EXPERIMENTS IN GASDYNAMICS OF EXPLOSIONS

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Introduction

Gasdynamics of explosions is, in our opinion, best denned as the science dealing with the interrelationship between energy transfer occurring at a high rate in a compressible medium and the concomitant motion set up in this medium. This subject has attracted a considerable amount of attention over the last decade for a number of reasons. First, there was a growing realization that this interrelationship is at the source of the explosion process and, in particular, that it governs the structure of detonation waves. At the same time it became evident that this topic also plays the key role in the whole field of hypersonics. This interest was then enhanced by the bearing our subject has on the mutual influence between exothermic chemical reactions and the dynamic behavior of combustion systems, by the work associated with the development of the gasdynamic laser, and by the exciting prospects the exploitation of laser technology offers in the eventual attainment of a controlled thermonuclear reaction.

As a consequence of this, there have been a number of books published on various aspects of the subject, especially in the Soviet Union (e.g., Shchelkin & Troshin 1963, Voitsekhovsky et al 1963, Soloukhin 1963, Zel'dovich & Raizer 1963–1966), and a number of international colloquia, devoted especially to its major topics, organized under the auspices of the International Academy of Astronautics (Oppenheim 1969, 1970, 1972a). Over the last year there were published two review papers (Korobeinikov 1971, Brode, Glass & Oppenheim 1971) and a monograph (Oppenheim 1972b) concerned exclusively with the fundamental features of the subject. These reviews dealt with the theoretical and the technological aspects, respectively. The present article is intended to supplement them with information on laboratory experiments.

The subject matter of gasdynamics of explosions evolved from investigations of detonation phenomena, shock-wave research, and blast-wave studies. Our review considers these three topics in turn, the latter two with particular reference, of course, to chemically reacting media. It should be noted that most of the experimental records reproduced here have been obtained in our laboratories. Also, in order to make our survey self-contained, there is a certain amount of overlap with our previous review papers (Lee, Soloukhin & Oppenheim 1969, Brode, Glass & Oppenheim 1971). The concomitant duplication has been, however, kept to the minimum and, in any case, the material is presented here from a different point of view and with a different purpose than before.

Detonation Phenomena

Almost one-hundred years ago an intensive program of study on combustion was carried out by the distinguished team of Mallard and Le Chatelier at the School of Mines in Paris. At the same time a remarkable development occurred at the School of Pharmacy in the course of research conducted by another team of great prominence, namely that of Berthelot and Vieille. Mallard was at that time a well-known professor of chemistry, the then young Le Chatelier later acquired fame as the pioneer of physical chemistry, Berthelot became recognized as the founder of organic chemistry, and it was as a consequence of these activities that Vieille developed the shock-tube technique for which he is today so well known.

As a consequence of their work, Mallard and Le Chatelier published a nearly 300-page-long classical treatise on combustion (Mallard & Le Chatelier 1883). In it they included a chapter on detonations where, with remarkable gallantry, they acknowledged Berthelot and Vieille as the "discoverers" of these

phenomena. The latter had published their findings a year earlier (Berthelot & Vieille 1882), thereby undoubtedly stealing the show since detonations obviously must have been of much greater importance to mining than to pharmaceutics. Exactly why the scientific team at the School of Pharmacy became involved in this study is not known, but the most plausible reason, one may speculate, is that their attention was directed toward these problems in connection with the vague concepts then prevailing that explosions are associated with some uncontrolled fission of organic molecules. Mallard and Le Chatelier must have had first-hand knowledge of such explosions, especially in connection with some of the experiments they performed where, in order to observe the behavior of flames en naturelle, they filled glass cylinders with combustible mixtures and ignited them at the closed end. From time to time such experiments must have led to some spectacular events associated with total annihilation of the apparatus, and it is perhaps to explain the reasons for such events that Berthelot and Vieille were brought into the act.

Their paper (Berthelot & Vieille 1882) was indeed most remarkable, in that it contained all the ingredients of a classical scientific contribution, namely, (1) an hypothesis about the real nature of a so-far mysterious phenomenon, (2) an experimental verification of the hypothesis, and (3) a prognostic theory permitting the determination of the salient feature of the phenomenon. The hypothesis was that detonation is a wave process characterized by a specific value of its propagation velocity. The experiments were performed in long lead tubes in which the detonation velocity was measured, with remarkable accuracy, to four significant figures. The sensing element and recording device they used is depicted in Fig. 1, reproduced from their paper. The wire inside the tube was covered with fulminate of mercury; the wavefront, upon reaching it, caused a little explosion that broke the electric circuit of the electromagnet in the "chronometer." This caused a rod to fall down. A second wire, placed a known distance downstream from the first, in a similar way caused the chronometer to let another rod fall. The latter, upon hitting the base, released a spring-loaded knife that made a nick on the first rod, marking the end of the time interval for the passage of the wave-front between the two stations. The theory was based on the premise that the heat released by combustion was transformed in its entirety into the kinetic energy of the medium. On the one hand, they neglected in their calculations to take into account the increase in the internal energy associated with the detonation process, and, on the other, they erroneously identified the wavespeed with the mass velocity of the medium. The two errors had a mutually compensating effect, bringing the results of the theory into satisfactory agreement with the experiment.

At the same time Mallard and Le Chatelier obtained the first streak photographs of the development of detonation, which, significantly enough, they published as the first three figures in their monograph (Mallard & Le Chatelier 1883) to demonstrate the growth of instabilities associated with accelerating flames.

