Study of Detonation in Condensed Explosives by One-Dimensional Channel Flow

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The detonation of mixtures of 80/20 ammonium nitrate/trinitrotoluene was studied using pseudo one-dimensional channel flow. A layer of explosive was confined symmetrically by two relatively massive metal plates whose shock velocity was higher than the detonation velocity of any of the explosive mixtures studied, thus producing a close approximation to a steady-state two dimensional detonation. From measurements of the plate velocity the flow could be analyzed as a pseudo onedimensional channel flow. The quantity $(dP/dp)^{\dagger}$ was found to have a maximum which served as an approximate experimental measure of the location of the Chapman–Jouguet point. The observed dynamic quantities were in substantial agreement with the Zeldovich–von Neumann–Doering model. Variation of the particle size of the ammonium nitrate produced a complex effect on reaction time, which could be adequately interpreted in terms of the three reaction steps in the detonation. Flash radiography provided a check on the one-dimensionality of the flow and allowed a rough estimate of the density profile behind the front, which was compared with the optical results.

INTRODUCTION

HE description of a detonation wave as a reaction initiated by a shock wave was put forward by Zeldovich¹ in 1940, and independently by von Neumann² and Doering³. The description was simplified by these idealizations: The system was assumed to be one-dimensional, and the detonation wave was assumed to consist of three separate parts-a shock wave, a steady reaction zone in which the reaction proceeds to completion, and finally a rarefaction wave which satisfies the rear boundary conditions. If a granular solid explosive is being treated as a continuous medium, the shock front must extend over a distance equal to several times the particle diameter. For fast reaction rates, the overlap of reaction zone and shock front could be important. The rarefaction may be eliminated by sustaining the detonation by a piston or in principle by supplying a suitable starting transient to form the steady reaction zone and to place the leading edge of the rarefaction wave far enough behind the shock front to allow essentially complete reaction, but only if one-dimensionality can be maintained through the steady zone.

In practice a rarefaction wave due both to lateral expansion and to the effect of the rear boundary generally overlaps the reaction zone, thus increasing the difficulties encountered in studying the reaction zone of detonations in condensed explosives. Part of the difficulty in studying the reaction zone is due to the fact that when the detonation closely approaches the one-dimensional steady-state Zeldovichvon Neumann-Deoring model, the measuring devices used to detect the reaction zone introduce transient or three-dimensional effects. In previous measurements of this type⁴ involving end-on impact of the detonation against a metal plate the impedance of the metal was not exactly equal to that of the reacting explosive, and a wave was reflected from the interface back into the detonation products. Despite the disturbance by the reflected wave, this and similar measurements of the reaction zone length have been the most reliable ones available for condensed explosives. Measurements made on long columns of explosive by x-ray transmission, optical interference, or wall pressure gages all suffer to some extent from deviations from one-dimensionality. In detonations in long columns of solid or liquid explosives, a close approach to a steady state may be attained, but an appreciable deviation from onedimensionality is caused by lateral expansion, which is appreciable even for charges confined by heavy metal walls. For unconfined or lightly confined solid explosives, the effects of lateral expansion are quite large.

Most of the experimental determinations of the reaction zone length in condensed explosives have been indirect. The experimental data comprise values of detonation velocity as a function of charge size, with the explosive either unconfined or confined by

¹ Y. B. Zeldovich, Zh. Eksperim. i Teor. Fiz. 10, 542 (1940). ² J. von Neumann, Office of Scientific Research and Development Report No. 549 (1942).

³ W. Doering, Ann. Physik 43, 421 (1943).

⁴ R. E. Duff and E. Houston, J. Chem. Phys. 23, 1268 (1955).

walls of different impedance. A number of approximate theories⁵⁻⁷ have been developed to relate data on the detonation velocity versus diameter to the reaction zone length. In most cases, and especially for unconfined charges, the system is extremely complex, consisting of a collection of stream tubes which differ widely in character. The situation is further complicated by the fact that the rate of chemical reaction depends strongly on the variables (e.g., temperature and pressure) which are influenced by the flow, and in turn the flow is strongly influenced by the energy released by chemical reaction.

To attempt a separation of the interacting effects of lateral expansion and chemical reaction, it is desirable, and perhaps even necessary, to observe the behavior of a single stream tube. The experimental difficulties in measuring the properties of an internal stream tube can be avoided if a system can be selected in which the flow is nearly the same throughout the cross section of the explosive, i.e., a system in which all the explosive lies in a single stream tube. Such a system can be analyzed as a pseudo one-dimensional channel flow with varying cross-sectional area. The requirements to be met are that the area must change gradually and the walls must be free of sharp changes in angle. The gradual area change can be insured by confining the explosive in realtively massive walls of high impedance material. A sharp angle will, however, occur at the line of contact of the detonation front with the wall if the detonation velocity appreciably



FIG. 1. Method of measurement of plate angle by the reflection of the image of a grid from the outside surface of the confining plate assembly.

