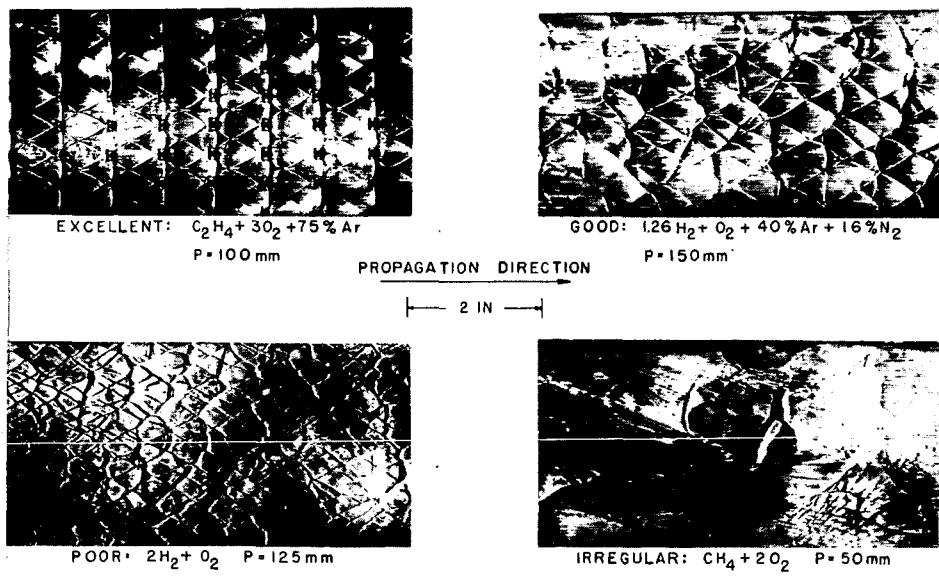


FIGURE 1. ARTIFICIALLY PRODUCED REACTIVE MACH STEM. (COURTESY REF. 37)

FIGURE 2. A FEW EXAMPLES OF SMOKE TRACK RECORDS. $3\frac{1}{4} \times 1\frac{1}{2}$ DETONATION TUBE.