Since the "discovery" there has been a period of eighty years of detonation research during which the wave was considered to be primarily a steady-flow phenomenon, while its physical properties became a subject of a thorough study. In the first twenty years, the theory of the plane, steady detonation wave became well established as a consequence primarily of the theoretical contributions of Hugoniot (1887–1889) and Chapman (1899), supported by the experimental investigations of Dixon (1893) and his many associates (Dixon et al 1903). In the next twenty years, the understanding of detonation phenomena was significantly advanced owing to the classical work of Crussard (1907) and Jouguet (1917), who formulated most of the fundamental properties of the one-dimensional steady wave. This was followed by a twenty-year period of refinements in experimental techniques associated mainly with the development of the rotating-mirror camera. Particularly noteworthy in this respect are the contributions of Campbell & Woodhead (1926), Payman (1928), Bone & Townsend (1927a,b), Bone & Frazer (1930), and Bone, Frazer & Wheeler (1935), as well as the investigations of the properties of detonations associated with the chemical nature of the medium carried out by Laffitte (1924), Egerton & Gates (1927), Sokolik & Shtsholkin (Shchelkin) (1933, 1934, 1937) and culminated by Shchelkin (Shtsholkin) (1939) and the theoretical studies of Becker (1922) and Lewis & Friauf (1930).





Collier a gorge: section normale a l'axe du tube.

Chronographe Le Boulenge: chronomietre

Fig. 1. Sensing element and recording device of Berthelot and Vieille

In comparison with flame studies that were conducted at the same time, the detonation research was simpler, and the progress made in this field more spectacular. The main reason for the relative simplicity was the fact that, according to the classical concept, the velocity of propagation of the onedimensional steady detonation wave was considered to be controlled by the gasdynamic choking, the Chapman–Jouguet condition, so that, unlike the flame, it could be regarded as virtually independent of the rate of chemical reactions. In accordance with these views, the detonation velocity was thought to be indicative only of the thermodynamic properties of the medium, and it is significant to note in this connection that its values for various substances were listed in the International Critical Tables (Bone & Townsend 1927b) as physical constants. The net effect of this was the belief that, except for the nonsteady phenomena associated with the development of the wave (Bone, Frazer & Winter 1927, Zel'dovich 1947, Ubbe–lohde 1949) and the spinning mode of its propagation observed near the limits (Bone, Frazer & Wheeler 1935, Zel'dovich 1946), the chemical reaction rates had virtually no influence on the detonation process.

Around the nineteen-forties the validity of this concept became a subject of extensive theoretical studies. The most popular model used for this purpose, called NDZ since it was proposed independently by von Neumann (1942), Doring &Buckhardt (1944), and Zel'dovich (1940-1944), was based on the assumption that the wave consists of a shock followed by an inviscid reaction zone terminated at the Chapman–Jouguet state. The flow throughout the system was considered to be steady, so that the locus of thermodynamic states within the wave was represented by a Rayleigh line. This led to many studies that

were concerned, on the one hand, with the coupling between the steady reaction zone and the nonsteady rarefaction wave behind it, putting in doubt the validity of the Chapman–Jouguet hypothesis (Brinkley & Kirkwood 1949, Kirkwood & Wood 1954) and, on the other hand, with the influence of transport properties, discrediting the Rayleigh line as an adequate representation of the thermodynamic process across the wave (Hirschfelder et al 1958–1959, Oppenheim & Rosciszewski 1963). At the same time, investigations of the stability and development of detonation revealed the importance of nonsteady shock-interaction processes ahead of the reaction zone (Schmidt, Steinicke & Neubert 1951, Oppenheim 1952–1953, Oppenheim & Stern 1959), while impressive programs of experimental research, represented by such notable contributions as those of Kistia–kowsky (e.g., Berets, Green & Kistiakowsky 1950, Kistiakowsky, Knight & Malin 1952, Kistiakowsky & Kydd 1956) and Manson (e.g., Manson 1947, 1948, 1958, Manson & Ferrie 1953, Manson et al 1963) brought detonation processes to a relatively high level of comprehension.

The state of the art existing in the nineteen-fifties was shown in the various textbooks on combustion (Jost 1940, Lewis & von Elbe 1951, Williams 1965, Van Tiggelen & De Soete 1968, Sokolik 1960, Khitrin 1957, Strehlow 1966) and on detonation (Taylor 1952, Zel'dovich & Kompaneets 1960), as well as in monographs (Mullins & Penner 1959) and review papers (Gross & Oppenheim 1959, Oppenheim 1961, Soloukhin 1963–1964). At the same time, development of blast-wave theory enhanced the understanding of gasdynamic flow fields associated with detonation waves, as reflected in the books of Courant & Friedrichs (1948), Sedov (1959), and Landau & Lifshitz (1959) and the series of monographs edited by Emmons (1958).

The placid image of a plane, steady-flow detonation wave, whose progress and structure are virtually unencumbered by the kinetics of fast chemical reactions, was shattered in the nineteen sixties. The first evidence contributing towards this revolution of the classical concept was provided by the independent studies of White (1961) in the United States and Voitsekhovsky et al (1958) and Denisov & Troshin (1959) in the Soviet Union; they demonstrated that the detonation wave is intrinsically multi-dimensional in structure and inherently nonsteady in character. White made his observations on the basis of sharp photographic records obtained by means of a Mach-Zehnder interferometer with a short-duration electric spark as the light source; Denisov and Troshin employed a carbon-soot technique for which, in a manner reminiscent of the gallantry of Mallard and Le Chatelier, they gave credit to Mach (Mach & Sommer 1877). This novel and relatively simple means for the study of detonation phenomena provided great impetus to experimental research in the field, with the work of Schott (1965a,b), Strehlow (1969), Mitrofanov & Soloukhin (1964), Topchian (1962), and Edwards, Parry & Jones (1966) representing the most prominent examples. The progress made in this respect was summarized in a number of books (Shchelkin & Troshin 1963, Soloukhin 1963, Voitsekovsky, Mitrofanov & Topchian 1963) and review papers (Oppenheim, Manson & Wagner 1963, Oppenheim 1965, 1967, Macpher-son 1967, Lee, Soloukhin & Oppenheim 1969, Strehlow 1968).