⁵ H. Eyring, R. E. Powell, G. H. Duffy, and R. B. Parlin,

Chem. Rev. 45, 69 (1949). ⁶ H. Jones, Proc. Roy. Soc. (London) A189, 415 (1947). ⁷ M. A. Cook, *The Science of High Explosives* (Reinhold Publishing Corporation, New York, 1958), pp. 125–128.

exceeds the weak shock velocity (or plastic sound velocity) of the wall material. In this case, the wall will be deflected by an oblique shock attached to the detonation front. On the other hand, if the detonation velocity is less than the sonic velocity of the wall material, the steady-state flow configuration will exhibit no shock waves in the wall, and the boundary of the explosive will have no discontinuities in angle. For example, with stainless-steel walls the detonation velocity must not exceed 4.5 mm/µsec, so that military explosives at high density cannot be studied in this way. Most explosives at densities less than 1.0 g/cc, and many commercial explosives of somewhat higher density, have sufficiently low detonation velocities to be studied.

EXPERIMENTAL

Experimental Method

The system chosen to meet the requirements for the application of a one-dimensional channel flow treatment consisted of a symmetrical sandwich of two parallel flat metal plates containing a layer of explosive, as shown schematically in Fig. 1. The explosive was initiated along the top edge by a linewave generator. The length and width of the explosive and plates were much greater than the thickness. Observations of plate motion were made as described below in a region half way between the lateral edges and near the end (bottom) away from the initiation end. The edges were confined by heavy steel bars. A typical assembly was 8 in. wide \times 12 in. high with a $\frac{1}{2}$ -in.-thick layer of explosive confined between two Type 304 stainless-steel plates each $\frac{1}{4}$ in. thick. Stainless steel was a reasonable compromise for the requirements of high density, high sonic velocity, and relatively low elastic strength.

A direct measurement of the velocity of the plate as a function of distance behind the detonation front during passage of the detonation enabled the determination of the width of the channel occupied by the reacting and expanding explosive and of the pressure exerted by the explosive against the plate. Since the plate configuration was a steady one moving along the plate at the detonation velocity D. a measurement of the angle θ by which the surface of the plate was deflected gave a direct measure of the velocity of the plate as a function of distance ybehind the detonation front. The deflection of the plates was obtained by observing the displacement of the image of a grid reflected in the polished outside surface of one of the plates. This method has the sensitivity required to determine the pressure in the region immediately behind the detonation

front where the lateral displacement is extremely small.

The source of the image was a grid of clear lines on relatively opaque 8×10 in. photographic film mounted vertically between glass plates and illuminated by an explosive argon flash lamp. The displacement of the image was observed with a Beckman-Whitley Model 189 Framing Camera. It was most convenient to mount the source grid parallel to the mirror plate as shown in Fig. 1. In this arrangement the length of the optical lever arm was identical for all points on the initial position of the plate and was given by the perpendicular distance from the plate to the grid. The deflection angle θ of the plate surface is then related to the vertical distance d by which the image is displaced by $\tan 2\theta = d/L$, where L is the plate-to-grid perpendicular distance. However, if the plate has been displaced outward a distance x, the relation is

$$\tan 2\theta = d/(L - x). \tag{1}$$

The displacement x was obtained by numerical integration of the experimental tan θ versus y data

$$x = \int_0^y \tan \theta \, dy, \tag{2}$$

where y is the distance along the undeflected plate surface. A grid of crossed lines was used to enable separation of the vertical displacement of the image from a small horizontal displacement arising from the nonperpendicular angle of view in the horizontal plane and from any slight horizontal curvature of the plate. The scale for image displacement measurements was based on the vertical size of the undistorted grid as measured on the first frames of the sequence. Separate reference distances were provided (as shown in Fig. 2) for measurement of position on the plate. The camera was focussed to give a sharp image of the grid. The small vertical aperture of the framing camera provided a sharp vertical location of points on the plate, the lateral blurring being of no consequence. Motion blur of the displaced image was minimized by shortening the exposure time by reducing the vertical aperture to the point where diffraction began to degrade the spatial resolution. The quality of the reflecting surface of the plate often deteriorated considerably upon deformation, especially in the region slightly behind the detonation front where the acceleration was beginning to decrease from its maximum value. Figure 2 illustrates a typical frame from a framing camera sequence. In practice, a large number of readings of image displacement d versus distance y



FIG. 2. Typical frame from framing camera sequence. The detonation front (D) is moving from top to bottom. The black rectangles on either side of the plate provide secondary distance references. Point A on undeflected image is equivalent to Point A' on the deflected image. The displaced grid lines at the base of cell are lines reflected from a small mirror in front of the assembly, used to provide a fixed frame of reference for measurement if the detonation runs to the end.

behind the detonation front were taken from several frames (after a steady state configuration has been reached) in order to minimize the effects of local fluctuations in the explosive. A typical set of experimental d versus y values is shown in Fig. 3. Equations (1) and (2) were then used to obtain θ and x as functions of y.

