Similarity and differences between conditions for initiation and failure of detonation

(reaction zone length/critical energy)

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ABSTRACT A theory is proposed to study the sensitivity of cylindrical explosives. Using the equations governing the conservation of mass, energy, and momentum, the equation of state, and Newtonian mechanics, we have derived the expression $D = D_0 e^{-a_1/d}$, in which the apparent length of the reaction zone, a , takes into account burned and unburned explosive and pressure release through the back and sides of the reaction zone. D is the detonation velocity and d is the diameter of the cylindrical charge. From the observed $D(1/d)$ curves and critical energy values, we can interpret failure and other phenomena associated with detonation.

A fruitful source of experimental data has been the measurement of the dependence, in the steady state, of detonation velocity in a cylindrical explosive on the radius of the explosive charge. In Fig. 1 (ref. 1, p. 42) we see detonation velocities (D) plotted semilogarithmically against the reciprocal of the diameters (d) of cylindrical charges for four typical explosives. Fig. 1 suggests the need of a theory to explain the diverse effects seen in this type of data. In an early paper (2) it was shown that for a one-dimensional detonation, using the familiar conservation of mass, energy, and momentum equations and the Abel equation of state, $P(V - \alpha) = RT$, one obtains

$$D^2 = (\Delta Q + \bar{C}_v T_0) \frac{2(\gamma^2 - 1)}{(1 + \alpha/V_0)^2} \quad [1]$$

and

$$P_1 = \frac{2R}{(V_0 - \alpha)} \frac{(\Delta Q + \bar{C}_v T_0)}{\bar{C}_v} \quad [2]$$

These are equations 6d and 6b of ref. 2; P_1 is the pressure at the Chapman-Jouguet surface and D is the detonation velocity. The symbols ΔQ , $\bar{C}_v$, T_0 , γ , α , V_0 , and R are the heat of reaction, the mean specific heat at constant volume, the ambient temperature, the ratio of specific heats at constant pressure and constant volume, the excluded volume in the Abel equation, the initial volume of the explosive, and the gas constant, respectively. From Eqs. 1 and 2 we see that D and P are closely related. In this report a theory is proposed to give the quantitative relationship between the detonation velocity and the charge diameter, from which we explain what happens in a steady-state detonation and at the failure point.

THEORY

In accord with Newtonian mechanics, the forward impulse pushing against an unbound explosive must be balanced by an oppositely directed impulse excited by escaping gases at the back of the reaction zone. It follows that the pressure driving the explosive is only half what it would be if escape of the

FIG. 1. $D(1/d)$ curves for trimethylene trinitramine (RDX) and RDX/2,4,6-trinitrotoluene (TNT) mixtures (from ref. 1, p. 42). Δ , 65 RDX/35 TNT, 1.71 g/cm³; $\circ$, RDX, 0.9 g/cm³; $\blacktriangle$, 60 RDX/40 TNT, 0.9 g/cm³; $\bullet$, 60 RDX/40 TNT, 0.5 g/cm³; all ratios are wt/wt.

rarefaction wave out the back were prevented. In Fig. 2 we indicate by $2P_0$ the hypothetical pressure that would exist if the pressure lost, P_0 , were added to the observed pressure P for a cylinder of infinite radius. The initial pressure, $2P_0$, will decrease for a cylinder of finite diameter d because of the increased escape area normal to the cylindrical axis and because of the length of the reaction zone (see Fig. 3) and the resultant incomplete reaction of the explosive. The reaction zone length, a , is the distance from the front of the shock wave to the Chapman-Jouguet surface, where the rarefaction wave begins.

We treat the burning zone of the explosive in Fig. 3 as cylindrical only as a matter of convenience, because we correct the calculated escape pressure by multiplying the normal surface area, $a\pi d$, by β_1 , which converts it to the effective escape area on a par with the area $\pi d^2/4$ of the base of the cylinder. When the length of the reaction zone, a , is short, β_1 will be close to unity.

Fig. 2 shows the effect of the escape area on the pressure as a function of $1/d$. Accordingly we write

$$\frac{2P_0 - P_0}{\pi d^2/4} = \frac{2P_0 - P}{\pi d^2/4 + \beta_1 a \pi d} \quad [3]$$

Solving for P , one obtains

$$P = P_0 \left(1 - \frac{\beta_1 4a}{d} \right) = P_0 \left(1 - \frac{2a_1}{d} \right), \quad [4]$$

in which $a_1 = 2\beta_1 a$.