The most important feature of this research was the particular attention given to transient phenomena. Before going into the details of the wave structure that emerged, let us consider first its most nonsteady feature: the formation. The development of the detonation wave in a slender tube was studied in particular by Salamandra, Bazhenova & Naboko (1959), Soloukhin (1961), and Oppenheim and his associates (Laderman & Oppenheim 1962, Urtiew & Oppenheim 1966). Following a laminar flame, the sequence of events associated with this process has been established essentially as follows:

(1) initial acceleration associated with wrinkling of the front surface;

(2) onset of turbulent front structure, when the flame acquires a characteristic "tulip" shape, followed by a relatively long period of turbulent flame propagation;

(3) actual transition to detonation triggered by "explosion in the explosion."

The last step is prompted by a boot-strap process of (1) the generation of pressure waves by the accelerating flame, (2) the increase of flame turbulence due to flow acceleration induced by the pressure waves, and in turn, (3) an increased intensity of the generation of pressure waves by the more turbulent flame. This process is depicted in Fig. 2 (reproduced from Meyer, Urtiew & Oppenheim (1970), representing a cinematographic sequence of schlieren photographs obtained by a stroboscopic laser system, described by Hecht, Steel & Oppenheim (1966), that uses an optical system developed by Oppenheim, Urtiew & Weinberg (1966). The medium was a stoichiometric hydrogen-oxygen mixture contained initially at a pressure of 1 atm, in a $1 \times 1,5$ -in. cross-section tube. The origins of both the time and the distance scales on the figure are arbitrary, the test section being actually about 10 feet away from the spark-gap located at the closed end and the recorded events occurring about 2 msec following ignition.

As the flame enters the test section in the top frame of Figure 2, it is already preceded by a number of shock fronts that were generated at earlier stages of flame acceleration. The shocks merge, causing further flow acceleration, and the intensity of turbulence increases, reinforcing the combustion process, so that finally the flow field ahead of the flame becomes full of pressure waves that are on the verge of collapse into new shocks, as is quite evident in the last frame. This, in turn, leads to an explosion in the exploding medium, as seen in Figure 3 (reproduced from Meyer, Urtiew & Oppenheim 1970), which represents a record of events directly following those of the previous figure. Actually, two such "explosions in the explosion" were recorded there, one appearing first at 715 µsec on the time scale and about 20 cm on the distance scale, the second at 730 µsec and approximately 30 cm. In this particular case both of the secondary explosions produced detonation fronts that are clearly visible as they propagate to the right in the lower frames. The detonation due to the first explosion will, of course, decay later, since the other, propagating ahead of it, will eventually consume all the unburned mixture, leaving only inert products behind. A double explosion, such as that recorded here, is, of course, a relatively rare occurrence, a single "explosion in the explosion" being in most cases quite sufficient to trigger the transition to detonation.

The duration of the process of the development of detonation, or the length of the so-called detonationinduction distance, is greatly prolonged if the pressure is decreased, the temperature increased, or a diluent added. In short, the rate at which the process develops is a sensitive function of the power density at which energy is released by the exothermic reaction of the chemical substance enclosed in the tube. It should be noted in this connection that in an unconfined space the development of detonation is also a sensitive function of the specific power of the initiation process, as described later in the section on Blast Waves.



Fig. 2 Cinematographic laser-schlieren records of the generation of pressure waves by a turbulent flame in a stoichiometric hydrogen-oxygen mixture maintained initially at a pressure of 1 atm in a $1 \times 1,5$ -in. cross-section tube



Fig. 3 Cinematographic laser-schlieren records of "explosions in the explosion" triggering the transition to detonation—a sequence of processes subsequent to those recorded in Fig. 2

Salient features of the multiwave structure of the detonation front are depicted in Figure 4. Figure 4a is an interferogram obtained by White of a self-sustained detonation wave in a stoichiometric hydrogen-oxygen mixture at an initial pressure of about 0.1 atm. Figure 4b shows soot traces, similar to those recorded by Denisov and Troshin, etched on the bottom wall of a $1 \times 1,5$ -in. cross-section tube, together with the corresponding cinematographic laser-schieren photographs of the wave as it propagates in the tube across the 1-in. side of the test section, obtained at Berkeley with the same medium and approximately the same initial conditions as in Figure 4a.