The configuration of the explosive-metal interface and the pressure along it were determined by neglecting the thickness of the plate. This assumption, while intuitively reasonable, is difficult to justify in a quantitative way. It would be possible to treat the plate as either an incompressible or compressible fluid in steady flow and compute both the configuration of and the pressure along the explosive-metal interface. However, it is not clear how to take account of the elastic strength of the plate in a detailed



FIG. 3. Example of typical experimental data of image displacement versus distance behind the detonation front.



FIG. 4. Frame illustrating elastic bending wave pattern set up in stainless-steel plate when the detonation velocity is less than about 2.0 mm/ μ sec.

way. It was therefore chosen to use the zero thickness approximation rather than to make an elaborate partial correction.

The energy dissipated in plastic deformation may be estimated roughly by computing the energy required to bend the plate to maximum curvature and to straighten it. In typical experiments, this estimate is only 1 or 2 cal/g of explosive, which is less than 1% of the observed kinetic energy of the plates per gram of explosive. When the detonation velocity D exceeds the shear wave velocity of the plate metal, the only elastic disturbance in the plate that travels ahead of the detonation and is likely to be excited is a plate compressional wave. This symmetrical wave causes no net lateral displacement of the plate, but only an elastic thickening of the plate. The motion is too small to be observed by measurement of the surface angle. When the detonation velocity is less than the shear wave velocity, elastic bending waves can travel ahead of the detonation. We have observed these (Fig. 4) in stainless-steel plates only when the detonation velocity is less than 2.0 mm/ μ sec, compared with a shear wave velocity of about 3.1 mm/ μ sec. The only other effect of the elastic strength would be to contribute to a slight smearing of the measured pressure-distance profile in the region of the detonation front, where the rate of change of curvature is the highest. In this region the applied pressures are much larger than the elastic strength of the metal used for confinement. The assumption was thus made that the elastic properties of the walls had a negligible effect on the measured trajectory of the plates.

Since the plate does not elongate, the velocity vector of the plate is at an angle $\frac{1}{2}\theta$ with respect to the perpendicular to the initial plane of the plate and has a magnitude V_p given by

$$V_{\rm p} = 2D\sin\left(\frac{1}{2}\theta\right),\tag{3}$$

where D is the detonation velocity. V_p was thus determined as a function of distance y behind the detonation front from the detonation velocity and the curve of θ as a function of y obtained as described earlier. The pressure on the plate as a function of distance behind the front was then obtained from

$$P \cos \frac{1}{2}\theta = m \, dV_{\rm p}/dt = mD \, dV_{\rm p}/dS, \qquad (4)$$

where m is the mass of the plate per unit area and S is the distance along the deflected plate surface,

$$S = \int_0^y (1 + \tan^2 \theta)^{\frac{3}{2}} \, dy.$$

The quantity S is also derived from the tan θ versus y curve. In the present experiments $\cos \frac{1}{2}\theta$ in Eq. (4) was essentially unity and was used as such in the analysis.

Analysis of Data

We obtain the dynamical behavior of the explosive in the detonation process from the pressure P, the lateral displacement x, and the detonation velocity D, by assuming that the flow of the explosive through the detonation can be analyzed as a pseudo onedimensional channel flow (Fig. 5). In a coordinate system in which the detonation is stationary, the mass flow equation is

$$A\rho q = A_0 \rho_0 D = A_0 \rho_1 q_1, \qquad (5)$$

where A is the cross-sectional area of the channel, ρ is the density, and q is the flow velocity. The subscript 0 refers to the explosive ahead of the detonation and the subscript 1 to the plane immediately behind the detonation front, where the cross-sectional area has not yet been increased by motion of the confining plates.

The equation of motion for one-dimensional channel flow is

$$-(1/\rho) dP/dS = q dq/dS.$$
(6)

For simplicity we set $P_0 = 0$ since $P_0 \ll P$. From





the equations describing the shock transition from state 0 to 1;

$$P_1 = \rho_0 D(D - q_1) = \rho_0 D^2 - \rho_0 D q_1, \quad (7)$$

$$q_1 = (\rho_0 D^2 - P_1) / \rho_0 D.$$
(8)

From Eqs. (6), (5), and (8),

$$dq/q_{1} = -(A/A_{0}\rho_{0}D)(dP/q_{1}),$$

$$\frac{q}{q_{1}} = -\left[\frac{1}{\rho_{0}D^{2} - P_{1}}\right]\int_{P_{1}}^{P}\frac{A}{A_{0}}dP + 1.$$

Integrating by parts

$$\frac{q}{q_{1}} = -\left[\frac{1}{\rho_{0}D^{2} - P_{1}}\right] \left\{ \left[\frac{A}{A_{0}}P\right]_{P_{1}}^{P} - \int_{A_{0}}^{A}\frac{P}{A_{0}}dA \right\} + 1.$$
(9)