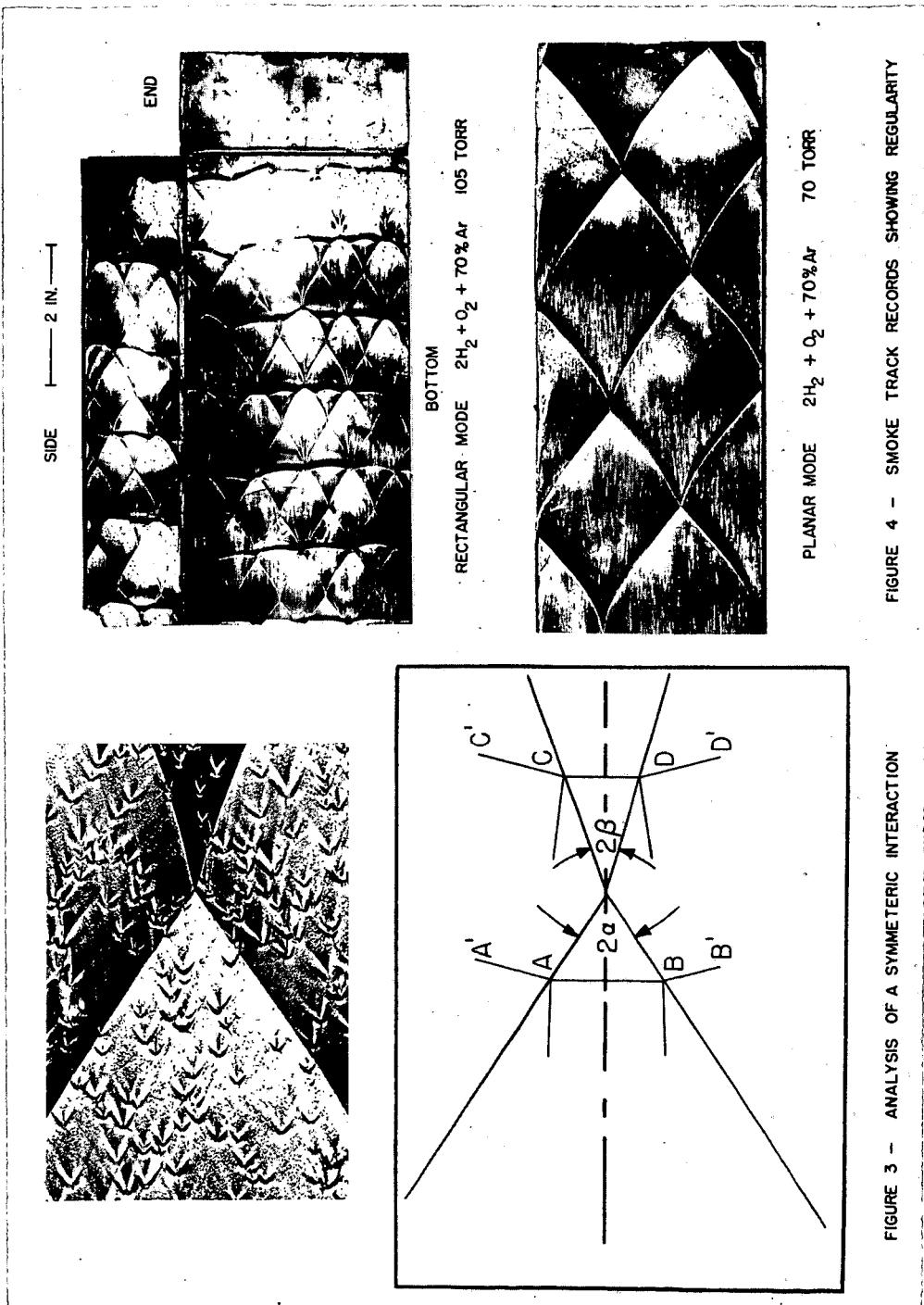


FIGURE 4 - SMOKE TRACK RECORDS SHOWING REGULARITY

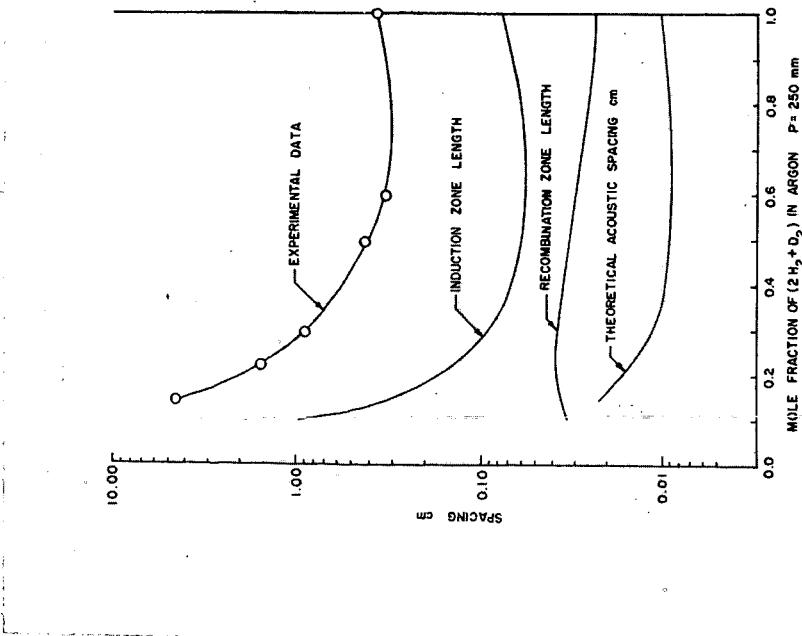
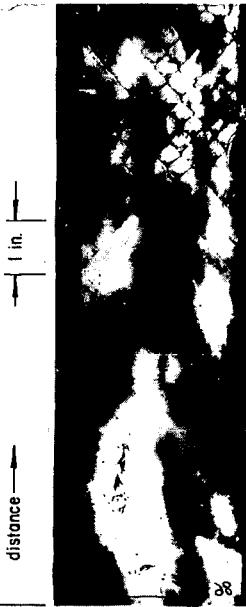
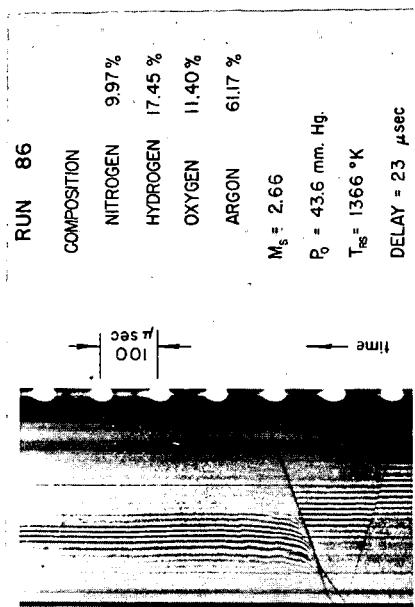