Fig. 4. Records demonstrating the multidimensional structure of the detonation wave

(a) Interferogram obtained by White with a stoichiometric hydrogen-oxygen mixture at an initial pressure of 100 torr

(b) Soot record obtained on the 1,5-in. sidewall and corresponding laser-schlieren photographs taken at a 5- μ sec interval across the 1-in. side of a 1×1,5-in. cross-section tube with a 4H₂+O₂ mixture at an initial pressure of 84 torr

To bring out the detailed structure of the wave, one has to decrease the power density of energy release. This is accomplished simply by diluting the reactive mixture with an inert gas such as nitrogen or argon. If one of the glass windows is covered with a thin layer of soot and the schlieren stop is removed, the cinematographic laser system yields a sequence of direct shadow photographs of the detonation front (propagating from left to right) combined with the traces (which remain stationary) that it has etched on one of the sidewalls. Such a record of a stoichiometric hydrogen-oxygen mixture contained in the $1 \times 1,5$ -in. cross-section tube initially at a pressure of about 0.1 atm. with nitrogen as a diluent, is presented here in Fig. 5. It appears clearly that the detonation front consists, in effect, of a set of curved intersecting shocks, while the traces on the sidewall delineate the trajectories of the intersection points. Since there are two such trajectories within the cross section of the tube, the case recorded in this figure is considered to represent a double-mode structure of the wave.

On a number of frames in Figure 5 one can observe double records of the front. This is due to the fact that its surface is actually distorted by wave-intersection processes similar to those recorded by soot imprints but propagating in the direction normal to the wall. It is this phenomenon, in fact, that produced the splashes that appear quite clearly in the soot record of Fig. 4b. The two records of the shock front correspond to the intersections of the shock surfaces with the two sidewalls of the test section. This is due to the fact that traveling shock fronts "can approach the wall only at right angles, so that their surfaces become then parallel to the axis of the optical system, exposing to it the highest refractive-index changes that produce the two shadow records.

Among other techniques, high-frequency pressure transducers and open-shutter photography are much used in modern detonation research. Both of these were described in some detail in the book of Soloukhin (1963). Best results for open-shutter photography are obtained with the use of an equimolar acetylene-oxygen mixture contained in an about 1/8-in.-wide channel at an initial pressure of the order of 0.1 atm. An example of such a record is given in Fig. 6, which corresponds to a triple-mode structure of the wave.



Fig. 5. Cinematographic laser-shadow photographs of a double-mode detonation in a nitrogen-diluted stoichiometric hydrogen-oxygen mixture at an initial pressure of 0.1 atm



Fig. 6. Open-shutter photograph of a triple-mode detonation in an equimolar acetylene-oxygen mixture in a $1/8 \times 1$ -in. cross-section duct

The records described so far represent just a sample of the great amount of experimental results that were obtained throughout the nineteen-sixties. Out of this emerged a novel understanding of the detonation-wave structure, which, in a simplified form, to emphasize the essentials, is shown in Fig. 7. The front at a given instant of time is represented by a continuous line, horizontal dashes behind the front denote zones of exothermic reactions, and broken lines are traces of the intersection points. The latter are, in fact, triple points of Mach reflections between the two head shocks as indicated by T on the diagram. The reason they etch their traces is associated with the effect of the slip line generated by the intersection. Since the line separates regions of different flow velocities, the flow on one side being supersonic while that on the other is subsonic, it gives rise in a real fluid to a considerable amount of shear that in turn creates a concentrated vortex of high-temperature and high-pressure gas behind the Mach stem. It is the rotating stylus action of such a vortex that produces the traces.



Fig. 7.Schematic diagram of a two-dimensional detonation-wave structure

The strongest shocks at the front are formed at the center line by collisions between triple points. They create regimes of highest temperature and density and, hence, fastest chemical reactions. The chemical induction times are then at their minimum, and the zones of exothermic processes are closest to the shock fronts. It is at these places that the wave system must receive the greatest impulse for its propagation, as indicated by the arrows in Fig. 7. Farther away from collision points the system is virtually in the process

of decay. The intensity of the shock front decreases, while the exothermic reaction zone lags more and more behind the shock, so that there are good reasons to believe (Lundstrom & Oppenheim 1969) that in most detonations it becomes completely decoupled before the next collision takes place. This periodic nature of the propagation process is actually reflected in the record of Figure 6, where, evidently, most of the illumination was obtained immediately after collisions, while later on the decay progressed so far that there are regions of complete darkness.

The most striking feature of these novel concepts concerning detonation phenomena is the realization that, in paradoxical contradiction to classical views, the process is strongly dependent on chemical rate processes. Not only is the local change in the rate extremely rapid, but the overall propagation of the wave is evidently controlled by the number density of collisions between triple points, the events that produce the largest changes in the local reaction rates. Concomitant to this is the appreciation of the role played in the detonation process by the power density, or specific power, at which energy is deposited in the compressible medium. It is, indeed, in this sense that the use of diluents affects the number density of triple points which, as has been shown by Strehlow (1970), is quite dependent on this parameter. Locally, according to the description of the wave-propagation process represented by Fig.7, the specific power of energy release must be considerably higher immediately after a triple-point collision than later when all rate processes are greatly slowed down.

Shock-Wave Research

A variety of nonequilibrium high-temperature molecular processes, such as vibrational excitation of molecules, dissociation, or chemical reactions, can be initiated easily by a shock wave. The nonisentropic compression and heating of the gas by the wavefront can be quite rapid, taking place in a transition time as short as 10-10 sec. This unique facility offered by the shock-tube technique has been widely employed for experimental studies in chemical physics of high-temperature phenomena. It should be noted, however, that with reference to chemical reactions it has been utilized primarily for the investigation of isothermal processes, such as those occurring during the induction period, while the exothermic process, representing after all one of their most significant features, remained to a large extent virtually unexplored.