Noting that

$$\int P \, dA = m(V_{\rm p})^2, \qquad (10)$$

when energy dissipation in the plates is neglected (m is the mass per unit area of each plate), and taking

$$A/A_0 = 1 + (2x/A_0), \tag{11}$$

approximations which are valid for small values of the angle of deflection of the walls, θ , we obtain

$$q/q_1 = [1/(\rho_0 D^2 - P_1)]$$

:[P_1 - P(1 + 2x/A_2) + mV_2^2/A_2] + 1. (12)

From Eq. (5):

$$1/\rho = V = Aq/A_0\rho_0 D = (q_1/\rho_0 D)(A/A_0)(q/q_1)$$
$$= [(\rho_0 D^2 - P_1)/(\rho_0 D)^2](1 + 2x/A_0)(q/q_1). (13)$$

The time t required for material to move from the detonation front (S = 0) to a distance S behind the detonation is

$$t = \int_0^s (1/q) \, dS. \tag{14}$$

Another quantity, $(dP/d\rho)^{\frac{1}{2}}$, obtainable from the analysis is of particular interest because it is influenced in opposite ways by expansion and by the energy release from chemical reaction. Because of this it serves as a useful measure of the reaction zone of the detonation. The rather complex effects of area increase and chemical reaction may be obtained from Eq. (5) in differential form and from Eq. (6);

$$\frac{q}{\rho}\frac{d\rho}{dq} = \frac{q^2}{dP/d\rho} , \qquad (15)$$

$$\frac{q}{A}\frac{dA}{dq} = \frac{q^2}{dP/d\rho} - 1.$$
(16)

First, consider the special case when the reaction rate is zero and only lateral expansion is occurring. Then $dP/d\rho = c^2$, so that

$$(q/\rho)(d\rho/dq) = -q^2/c^2,$$
 (17)

$$(q/A)(dA/dq) = (q^2/c^2) - 1.$$
 (18)

For subsonic flow, an increase in area is associated with a decrease in flow velocity and an increase in pressure and density; for supersonic flow, the reverse is true.

The effect of an exothermic chemical reaction can be obtained by assuming first that the pressure Pis held constant. In this case the velocity q is also constant. Therefore, the area A must be increased to accommodate the increased volume produced by the chemical reaction. If the area A is then adjusted back to its original value with no further chemical reaction, then from Eqs. (17) and (18) the flow velocity increases and the density and pressure decrease in subsonic flow; again the reverse is true in supersonic flow. Chemical reaction and lateral expansion thus have opposite effects on the flow variables; furthermore, the effects of both reaction and expansion are reversed as the flow passes through the sonic or Chapman-Jouguet surface.

Immediately behind the detonation front before appreciable acceleration of the confining plates has occurred,

$$dA/dS = 0, \qquad dP/d\rho = q^2. \tag{19}$$

The behavior of $(dP/d\rho)^{\frac{1}{2}}$ through the detonation is indicated schematically in Fig. 6. We first consider the idealized case (a) of a reaction zone with no lateral expansion until reaction is completed, followed by a zone of lateral expansion with no further reaction occuring. In the reaction zone the flow is subsonic; q rises to meet the (usually) falling ccurve at the Chapman-Jouguet state where reaction is complete. In this zone $(dP/d\rho)^{\frac{1}{2}}$ coincides with the q curve [Eq. (19)]. In the expansion zone the flow is supersonic, q continues to increase, and P, ρ , and c decrease. In this zone $(dP/d\rho)^{\frac{1}{2}}$ coincides with c. Thus, $(dP/d\rho)^{\frac{1}{2}}$ reaches its maximum value at the Chapman-Jouguet state where reaction is complete. Considering now the actual case in which reaction and expansion are occurring simultaneously, we note in Fig. 6(b) that the q and c curves again cross at the Chapman-Jouguet state or sonic point, where reaction is now only partial. The quantity $(dP/d\rho)^{\frac{1}{2}}$ initially coincides with q immediately behind the front, but later falls below the q curve because of lateral expansion. Eventually, as reaction



FIG. 6. Schematic variations of sound speed (c), flow rate (q), and $(dP/d\rho)^{\frac{1}{2}}$ with distance behind the detonation front for two detonation cases: (a) case of reaction zone with no lateral expansion until completion of reaction; and (b) case of overlap of reaction and expansion regions.

proceeds to completion downstream from the Chapman-Jouguet state, $(dP/d\rho)^{\frac{1}{2}}$ approaches the falling *c* curve. Thus, $(dP/d\rho)^{\frac{1}{2}}$ goes through a maximum which will generally lie near the sonic point. The exact location of the sonic point cannot be obtained from the experimental data since the sound velocity cannot be determined (unless the equation of state of the mixture is known). From the experimental approach, the maximum in $(dP/d\rho)^{\frac{1}{2}}$ appears to be a reasonable measure of the length of the effective reaction zone.