As long as $2a_1/d \ll 1$ this is equally well written as

$$P/P_0 = e^{-2a_1/d} \quad [5]$$

The latter expression is satisfactory until the losses perpendicular

Abbreviation: TNT, 2,4,6-trinitrotoluene.

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FIG. 2. Pressure versus reciprocal of cylinder diameter.

to the cylinder axis cause a critical change in the slope of the line D versus $1/d$, as will be explained later, when the line gets a new slope a_2 . Returning to Eqs. 1 and 2, we note that simply changing the diameter of our cylindrical explosive leaves all the quantities in these equations unchanged. It therefore follows that we can replace P/P_0 by D^2/D_0^2 . We therefore write

$$D^2/D_0^2 = e^{-2a_1/d}. \quad [6]$$

Now when increasing $1/d$ decreases D sufficiently to cause a sudden change in the slope of the line D versus $1/d$, we will indicate this by rewriting Eq. 6 as

$$(D/D_0)^{\beta_2} = e^{-a_1/d} \quad [7]$$

or

$$\ln(D/D_0) = -\frac{a_1}{\beta_2 d} \equiv -\frac{a_2}{d}. \quad [8]$$

Here $a_2 = a_1/\beta_2 = 2(\beta_1/\beta_2)a$.

We next consider how β_2 can be sometimes greater and sometimes less than unity, as is in fact observed. As long as $\ln(D/D_0) = -a_1/d$ is small and a_1 keeps a constant value, this equation is understandable as arising from the simple escape theory, which leads directly to the equation $D/D_0 = 1 - a_1/d$. When the curve suddenly shifts to a new straight line at some value of $1/d$ with a greater slope corresponding to $\beta_2 < 1$ or a lesser slope corresponding to $\beta_2 > 1$, this can be understood as follows. As D grows smaller it may still be large enough to excite some sites that now have time, with a smaller D , to develop into burning centers and make a contribution to the burning rate which was not previously possible. In this case, in which $\beta_2 > 1$, the apparent reaction zone length grows more slowly than before with an increase in $1/d$, and we have $a_2 < a_1$. On the other hand, if a certain part of the burning centers suddenly phase out at a certain $1/d$ the slope will then grow more steeply, and we have $a_2 > a_1$. The abruptness of the shift from a slope a_1 to a_2 at a particular value of $1/d$ indicates an abrupt change in the surface area burning. Until we can determine the values of β_1 and β_2 separately we will write $2\beta_1/\beta_2$ as a single term β .

We justify the success of the exponential form of Eq. 7 above by expecting phonon and photon behavior to be analogous. Thus, burning centers consist of a variety of structures excited by absorbing energy from the pressure wave as it passes over them just as a pulse of light excites molecules as it gives up energy to excite the molecules. There is a third change in the

FIG. 3. Reaction zone in the detonation of a cylindrical charge.

pressure with $1/d$ due to holes flowing into the explosives, changing the density. This effect may be evaluated by studying change in density. A fourth effect arises from a change in the number of burning centers ignited. A fifth effect arises from the burning surface area that can develop before being snuffed out by the rarefaction waves. Effects four and five will be reflected in the amount of unburned material. For the radius r of the explosive, when the transient has settled down to the steady state, we can write for the pressure dependence

$$\frac{dP^n}{d(1/r)} = \kappa P^n, \quad [9]$$

in which κ is analogous to Beer's coefficient for photons. Thus

$$P^n = P_0^n e^{-\kappa/\tau} = P^n e^{-\kappa'/d}. \quad [10]$$

Because of the proportionality of P to D^2 as long as only d is changed, we can write

$$\ln(D/D_0) = -\frac{\kappa'}{2n} (1/d), \quad [11]$$

which is essentially the same as Eq. 8. In all cases treated it is interesting to find that a plot of experimental data using Eq. 8 or Eq. 11 and starting at $1/d = 0$ gives two straight lines. At the intersection of the first line with the second line there must be a change in mechanism. The shock wave velocity, D , at this value of $1/d$, has dropped to a value such that the rarefaction wave snuffs out certain centers, leaving unburned explosive. The final line terminates when the steady state velocity, D , has dropped to a value that can no longer support a detonation of any kind. The slope of the final line is sometimes less and sometimes more than that of the first line, as one sees from the figures given and many others not reproduced here.