It was in fact the detonation research that was most instrumental in showing both the significance of the dynamic properties of the exothermic process and the remarkable capability of the shock-tube technique as the most suitable means for their study. Research efforts in this respect have been concerned with the investigation of chemical kinetics associated with the induction process, using the shock-tube technique (Schott & Kinsey 1958, Skinner & Ringrose 1965, Gardiner & Wakefield 1970) as well as stationary shocks in a supersonic flow field (Gross 1959, Nicholls 1963, Rhodes, Rubins & Chriss 1964, Rubins & Rhodes 1964, Rubins & Cunningham 1965, Richmond & Shreeve 1967), and with its influence on the nonsteady detonation phenomena such as the transition to detonation (Saytzev & Soloukhin 1958,1962, Voevodsky & Soloukhin 1965), its multi-wave structure (White & Moore 1965), spin (Borisov & Kogarko 1963), and limits (Patch 1961).

One of the most interesting results to emerge from all this is the novel concept of the strong-ignition limit—a line of demarkation between two different modes of autoignition on the classical explosion plane of initial pressures and temperatures. At lower temperatures the occurrence of autoignition manifests itself first in the form of a flame kernel, while at higher temperatures it appears first as a blast wave associated with a shock front. The blast wave is formed by the higher specific power the exothermic process attains under such conditions; this phenomenon, in fact, explains the "explosion in the explosion"—the necessary factor in the transition to detonation that was described here in connection with Fig. 3.

The strong-ignition limit is due to the same physical process as that governing the classical secondexplosion limit: the onset of the chain-branching mechanism. But, as is typical of criteria for gasdynamic explosions, it depends on a specific value of the rate at which the induction time, terminated by this phenomenon, changes with temperature, or more specifically $(\pi\tau/\pi T)p$, where τ is the induction time, T the temperature, and p the pressure, rather than on the value of the induction time itself that determines the second-explosion limit. This property is demonstrated in Fig. 8, reproduced from the paper of Meyer & Oppenheim (1971a). Besides the classical explosion limit, the diagram contains lines of constant induction time, and constant gradient of induction time with respect to temperature, both evaluated on the basis of kinetic schemes deduced from shock-tube data (Skinner & Ringrose 1965, Gardiner & Wakefield 1970). Included also are a number of schlieren photographs to illustrate the significant difference between the mild and strong ignition. The strong-ignition limit delineating the regimes of these two modes of ignition is represented by the shaded zone, from which it appears that for the hydrogen-oxygen mixture for which this diagram was constructed this limit corresponds specifically to $(\pi\tau/\pi T)p = -2 \mu sec/K$. Subsequently, the reason for this criterion was rationalized by Meyer & Oppenheim (1971b) on the basis of the coherence requirement for the onset of exothermic reaction centers whose dynamic properties were established earlier by Zajac & Oppenheim (1971).

More recently, it has been established by Vermeer, Meyer & Oppenheim (1972) that, for hydrocarbon fuels, the strong-ignition limit coincides with a line of $(\pi/\ln\tau/\pi T)p = \text{const}$ rather than $(\pi\tau/\pi T)p = \text{const}$. In fact, for stoichiometric n-heptane-oxygen and i-octane-oxygen mixtures with 70% argon dilution, the value of this parameter was found to be in the vicinity of -0.01K-1. This is in conceptual agreement with the Shchelkin criterion, which, as described in the next paragraph, is considered to govern the onset of flow instabilities in a gas mixture undergoing an exothermic chemical reaction. The fact that seemingly different criteria apply to the strong-ignition limit for different chemical systems was explained on the basis of the analysis of Meyer & Oppenheim (1971b), from which it appears that the criterion should actually be proportional to δ -1($\pi\tau/\pi$ T)p, where δ is the duration of the exothermic power pulse (see Fig. 10), as measured by, say, its half-width or rise-time, and that therefore for the hydrogen system δ is evidently relatively invariant, while for the hydrocarbon mixtures it is in effect proportional to τ .

Flow instabilities that are set up behind shock waves in exothermic systems have been a subject of extensive investigations using experiments performed in ballistic ranges filled with explosive gas mixtures (Ruegg & Dorsey 1962, Behrens et al 1965, Chernyi 1968, Chernyi & Gilinskii 1970, McVey & Toong 1971). More recently, a criterion for the onset of such instabilities has been deduced by Soloukhin & Brochet (1972) on the basis of shock-tube experiments using argon-diluted equimolar mixtures of NH3 and O2. The criterion is based on the postulate proposed by Shchelkin (1959) that the instabilities occur when the change in the induction period becomes, owing to a sufficiently high rate of energy release (or, more specifically, exothermic power density), of the same order of magnitude as the induction time itself. As an example of the manner in which this investigation was conducted, experimental records showing the development of flow instabilities behind a shock wave are represented in Fig. 9.

To illustrate recent trends in the study of exothermic processes by the use of shock waves, let us consider, for example, the progress made in the kinetics of combustion systems using nitrous oxide as an oxidizer (Lin & Bauer 1969, Henrici & Bauer 1969, Garnett et al 1969, Soloukhin 1971). In such systems a chain reaction is initiated by the oxygen atoms generated by the bimolecular collision step N2O+M \rightarrow N2+O+M, for which the rate constant k1 is well known (Jost et al 1964–1966, Borisov 1968). The oxygen atom is consumed by subsequent collision with nitrous oxide, forming N2+O2 or 2NO. The respective rate constants k2 and k3 were also determined by Garnett et al (1969) and by Lin & Bauer (1969), enabling a quantitative evaluation of the complete system associated with the generation of atomic oxygen. It was then demonstrated (Soloukhin 1971) that, in several reacting systems involving hydrocarbons as fuels, the temperature coefficient of the total rate of the initiating step is determined essentially by the difference between the activation energies of the first and second or of the first and third step, both being of the order of 33 kcal/mole. However, when hydrogen is used as fuel, then at moderate temperatures the process is governed by the competitive consumption reaction: H+N2O \rightarrow OH+N2. This reaction mechanism was

studied quantitatively by Henrici & Bauer (1969), who were able on this basis to obtain quite accurate data on rate constants for other elementary steps in the reaction scheme for which, until then, only rough information had been available.