One would expect that usually the pressure will fall monotonically behind the detonation front, since lateral expansion gradually increases from an initial zero value; whereas, reaction is usually high behind the front. In this case, reaction dominates lateral expansion in the subsonic zone, while the reverse is true in the supersonic zone. However, the reaction rate may be very low for an appreciable distance behind the front (in an "induction" zone), in which case the pressure may rise in the subsonic zone because of the dominance of lateral expansion over reaction. Eventually the reaction rate must speed up in order that the sonic velocity may be passed.

Effects of Explosive Thickness and Confinement

The detonation process can be influenced on the one hand by changing some property of the explosive

to vary the reaction rate, and on the other hand by changing the thickness of the explosive and the degree of confinement, which affect the rate of lateral expansion. In the experiments described in this paper, the reaction rate of the explosive was varied by changing the particle size of one of the ingredients (AN) while keeping the composition (80% AN and 20% TNT) and the density fixed. If the thicknesses of the explosive and of the confining plates are fixed, the relative rates of chemical reaction and lateral expansion will be changed as the particle size is changed. The extent of chemical reaction occurring ahead of the Chapman-Jouguet point will be changed, with a consequent change in the detonation velocity. The altered conditions of pressure and temperature as a consequence of the change in detonation velocity will influence the rate of reaction. Thus, a change in particle size produces a complex effect on the measured reaction time. The direct effect of particle size on reaction rate can be obtained if the thickness of the explosive layer and the degree of confinement can be modified in such a way that the relative rates of reaction and lateral expansion remain unchanged. If the change in particle size changes the time scale of the reaction without changing the form of the reaction rate curve then a change in the thicknesses of explosive and confining plates by the same scale factor by which the time scale of the reaction has been changed will not change the relation between reaction and lateral expansion. Consequently, the detonation velocity will be unchanged. The ratio of measured reaction times should equal the scale factor.

If, on the other hand, the change in particle size changes the form of the reaction rate then a change of the thicknesses of both explosive and confining plates by the same factor will not maintain an unchanged ratio of reaction rate to lateral expansion throughout the reaction zone. For a given thickness ratio, the ratio of particle sizes which keeps the detonation velocity unchanged will differ from the particle size ratio which gives a ratio of reaction times equal to the thickness ratio. In discussing the present experimental results, we designate as "scale shots" those pairs of shots made with different particle sizes and different thickness for which the detonation velocity is substantially unchanged. In every case, the thickness of the confining plates was changed by the same factor as the thickness of the explosive.

In indirect methods of deriving the reaction time from measurements of the effects of explosive diameter on the detonation velocity, for example, by using the Eyring curved front theory, the reaction zone length is usually assumed to be unaffected by the change in detonation velocity. Since the method provides no information on the reaction zone length for a single value of the diameter, some such arbitrary assumption is necessary.

Explosives Studied

The explosive selected for systematic study by the method described was a mixture of 80% AN and 20% TNT. A two-component mixture was chosen because it was felt the effective reaction time in this type of material could be adjusted to fall in a convenient range for experimental observation. This expectation was based on the fact that over half the total energy release in 80/20 AN/TNT is derived from the mixing and interaction of the products of the decomposition of the separate components.⁸ If the particle size of one component is much larger than that of the other, the mixing process will be dependent on the particle size of the component with the larger size. In the work reported here, the particle size of the major component AN was varied over a range of diameters (0.006-0.12 cm) which was very large compared to the constant size of the TNT particles $(2-5\mu)$. In these experiments, it is likely that the rate of decomposition of the TNT was so fast that it could not be resolved by the experimental technique. The measured values of pressure and density just behind the detonation front thus probably do not correspond to compressed unreacted explosive but rather to a compressed

TABLE I. Ammonium nitrate particle sizes used in this study. (TNT particle size was approximately $2-5\mu$ in all cases.)

Mix	Ammonium Nitrate Particle Diameter (cm)	$\begin{array}{c} \text{Detonation Velocity} \\ (\text{mm}/\mu\text{sec}) \end{array}$
0 A B C D E F G	$\begin{array}{c} 0.0053 - 0.0061 \\ 0.0124 - 0.0149 \\ 0.0297 - 0.0350 \\ 0.0419 - 0.0500 \\ 0.0589 - 0.0711 \\ 0.0711 - 0.0833 \\ 0.0833 - 0.1001 \\ 0.1001 - 0.1191 \end{array}$	$\begin{array}{r} 4.43 \\ 4.15 \\ 3.98 \\ 3.82 \\ 3.67 \\ 3.38 \\ 2.83 \\ 2.67 \end{array}$

mixture in which much of the TNT has already reacted.

The TNT was prepared by a proprietary process from commercial material. The AN was prepared from commercial material by drying and double screening. The cells were loaded by vibrator packing the measured weight of material into the known cell volume. The mix designations are listed in Table I.