FIGURE 5 - TRANSVERSE WAVE SPACINGS IN THE $2\text{H}_2 + \text{O}_2$ SYSTEM

FIGURE 6 - THE APPEARANCE OF TRANSVERSE WAVES DURING INITIATION BEHIND A REFLECTED SHOCK

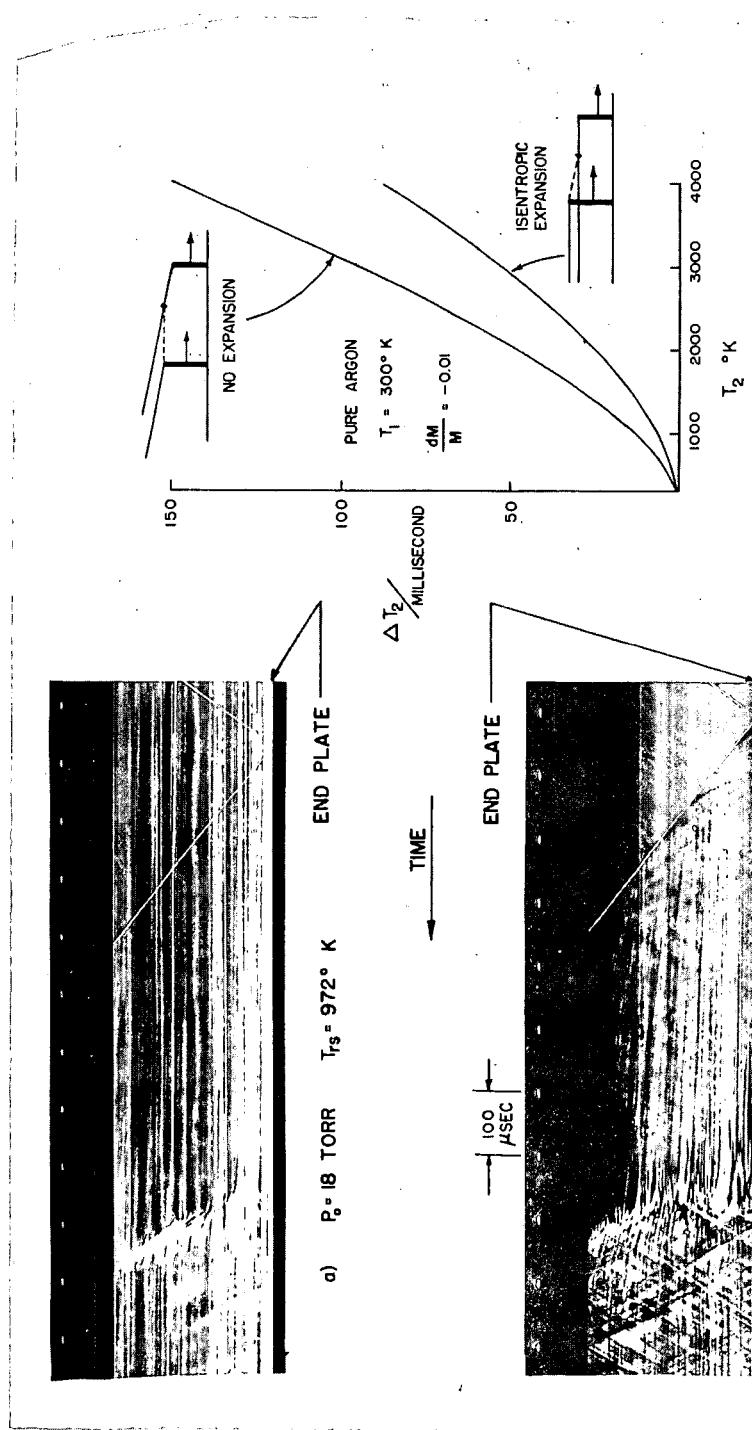


FIGURE 7 - HOT SPOT INITIATION IN $0.08\text{H}_2 + 0.02\text{O}_2 + 0.90\text{Ar}$ MIXTURES
BEHIND REFLECTED SHOCK
COURTESY A. COHEN, BALLISTIC RESEARCH LABORATORIES.