Fig. 8. Mild and strong explosion regimes as a function of the pressure and temperature of the induction process. The "strong-ignition limit" delineating the boundary between the two regimes is represented by a horizontally cross-hatched zone

The advantages offered by the shock-tube technique in gathering such data should be considered today of particular importance in view of the availability of computer facilities that can be exploited to determine the reaction mechanism in complex systems.

As to the insight into the exothermic zone itself, an excellent means has become available recently in the form of laser-shear interferometry, developed by Jones, Schwar & Weinberg (1971). The essential requirement for the success of this technique is a certain minimum coherence length of the light beam. As a consequence of this property, any point in the train of wavefronts maintains, over a certain length of the optical path, an invariant phase relationship with all other points. Thus, if the beam is split into two parts that are then superimposed over each other with a certain amount of overlap or "shear," an interference pattern in the region of overlap is obtained. If this region is selected in such a way that one component contains the part of the test section with phase disturbance while the other is devoid of it, as for instance the flow field ahead of a shock front, one obtains a so-called "finite-shear" interferogram of the disturbed field itself. The maximum attainable amount of shear is limited by the minimum available coherence length of the light beam.



Fig. 9. Interferograms and pressure records of a set of shock-reaction-zone systems in mixtures of X (0.5NH3+0.5O2) + (1 - X)Ar (time base marked in 10-µsec intervals), (a) X = 0.15, p1 = 0.01 atm, M1 = 5.6: stable one-dimensional shock, induction zone, and exothermic rarefaction wave (deflagration), (b) X = 0.175, p1 = 0.01 atm, M1 = 5.5: steady induction zone and nonregular exothermic wave, (c) X = 0.175, p1 = 0.03 atm, M1 = 4.7: both induction zone and exothermic wave are distributed, (d) X = 0.2, p1 = 0.01 atm, M1 = 5.8: shock front becomes nonplanar and is followed by strong pressure waves, causing nonsteady hock propagation, (e) X = 0.2, p1 = 0.03 atm, M1 = 5.25: unstable, multidimensional detonation wave

In practice, the splitting of the beam can be accomplished by means of a diffraction-grating/lens system combined with a stop that lets through only the two first-order diffraction beams. The grating has a spacing of about 40 lines per millimeter, and the interferometric pattern is recorded in the form of a distorted grating on a film emulsion of an equivalent space resolution. In order to obtain an explicit interferogram, one has to use an additional exposure of the test section without the presence of the event to be observed. The final result obtained in this manner is then produced in the form of the Moire pattern of the records of the two exposures, or, more exactly, a combined grating that is selectively obliterated by the Moire pattern. This is finally reconstructed by the use of a simple lens system provided with a stop that lets-through only the first-order diffraction beam, with all the nonessential information thus filtered out.

Added flexibility is attained by the use of two photographic negatives, instead of one, to record separately the reference and test exposures. In this way one can reconstruct from a single record of the observed events a schlieren photograph, as well as a zero-fringe or a finite-fringe interferogram, the latter with any fringe spacing one wishes to obtain. Thus one can trace sharply the loci of maximum gradients of refractive index, typical of schlieren records; the loci of constant optical paths corresponding usually to lines of constant density, as in zero-fringe interferograms; and, finally, the space variation in optical paths, as in finite-fringe interferograms, usually yielding the density profiles, but this time with an adjustable space resolution across the test section due to the controllable fringe spacing. An example of such records,

with the deduced heat-release profile and the corresponding exothermic power pulse, is provided by Fig. 10.

As the shock-wave and shock-tube techniques become more developed, it becomes possible to exploit a whole variety of complex wave-interaction processes for studies of nonequilibrium molecular phenomena in high-temperature gases. Fast expansion of a shock-heated reactive-gas sample by passing the gas through a rarefaction wave or nozzle provides, for instance, a highly effective means for quenching a nonequilibrium relaxation process associated with



Fig. 10. Laser-shear interferograms of an exothermic reaction zone behind a reflected shock in an argon-diluted stoichiometric hydrogen-oxygen mixture and the deduced heat-release and exothermic-power-pulse profiles

either a chemical reaction or a molecular or radiative excitation. Various "negative-temperature-impulse" techniques have been developed in the studies of recombination phenomena and rapidly "frozen" chemical reactions (Resler 1966, Hurle & Russo 1967, Soloukhin 1967), and extremely rapid gas-cooling rates of an order of 106–108 K/sec have thus been achieved.

The most exciting application of these methods has come to the fore in connection with the development of gasdynamic lasers. On the basis of earlier suggestions that established the basic principles for the achievement of the population inversion necessary for laser emission in a rapidly cooled molecular system (Hurle & Hertzberg 1965, Konyukhov & Prokhorov 1966, Basov et al 1963), gasdynamic lasers using shock-tube techniques have been successfully developed (Kuehn & Monson 1970, Gerry 1970–1971, Dronov et al 1970, Russell et al 1971, Karniushin & Soloukhin 1972) with the use of thermally pumped CO2-N2-He gas mixtures.

