# INDETERMINACY AND STABILITY OF THE DETONATION MODE IN A BOUNDED MEDIUM

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It was shown in [1] that in the presence of nonmonotonic heat release the detonation mode in an unbounded medium can be indeterminate. Certain properties of the number of detonation modes and their stability with respect to transition from one mode to another were also investigated there.

The lateral scattering of products of detonation affects the latter's rate in a manner qualitatively similar to that of heat losses or of an endothermic reaction. Noting this similarity and the results cited in [1], we consider the case of monotonic heat release on the assumption that lateral scattering is the only source of losses.

The effect of lateral scattering on the rate and stability of detonation, as well as the existence of a critical diameter of the charge, was first confirmed experimentally by Khariton and Rozing [2,3]. The critical diameter and its dependence on specific properties of an explosive were subsequently investigated in numerous papers (see [4,7] and other publications). Another interesting aspect of the link between the stability of detonation and the scattering of reaction products is the low detonation rate observed under certain conditions [4,5,8–12]. This phenomenon has not yet been fully explained. There is no doubt that the low detonation rate is, if not in all cases at least in the majority of them related to two or more heat-release stages taking place at substantially different rates [5,10–13]. However, the question of the character and limits of stability as well as of the low sensitivity of the detonation rate to variation in external parameters [10,11] remains unanswered. Attempts at a theoretical explanation of the low detonation rate were made by Eyring et al [14], but their results contradict experimental data [11,12] (see also §3 below, and [15]). The problem of indeterminacy of the detonation rate has been partially considered by Schall [16]. A critical review of [16] appears in [1].

In connection with all this it is interesting to investigate the total number of detonation modes and their stability during transition from one mode to another. In view of the recently revealed instability of a plane (smooth) detonation front [17], we point out that in the following we consider the stability of any detonation front which is steady-state on the average. The detonation rate, expressed in terms of the heat of reaction, is the same as that of a smooth front. It is known that "turbulent pulsations" of an uneven front alter the detonation rate only very slightly [18,19], and that such alterations are not always present [20]. Unevenness of the front also does not contradict the concept presented in [3] of a relation between the critical diameter and the width of the reaction zone [6,7]. The quantitative expression of the criterion given in [3] depends on specific properties of the explosive. In the case of high activation energy the induction time for large diameters close to the critical one increases very greatly in the direction from the charge axis toward its periphery, owing to considerable curvature of the front. Under these conditions the effective induction time can be considerably longer than for a straight shock wave [21].

The problem is formulated in an approximation to a given streamtube shape in §1, which also deals with investigation of the general properties of detonation modes in a bounded medium. These properties are illustrated in §2 for a simple model of detonation with one or two irreversible chemical reactions. Section 3 is devoted to discussion of the results and to the conclusions.

Systematic mathematical analysis of detonation with lateral scattering is extremely difficult. This explains the present lack of a rigorous theory for non-one-dimension detonation, in spite of numerous investigations of this problem. Several existing approximate theories [14,22–25] give a qualitatively correct description of increase in the detonation rate with increasing diameter of a cylindrical charge or with decreasing front curvature [26].

However, knowledge of the exact pattern of expansion of the explosive in the reaction zone is not necessary for the analysis of the number of modes and their stability. The mode and stability of detonation depend on the relationship between the rate of expansion and that of the chemical reaction. Both of these rates are defined for a given charge diameter by the shock-wave intensity and by the complete range of gas-dynamic parameters behind the compression shock. The rate of a chemical reaction is usually much more affected by the shock-wave intensity than by the effect of lateral scattering. Consequently, in investigations of the number of detonation modes and of their stability, it is natural to consider the expansion law (the shape of the streamtubes) as given and independent of the wave intensity, except in the case of weak waves (see below). Any relatively minor dependence of the streamtube form on the wave intensity result only in a small variation of the detonation rate, while the functional relationships remain qualitatively unchanged.

**§1.** Let us consider a detonation process in an infinitely long cylindrical charge and assume that the streamtube cross section $\sigma$ is a given function of the distance x from the shock-wave front initiating the detonation.