DISCUSSION

Returning to Fig. 1, there are a number of points that should be considered.

(i) We first notice that the intercepts on the $\ln(D/D_0)$ axis of these curves of $\ln(D/D_0)$ versus $1/d$ are arranged in the order of initial density, where the change in d is the only change in the explosive. Because kinetic energy density is pressure and is therefore proportional to D^2 , this was to be expected from the theory.

(ii) When D_0 is high (see top curve of Fig. 1), the slope of the curve is slight, suggesting fast reaction—i.e., short reaction zone length. This is understandable if a plasma or some other reactive burning sites are formed as a result of the shattering blow ac-

FIG. 4. $D(1/d)$ curves for Baratol (from ref. 3, p. 130). Δ , Colloid milled BaNO_3 ; $\circ$, $-65 + 100$ mesh BaNO_3 .

FIG. 5. $D(1/d)$ curves for 50 TNT/50 NaNO_3 (from ref. 4, p. 131). Δ , Cast sodatol; $\circ$, loose sodatol.

companying the shock wave, giving many burning surfaces with low activation energies so that burning is very rapid.

In Fig. 2 we see the behavior featured in the top curve of Fig. 1. In the bottom two curves of Fig. 1, which involve mixed explosives, a rapid drop with $1/d$ is, as stated above, probably due to more widely spaced burning centers arising with smaller D s and therefore slower burning and a longer apparent reaction zone. In Fig. 4 at $1/d = 0.20$ for the upper curve and at about 0.14 for the lower curve we see the same rapid changes to less steep slopes, with failure occurring at higher values of D . A sharp increase in the curvature of the shock wave front would also cut down losses out the side by decreasing the lateral escape area, but were this the case it would leave unexplained the abrupt change in the slope.

Transition to Steeper Slopes at Lower Values of $1/d$. Transitions to steeper slopes are also observed (see Fig. 5). Incomplete burning due to initiating fewer burning centers, giving incomplete burning to products, is apparently one way for these steeper slopes to occur. Other features are suggested by the data shown in Fig. 6.

Stability of Detonations in Cylinders with Small Diameters. With loss of explosive out the side and decrease in density of the outside layers, these layers tend to lag behind the center, giving a curved front. If the outside layers did lag behind the center, the shock wave would be disrupted. There is not disruption because phonon waves bouncing off the curved front are reflected back toward the center, with a resultant equalizing of kinetic energy density of surface and center layer keeping the shock wave moving together. This slowing down of center layers by a drag of outside layers simply increases curvature and therefore mixing of layers. It is thus clear how the curvature of the shock wave front is a measure of decreasing velocity of the shock wave from its ideal flat front value for $r = \infty$.

Maxima Arising in D from a Change in the Mix of Particle Sizes in the Charge. Large particles develop greater shear from a shock and so are more apt to initiate burning centers. Typically, burning will spread over much of the internal surfaces from such centers so that the burning rate will be fastest when there are enough large particles to supply sufficient initiating

FIG. 6. $D(1/d)$ curves for $\text{Pb}(\text{NO}_3)_2/\text{TNT}$ and 2,4-dinitrotoluene (from ref. 4, pp. 132 and 129). Δ , 70/30 $\text{Pb}(\text{NO}_3)_2/\text{TNT}$, -28 + 48 mesh $\text{Pb}(\text{NO}_3)_2$; $\circ$, -65 + 100 mesh 2,4-dinitrotoluene.

centers and enough small particles to supply ample internal surface. Thus one expects the burning rate to pass through a maximum with a best mix of large and small particles, as is in fact observed. This type of analysis seems to account for the observed qualitative behavior of explosives, but only as it is made quantitative can it properly weight alternative causes of the various phenomena. This will require, among other things, an appropriate equation of state that will vary with the mechanism of burning and the products formed.

Initiation in a Steady-State Detonation and de Novo Initiation. Walker and Wasley (4) have shown that total energy fluence—i.e., the violence of a shock times the time the pressure is applied—fixes a line that separates the domain of detonation from the domain of failure to detonate. For a different type of explosive, B. Brown and others at Hercules (personal communication) find a different slope for this fluence line. The two sharply intersecting lines in our graphs of D against $1/d$ observed regularly for detonation suggest an unexpectedly abrupt change in the detonation mechanism. This is understandable if two intersecting lines arise from two different types of initiating centers with different fluence conditions.