Experimental Results

The results [pressure P, density ρ , flow velocity q, and $(dP/d\rho)^{\frac{1}{2}}$] obtained by the grid reflection technique and the one-dimensional channel flow analysis on two of the eight mixes for $\frac{1}{2}$ -in.-thick layers of explosive confined by $\frac{1}{4}$ in. stainless-steel plates are shown in Fig. 7. For comparison of the different mixes, the curves of P versus S are shown in Fig. 8,



FIG. 7. Pressure, density, flow rate, and the derived quantity $(dp/d\rho)^{\frac{1}{2}}$ versus distance behind the detonation front for two of the mixes studied (see Table I).

⁸ Reference 7, p. 307.



FIG. 8. Summary of pressure versus distance for all experiments in the large cells. Mix designations are indicated for each case.

and the curves of P versus specific volume V are shown in Fig. 9. Values of the effective reaction time t obtained by integrating Eq. 14 from the detonation front to the position of maximum $(dP/d\rho)^{\frac{1}{2}}$ are shown versus particle diameter d, of the AN in Fig. 10. t was found to vary with both particle diameter and "cell size" (explosive thickness and confining plate thickness changed by the same factor, 0.444). The heavy curve drawn through the horizontal bars (indicating the AN particle size range) shows a complex relation of effective reaction time to AN particle size for a fixed cell size. The circles



FIG. 9. Pressure versus specific volume for all experiments in the large cells. Rayleigh lines connect the state just behind the front to the initial (uncompressed) volume.



FIG. 10. Reaction time versus ammonium nitrate particle diameter. The horizontal bar lines represent the particle size ranges of the experiments in the large cells $(\frac{1}{2}\text{-in.-wide explo$ sive layer) and the corresponding reaction times. The circledpoints indicate the reaction times for the scale shots at theaverage value of particle size. Straight lines have been usedto connect the origin to the points on the scale shots andfrom the scaled points to the points on the large cell curvewhere the reaction time is in scale (1/0.444) with that of thesmaller cells. The dashed line represents the probable shapeof the reaction time curve for the smaller cells on the assumption that the observed scaling holds over the entire range ofparticle diameters.

indicate shots made with the smaller cell size, and the tie lines through two of these points connect them to points on the curve for the larger cell size where the reaction time is increased over the scale shot by the same factor as the cell size. It was found that the detonation velocities for the scale shots were within experimental error of the interpolated detonation velocities of the large shots (Tables I and II). As noted earlier, this indicates that only the time scale and not the form of the reaction varies in the scaled shots.

The amount of reaction occurring in the effective reaction zone between the detonation front and the peak of $(dP/d\rho)^{\frac{1}{2}}$ apparently decreases as the particle size of the AN increases. Some idea of the extent of reaction would be very helpful in inter-

TABLE II. Summary of results of scale shots.

Shot Designation	Detonation Velocity $(mm/\mu sec)$	"Full-Scale" Detonation Velocity ^a (mm/µsec)
A'	4.15	4.13
F'	3.17	3.12
H'	2.48	
Ι'	1.97	

 Interpolated detonation velocity of full size shot at that particle size for which the reaction time was increased by the scale factor 1/0.444.

preting the change in reaction time with particle size. Accordingly, an attempt was made to estimate the amount of reaction occurring for each amatol mix in the following way. It was assumed that complete reaction had occurred by the time at which V = 1.2 cc/g for the smallest particle size. [This volume was much larger than that at which the peak in $\left(\frac{dP}{d\rho}\right)^{\frac{1}{2}}$ occurred for any of the particle sizes.] The area under the PV curve for Mix 0 from V = 1.2 cc/g to zero pressure was obtained as the difference between the total energy release and the measured area from the shock to V = 1.2 cc/g, less the shock energy $\frac{1}{2}P_1(V_0 - V_1)$. It was then assumed that the isentropic expansion from V = 1.2cc/g followed a constant γ law, for which E = $PV/(\gamma - 1)$ then determined the effective value of γ . The value of γ thus determined for the finest AN particle size was then used to determine the area under the "tails" of all the other PV curves from V = 1.2 cc/g to zero pressure. It felt is that this calculation (see Fig. 11) afforded a reasonable, if crude, estimate of the energy release up to V = 1.2cc/g.

Radiographic Experiments

In addition to the optical work, flash radiography was employed, for a twofold purpose, (1) to examine the major assumption made in the work, that of negligible variation of properties on cross-channel lines, and (2) to obtain a crude check on the density profiles obtained in the optical work. The explosive was viewed parallel to the confining plates and was loaded to the same density as in the optical work.

In adapting the one-dimensional channel flow arrangement to allow radiography, the width of the channel was reduced from 8 to 4 in., and the lateral edges were covered with 0.120-in.-thick glass rather



FIG. 11. Energy versus particle diameter of ammonium nitrate. Illustrated are the curves obtained from the experimental P-V data. The total energies to be expected from the various reaction steps are indicated.