In such an apparatus, an equilibrium gas mixture heated behind the reflected shock wave is expanded through a supersonic nozzle into a laser cavity. Here a vibrationally nonequilibrium system of carbon dioxide and nitrogen molecules is created in the gas flow as a result of the rapid gasdynamic cooling of the gas downstream of the nozzle in an expansion time that is shorter than the corresponding vibrational relaxation times of the upper laser levels of the molecules. By the use of helium or water as a catalyst, the lower laser level is depopulated in less time than that required for expansion through the nozzle. The shock-tube technique thus furnishes an effective simulation of the process, yielding fundamental data for the development of a continuous-wave gasdynamic laser of high power output.

Shock-tube experiments provide basic information on both power and negative absorption (gain) obtainable in a gasdynamic laser. Thus at the Institute for Pure and Applied Mechanics in Novosibirsk we were able to obtain laser power output at about the one-hundred-watt level in a shock tube of 3.5×13 cm cross section and nozzle-throat ratio of 16-30, at an equilibrium gas pressure of 8 atm and temperatures of 1200-1600 K.

Explosion and detonation phenomena have also been used in gasdynamic-laser experiments. Direct observations of the population inversion behind an incident overdriven detonation wave propagating in a mixture of F2O+H2 have been made by Gross et al (1969). It should be noted also that direct creation of population inversion behind a normal shock wave was predicted to be achievable by means of shock-initiated dissociation or vibrational excitation (Oraevskii 1965, Anderson & Madden 1971). In the latter case, for instance, two laser transitions (50 and 20μ) may be created in carbon dioxide by molecular vibrational-energy exchange only. At actual experimental conditions, only relatively wide non-equilibrium zones are accessible in these cases, since the diffraction losses from the laser cavity make the lasing regime quite critical.

In explosion-pumped gasdynamic lasers (Yatsiv et al 1971, Tulip & Seguin 1971 a,b, Dzhidzhoevetal 1971), the detonative combustion of various systems was used, yielding a temperature of 1000–1700 K of the products ahead of the nozzle at stagnation pressures of 5–15 atm. Since a small amount of water concentration ,is admissible in the lasing molecular systems, a whole variety of combustion systems was found to be appropriate, as, for instance, HN3, CO, C2H2, C6H6, C3H8, etc, mixed with oxygen and nitrogen (Meinzer 1971, Tulip & Seguin 1971a,b).

Thus, relatively simple means for effective direct conversion of heat to coherent radiation is available today as a consequence of the application of the shock-tube technique to the development of thermally pumped gasdynamic lasers. All this work was instrumental, moreover, in initiating a variety of important fluid-dynamic studiea of steady as well as nonsteady expansion flows involving a number of lasing nonequilibrium molecular gas systems. The resulting experimental investigations, combined with calculations of the nonequilibrium flow systems, should lead to further progress in the acquisition of important molecular-gas-kinetics data, as has been the case in this field over the last two decades.

Blast-Wave Studies

Experimental studies of blast waves were handicapped for a long time by the difficulties associated with their initiation at a sufficiently small scale to be contained under laboratory conditions.

The traditional means used for the deposition of energy, i.e., electric sparks or exploding wires, were inadequate as a consequence either of the relatively long time required to discharge the energy and the excessive amount of space over which it was deposited, or of the resulting chemical and electronic debris that tended to contaminate the flow field. This situation has been changed drastically by the advent of pulsed lasers. Since the initial announcement of Terhune (1963) that electrical breakdown can be induced in a gas by the focusing of a Q-spoiled laser pulse, the subject has received a great deal of attention, as reflected in a comprehensive review paper of Meyerand (1967). Even though the exact breakdown mechanism is still a controversial matter, it has been established that the initial presence of a few free electrons is an important part of the process. At low densities, however, these free electrons are in short supply and it becomes necessary to introduce a metal wire close to the focal point to provide electrons that are stripped off by the electric field at the focus (Lee & Knystautas 1969, Bach, Knystautas & Lee 1969). An example of a laser-induced breakdown in air at atmospheric pressure, where the initial ion concentration is sufficiently high to render the use of an artificial source in the form of a metal surface unnecessary, is shown in Figs. 11 and 12. Fig. 11 is an open-shutter photograph of the spark exhibiting its characteristic-elongated shape (Meyerand 1967, Weinberg & Wilson 1971), while Fig. 12 is a set of

cinematographic schlieren photographs, taken at l-µsec intervals, recording the development of the blast wave in the presence of a plasma ball that retains an essentially constant radius after it becomes detached from the shock front. (The sequence in which the consecutive frames are presented is from top to bottom in successive rows from left to right.) As becomes evident from this record, although the 20- to 50-nsec duration of energy deposition can be considered to have been virtually instantaneous in comparison with the l-µsec interval between the frames, the relatively long life of the plasma ball



Fig. 11. Open-shutter photograph of an electric breakdown produced by a laser pulse in air at a pressure of 1 atm

tends to contaminate the medium as well as complicate the pattern of energy release, causing the resulting gasdynamic flow field for all practical purposes to be analytically untractable.



Fig. 12. Cinematographic laser-schlieren photographs of a blast wave generated by laser breakdown recorded in Fig. 11. Initial pressure 1 atm; time interval between frames 1 μ sec. The sequence of events is from top to bottom, left to right. Seen in the background are two vertical marking lines set at a distance of 2 in

An alternative method that circumvents this difficulty is the use of a thin wire placed in the path of the laser bearm at a small distance from the focal point. No electric breakdown takes place in this case; instead the irradiation of the wire results in a local sublimation associated with a small plume of vaporized metal (Ready 1963, Honig 1963, Honig & Woolston 1962), as shown in Fig. 13. However, the

accompanying schlieren record, Fig. 14, taken in an equimolar acetylene-oxygen mixture at 120 torr, indicates a virtual absence of such a plume. Apparently, in view of the small size of the laser beam close to the focal point, the wire acts as an extremely efficient heat sink for the vaporized metal, causing its rapid recondensation and thus leaving the flow field relatively free of contaminants.