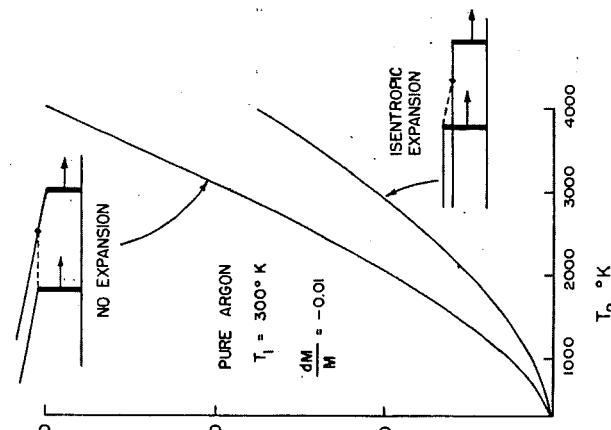


FIGURE 8 - TEMPERATURE CORRECTION FOR
SHOCK ATTENUATION

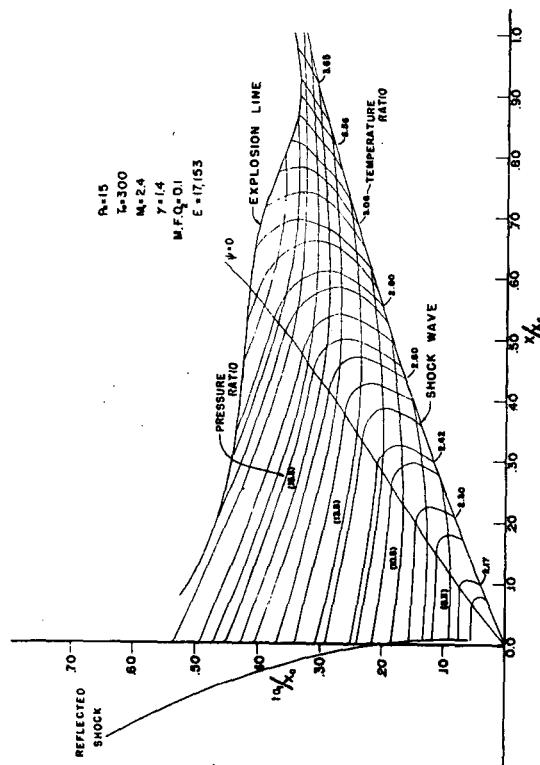


FIGURE 10 - NONSTEADY FLOW BEHIND A STEP SHOCK PROPAGATING INTO A CONVERGING CHANNEL. REF. 57

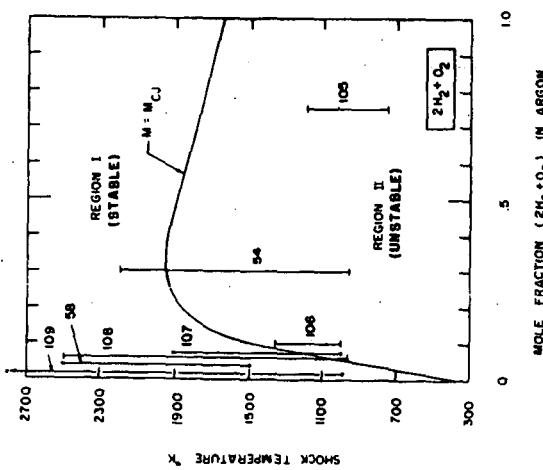


FIGURE 9 - STABILITY REGIONS DEFINED BY THE CJ CRITERION