FIG. 12. Radiographs of detonation in one-dimensional channel flow for mixes G (left) and C (right). The aluminum "step wedges" are visible in the lower parts of the radiographs. The detonation is proceeding in both cases from top to bottom. The radiographs illustrate the relatively small expansion of the stream tube with this confinement.

than massive steel. The cells were again formed by parallel $\frac{1}{4}$ -in.-thick plates of 304 stainless steel. The method of radiography and film protection used has been described previously.⁹

The density of the explosive behind the detonation front was obtained by measuring the optical density of the radiograph with a Jarrel-Ash Model 21-000 Microphotometer at a slit size of 1.5 mm \times 15 μ and comparing the readings for the explosive with readings from an aluminum step wedge in the field of view. The aluminum step wedge had been calibrated against a step wedge of the same explosive at the same density with the same x-ray flash voltage (500 kV) used in the detonation shot. Two of the radiographs are shown in Fig. 12. Densitometer readings across the 0.5-in.-wide channel at any fixed distance from the detonation front indicated no detectable density variations on cross-channel lines.

Curvature of the detonation front made it impossible to obtain from the radiograph an unambiguous, density profile downstream from the detonation front. Whereas, in the optical work measurements were made near the center line of the plates where curvature is minimal, the x-ray path extended through the full width of the explosive, including

⁹ A. S. Balchan, J. Appl. Phys. 34, 241 (1963).



FIG. 13. Comparison of radiographic and "smeared" optical density profiles for Mix C.

the edge zones where the curvature is greatest. The reduced width and edge confinement required in the radiographic work further enhanced the curvature effects. A comparison of optical and radiographic results was obtained by computing an apparent density profile in the narrow cells from the optically obtained density profile and the curvature of the front. The curvature in the smaller cells was measured by a modified grid reflection technique with explosive shots identical to the x-ray shots. It was assumed that the optically obtained density profile did not vary along the x-ray path, the whole profile being displaced downstream in accord with the curvature of the front. The apparent density profiles obtained by averaging along the x-ray path were compared with the radiographically observed profiles for four different explosive mixtures. For brevity, one typical comparison is shown in Fig. 13; agreement in the other three experiments was as good as that illustrated.

It was concluded that the radiographic experiments provided adequate justification for the onedimensional flow assumption and indicated that no gross errors in measurements of flow properties were made in the optical work.

DISCUSSION OF RESULTS

The most obvious feature of the present results is the occurrence of a relatively thin high-pressure zone immediately behind the shock front in all but the coarsest amatol mixes; the zone is most clearly developed where the AN particle size is smallest. The dominance of chemical reaction in the finegrained mixtures is clearly demonstrated in the P-Vcurves (Fig. 9), where the pressure drops to about half the peak value along a line which barely deviates from the Rayleigh line. As shown in Fig. 6, in the Zeldovich-von Neumann-Doering model the quantity $(dP/d\rho)^{\frac{1}{2}}$ rises along the velocity (q) curve until it reaches its maximum value at the Chapman-Jouguet point, where the velocity is sonic, and where, in the Zeldovich-von Neumann-Doering model, reaction is complete. In the more fine-grained mixtures, $(dP/d\rho)^{\frac{1}{2}}$ follows the q curve almost to its maximum value and then falls sharply as q continues to rise because of lateral expansion. The location of the maximum $(dP/d\rho)^{\frac{1}{2}}$ must therefore lie close to the Chapman-Jouguet point in the fine-grained explosives.

As the AN particle size is increased, the pressure profile in the reaction zone becomes less steep; and for the coarsest size studied (Mix G), the pressure is almost flat. Apparently in the coarse mixtures the rate of energy release behind the front is so low that it barely predominates over lateral expansion. Since the rate of lateral expansion rises steadily from an initial zero value, the rate of chemical energy release must be rather low near the detonation front and probably increases downstream, at a rate which is necessarily slower than the rate of increase of the rate of lateral expansion. since the two effects must be equal at the Chapman-Jouguet point. As pointed out earlier, the pressure can show a rise in the initial part of the reaction zone, but this was not observed in the range of particle sizes studied.

Since the decomposition of the very fine TNT component should be fast, even in the mixtures with coarse AN, it appears likely that the TNT reaction is occurring too fast to be resolved adequately. In the coarse AN mixtures it is quite possible that the TNT reaction is occurring partly in the shock front zone, which must extend over a few particle diameters, i.e., a few millimeters.

The complex variation of effective reaction time τ versus AN particle diameter for fixed explosive thickness $(\frac{1}{2}$ in.) and confinement $(\frac{1}{4}$ -in. stainless-steel plates), shown by the heavy curve in Fig. 10, can be accounted for qualitatively on the basis of the three-stage chemical energy release and the effects of greater lateral expansion relative to the extent of reaction and lower detonation velocity as the particle diameter increases. We use as a guide the curve of Fig. 11 which shows the rough estimate of chemical energy release up to the point where the volume was 1.2 cc/g, and we note that the energy release in the effective reaction zone is less than this curve. In the range of small particle