The Length of Reaction Zone near the Failure Point. The critical energy for detonation is given by (4)

$$E_c = \frac{tP^2}{\rho_0\mu_s(10)}, \quad [12]$$

in which t is the shock pulse width in μs , P is the average shock pressure in kilobars (1 kbar = 10^8 pascal), ρ_0 is the density of the unshocked explosive in g/cm^3 , μ_s is the shock velocity in the explosive at pressure P in $\text{cm}/\mu\text{s}$, and the factor 10 is used to give the critical energy E_c in J/cm^2 .

If the detonation velocity D is given, the value of P , μ_s , and t can be estimated approximately for condensed explosives as follows:

$$P = \rho\mu_s\mu_p = \rho_0D(1/4D) = 1/4\rho_0D^2 \quad [13]$$

$$\mu_s = D - \mu_p = 3/4D \quad [14]$$

Table 1. Reaction zone length and the value of β at failure conditions

Explosive	E_c^* cal/cm ²	ρ_0^* g/cm ³	$D_i^\dagger$ cm/ μs	a , mm	βa , mm	β
TNT	≈ 100	1.6	0.6285	0.19	1.1	5.8
Composition B	≈ 150	1.715	0.69	0.23	2.94	12.8
Tetryl	42	1.66	0.6651	0.07	0.76	10.9

* From ref. 3. One cal = 4.184 J.

† From ref. 4, figures 6-4a (p. 129) and 3-11 (p. 53), and ref. 1, figures 1.5.2 and 1.5.3 (p. 42). The values for D were density corrected.

Table 2. Effect of pressure and density on the length of reaction zone and the value of β

Explosive	E_c , cal/cm ²	ρ , g/cm ³	P , kbar	D , cm/ μ s	a , mm	β
TNT	100	1.0	3.0	0.3487	1.01	1.09
			46.7	0.435	0.65	1.69
			60	0.4931	0.51	2.17
			70	0.4671	0.43	2.53
			90.7	0.5318	0.33	3.33
		1.3	120	0.6116	0.25	4.34
			130	0.5738	0.24	4.71
			156	0.6285	0.19	5.80
			180	0.6752	0.17	6.51
Composition B	150	1.6	146	0.5954	0.33	9.03
			169.2	0.6546	0.27	10.9
			190	0.6937	0.24	12.25
		1.715	170	0.6338	0.27	10.96
			201.5	0.69	0.23	12.6
			230	0.7372	0.2	14.83
Tetryl	42	1.05	40	0.3929	0.32	2.38
			57.0	0.4691	0.22	3.45
			70	0.5197	0.18	4.17
		1.30	80	0.4994	0.16	4.76
			96.7	0.5491	0.13	5.76
			110	0.5885	0.12	6.48
		1.66	160	0.6249	0.08	9.53
			180.2	0.6651	0.07	10.9
			200	0.6987	0.06	11.9

and

$$a = Dt, \quad [15]$$

in which μ_p is the particle velocity.

From $D(1/d)$ curves we can calculate the detonation velocity near the failure point, $D_{\min}$. By using Eqs. 12–15, the length of the reaction zone, a , can be calculated. Then, from the slope of $D(1/d)$ curves, we obtain the product of β ($= 2\beta_1/\beta_2$) and a and, consequently, obtain the value of β . The calculated values of a and β for three explosives—TNT, composition B, and tetryl—are given in Tables 1 and 2. Notice that:

(i) For these explosives, the one with the lowest critical energy is associated with the shortest reaction zone. This indicates that an explosive with a short reaction zone needs less energy to initiate detonation.

(ii) For the same explosive, the length of reaction zone decreases with an increase in the average shock pressure. A higher pressure sets up a stronger shock wave and creates more reaction centers. As a consequence, the reaction can be completed within a shorter length of time. An increase in the explosive density causes an increase in the length of reaction zone. However, an

explosive with higher density requires higher pressure to detonate. The latter effect usually overshadows the effect of density, and the resultant effect shows a decrease in the length of reaction zone.

(iii) The values of β for these explosives are higher than unity—i.e., $a_2 > a_1$. This suggests that incomplete burning plays a large part in the failure of detonating these explosives below the critical diameter.

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