The advantage of this method lies in its relative simplicity and low cost; the disadvantage is associated with its limitation to relatively weak blast waves (front Mach number being at most of the order of 2). This difficulty can be circumvented by resorting to the use of a picosecond, mode-locked, gigawatt laser. Such an apparatus is expensive both in initial cost and in operation. It has, nonetheless, been used with great success by Bach et al (1971) to evaluate induction periods in a nitrogen-diluted stoichiometric acetylene-oxygen system under the non-isothermal conditions existing in the blast wave.

Laser-initiated blast waves were used by Lee and his associates (Lee & Kny-stautas 1969, Bach, Knystautas & Lee 1969) to study the transition to detonation under spherical flow conditions. They were able to demonstrate the critical influence on the development of the process of the power density at which the initiation energy is deposited in the explosive gas. This is illustrated by two sets of schlieren records, reproduced in Fig. 15. The first set represents the sequence of events associated with a subcritical level of initiation energy in an equimolar acetylene-oxygen mixture. As the process progresses, the combustion front separates from the shock wave, and from then on the distance between the two increases. Such a wave system is actually in a state of decay. The reaction zone soon becomes completely decoupled from the shock front, whose strength continues to decrease as if it were a front of a blast wave of an essentially constant energy content. With the use of a focused laser beam as the igniter, the power density of ignition energy can be varied simply by modifying the energy of the laser pulse. Thus, in the records presented in Fig. 15b, the specific power of ignition energy was just sufficient to promote the onset of a secondary explosion that produced a wave-intersection process that eventually engulfed the head shock front, thus preventing the separation of the combustion front from the shock that led to the decay in the previous case.

More recently, Soloukhin & Ragland (1969) demonstrated that similar effects can be obtained by using the exhaust of a small detonation tube as an explosive igniter. The sequence of schlieren photographs obtained by them with the use of a stoichiometric hydrogen-oxygen mixture is shown in Fig. 16a, while the corresponding records obtained with an equimolar acetylene-oxygen mixture at the same initial pressure and temperature are given by Fig. 16b. The process recorded in Fig.16a evidently corresponds



Fig. 13. Open-shutter photograph of a metal-plume explosion produced by a focused laser pulse in air at 120 torr

to that of Fig. 15a, while the records of Fig. 16b are, in effect, similar to those of Fig. 15b.



Fig. 14. Cinematographic laser-schlieren photographs of a blast-wave combustion-front system produced in an equimolar acetylene-oxygen mixture at an initial pressure of 120 torr by the plume explosion shown in Fig. 13



Fig. 15. Ignition of an equimolar acetylene-oxygen mixture produced by a laser pulse. (a) Case of subcritical ignition yielding a decaying blast-wave combustion-front system. (b) Case of a just-supercritical ignition, when the decay is prevented by the onset of a secondary explosion that leads to the establishment of a multidimensional detonation front

The novel experimental findings described in this section were associated, of course, with significant advances in blast-wave theory. However, their description is outside the scope of this paper; moreover, they were reviewed recently by Korobeinikov (1971). It is of interest to note, though, that most of the current studies he describes are concerned explicitly with the interpretation of the phenomena presented in the last two figures.



Fig. 16. Ignition of an explosive gas mixture produced by a detonation wave. The small detonation tube used as the igniter was 10 mm i.d. and 13 mm o.d. (a) Subcritical ignition obtained in an equimolar hydrogen-oxygen mixture at an initial pressure of 0.07 atm. (b) Supercritical ignition obtained in an equimolar acetylene-oxygen mixture at the same pressure as (a)

Closing Remarks

In making our way through the various topics concerning recent accomplishments in experimental studies of gasdynamics of explosions we went full circle, ending with a record pertaining to detonation phenomena, which were considered at the outset. This emphasizes the close interrelationship between the three major fields of scientific endeavor of which our subject is primarily composed: detonations, shocks, and blast waves. Of course, this is not surprising since all three are intimately involved in most explosions. The fact, however, that they are now being brought together to bear upon the study of this subject in a more coordinated way than ever before should have a profound influence on its future development.

The most important feature of current studies is the particular attention paid to transient processes and the concomitant progress made in the development of novel experimental means especially suited for this purpose. In particular, in striking contrast to classical concepts, it became clear that the exothermic processes associated with fast chemical reactions are in a significant impedance mismatch with the gasdynamic flow phenomena so that, except for the energetically trivial cases of extremely low specific powers, the two cannot coexist under steady-flow conditions. Associated with this is the realization that chemical kinetics plays a vital role in detonation phenomena, in spite of the fact that its influence on the overall propagation rate is quite insignificant. As a consequence of this notion our subject has become of central importance to the study of the dynamic properties of exothermic processes, whose understanding has so far been seriously handicapped by the lack of adequate experimental facilities.

The most exciting prospects for the future are associated with possibilities of exploiting the knowledge of explosion phenomena for the development of such interesting devices as the gasdynamic laser and the apparatus based on the use of lasers to achieve controlled thermonuclear reaction. Despite the obvious one-sidedness of our review, restricted as it is so much to the description of experiments performed in our own laboratories, we hope that bringing together the most critical features of the subject will be of assistance to a reader engaged in one of these fascinating endeavors.

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