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diameter (Mixes 0 and A) reaction is nearly complete at $(dP/d\rho)^{\frac{1}{2}}$ maximum. The steep (somewhat less than proportional) variation of τ with d results from the decrease in the rate of the mixing reaction of the products of decomposition of TNT and AN. Further increase in diameter, from A through B; C, D, and up to E, produces only a slow increase in γ . However, in this range the extent of reaction [up to the maximum $(dP/d\rho)^{\frac{1}{2}}$ decreases considerably. Thus the reaction rate must be decreasing more rapidly with increasing particle size than would be indicated by the increase in reaction time alone. For still larger AN sizes, from E to F and G, the reaction time τ increases strongly with increasing particle size, at a rate which is more than proportional. The estimated energy release indicates that very little of the mixing reaction is occurring in the reaction zone in Mixes F and G. Presumably the decomposition of AN particles, in an atmosphere of the hot products of decomposition of TNT, is the dominant reaction in the neighborhood of $(dP/d\rho)^{\frac{1}{2}}$ maximum. The strong dependence of τ on grain diameter is indicative of a strong dependence of the rate of AN decomposition on grain size.

As discussed above, information on the direct effect of AN particle size on the reaction rate can be obtained by making scale shots. Four such shots, designated A', F', H', and I', were made in which the thickness of the explosive layer and the thickness of the confining plates were reduced by a factor of 0.444. (A' and F' were approximate scale shots of mixes A and F; H' and I' had no corresponding full-size shots.) If the change in AN size produced only a change of time scale of the reaction then at that particle size ratio that changes the reaction time τ by the scale factor the detonation velocity should be unchanged. Shots A and A' gave indentical detonation velocities; and with a particle size ratio of 0.42, compared with the cell size ratio of 0.444, the ratio of reaction times was the same within experimental error. Since reaction is nearly complete, this scaled pair of shots indicates that the reaction time of the "mixing reaction" of the products of decomposition of AN and TNT depends about proportionally on the average diameter of the AN particles under identical conditions of reaction. This finding is in agreement with the results of Eyring, obtained from variation of detonation velocity with charge diameter, on an amatol of somewhat different composition.

Boyer and Grandey¹⁰ calculated the initial tran-

sient for a one-dimensional detonation of an explosive consisting of fuel-rich and oxidizer-rich components, using reasonable values of reaction rate constants. They treated the mixing as a laminar diffusion flame. Such a model gives a very strong dependence of reaction time on particle size (r proportional to the square of the particle diameter d). With their reasonable values for the physical constants involved, their calculations indicate that the mixing reaction will contribute substantially to the detonation when the particle diameter is not more than about 10^{-5} cm. Both the magnitude and the linear dependence of r on d found in the amatol experiments indicate that the mixing cannot be occurring by laminar diffusion. Turbulence would tend to make the (eddy) diffusivity proportional to particle size, thus increasing the rate of mixing at larger particle sizes and tending to make the reaction time proportional to d rather than d^2 . Turbulent mixing could occur as a result of an appreciable difference in flow velocity between the AN particles and the TNT decomposition products. If the two velocities are the same immediately behind the front then a difference could be set up as reaction releases energy and material into the gas stream. The solid AN particles would tend to lag behind by an amount dependent on the drag of the accelerating gases. By assuming each particle was accelerated by a constant effective drag pressure obtained by using a drag coefficient for spheres of about unity (corresponding to turbulent flow), it was estimated that a difference of velocities of several hundred meters per second could be generated, which is sufficient to give a Reynold's number well into the turbulent range in granular beds even for values of the viscosity coefficient as high as 10^{-2} poises.

In comparing the scale shot designated F' with the $\frac{1}{2}$ in. full-size shots, the AN size ratio which makes the ratio of reaction times equal to the cell size scale (0.444) is about 0.54, corresponding to a full-scale shot lying between E and F at a particle diameter of 0.085 cm (Fig. 10). The detonation velocities (3.17 mm/ μ sec for F' compared with an interpolated large cell value of about 3.10-3.14 mm/μ sec) agree within experimental error. From the estimated amount of energy release in the reaction zone, it is probable that the main energyreleasing reaction at $(dP/d\rho^{\frac{1}{2}})$ maximum is the decomposition of AN. The reaction time for this reaction under identical conditions of reaction is proportional to $d^{4/3}$. A constant linear rate of grain burning would be expected to make τ proportional

¹⁰ M. H. Boyer and R. Grandey, *Detonation and Two-Phase Flow* (Academic Press Inc., New York, 1962), p. 75.

to d. The difference, if significant, may be associated with initiation and forcing of the decomposition of the outer layers of the AN particles.

Indirect methods based on the variation of detonation velocity with charge diameter have not been capable of developing information on the rates of any but the final reaction step when several reaction stages are involved in a detonation. The present direct method can give information on the rates of earlier reaction stages, provided the corresponding energy release is sufficient to maintain the detonation. The small-scale shot I' detonated at a steady velocity of 1.977 mm/ μ sec and indicated approximately an effective energy release corresponding to that of the TNT alone. The fact that the reaction time curve for the scale shots in the region of large AN particle sizes appears to be flattening indicates that we are approaching the particle size range in which the TNT alone is reacting.

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