With other conditions equal, the velocity of a detonation wave is the higher, the greater is the heat release and the smaller the losses from lateral scattering in the subsonic zone of flow. In this respect the above phenomenon is similar to the detonation process in the unbounded medium in the presence of a nonmonotonic chemical reaction. However, this problem differs significantly in that it results in non-one-dimensional motion, and this leads to a different expression of the laws of conservation. In particular, the particle trajectory in a steady-state shock wave in the pressure-volume plane is no longer defined by the Michelson straight line, and the detonation-wave velocity cannot be determined from the known condition of tangency to the adiabatic curve of maximum heat release by the detonation. For the same reasons losses due to lateral scattering do not result in any appreciable heat losses.

Nevertheless, continuous function $f(D)$ of the shock-wave velocity can be introduced, as in [1], for a bounded medium. When equal to zero, this function defines the velocities $D_i$ of steady-state modes, and when different from zero, it indicates the direction of development of the nonsteady-state process. To prove this we consider a continuity equation together with an Euler equation for steady-state motion in a system of the wave-front coordinates

$$D\rho_1 = u\rho s \equiv j = \text{const},$$

$$udu/dx = -dP/\rho dx, \quad s \equiv \sigma(x) / \sigma(0). \tag{1.1}$$

Here D and u are, respectively, the gas velocities in front of and behind the wave (D is the detonation-wave velocity in a laboratory system of coordinates),

$\rho_1$ and $\rho_2$ are the gas densities in front of and behind the wave, and P is the pressure. We assume the gas motion to be adiabatic [4].

We denote by $\alpha q (0 \leq \alpha \leq 1)$, that part of the heat q released during an irreversible chemical reaction up to a given instant of time, and we represent pressure variation in an adiabatic process with an irreversible chemical reaction in the form

$$dP = c^2 d\rho + (\partial P / \partial \alpha)_\rho \, d\alpha, \tag{1.2}$$

where c is the speed of sound under conditions of a frozen irreversible reaction.

Combining (1.1) and (1.2), we obtain

$$(u^2 - c^2) \frac{d\rho}{\rho dx} = \frac{1}{\rho} \left(\frac{\partial P}{\partial \alpha}\right)_\rho \frac{d\alpha}{dx} - u^2 \frac{ds}{sdx}. \tag{1.3}$$

For a detonation process to be steady the right-hand side of Eq. (1.3), subsequently denoted by $\psi$, must, at the Jouguet point

$$u = c \tag{1.4}$$

satisfy the equation

$$\psi = 0. \tag{1.5}$$

Equations (1.4) and (1.5) define the velocity D of a self-sustaining detonation wave. Equation (1.5) with Eq. (1.4) is usually formulated as the condition for compensation of pressure increase during the reaction by decrease with lateral scattering at the Jouguet point [5, 14]. For a detonation in an unbounded medium the relation $ds/dx = 0$ and the simultaneous fulfillment of Eqs. (1.4) and (1.5) indicates the known condition for completion at the Jouguet point of an irreversible monotonic reaction (or the maximum heat release in a nonmonotonic reaction) [4, 27].

We introduce into our analysis the following function of the shock-wave velocity:

$$f(D) = \frac{1}{\rho} \left(\frac{\partial P}{\partial \alpha}\right)_\rho \frac{d\alpha}{dx} - c^2 \frac{ds}{sdx}\Big|_{x=x_0}. \tag{1.6}$$

Here $x_0$ is a quantity dependent on D and equal to the smallest of the two values of $x_1$ and $x_2$, defined as follows: $x_1$ is the point $x \geq 0$ at which $u = c$; $x_2$ is the point corresponding to the smallest $x \geq 0$ at which $\psi \leq 0$, and such that $\psi < 0$ when $x_2$ (if the point $x_1$ does not exist, by definition $x_0 = x_2$; if $\psi \leq 0$ for all $x \geq 0$, obviously, $x_0 = 0$).

Function (1.6) exists and can be determined for any D and a given law s(x) of streamtube expansion by our solving the Cauchy problem for the system of ordinary differential equations (1.1), which must be supplemented by equations of adiabaticity (or by the Bernoulli equation) and of chemical kinetics. By virtue of continuous dependence of the solution of the Cauchy problem on the initial conditions, this function is continuous and has everywhere a first derivative, except at points of transition from $x_0 = x_1$ to $x_0 = x_2$. At these points $f(D)$ has breaks and, consequently, two derivatives: $f'_-$ and $f'_+$.

A property of $f(D)$ at small D close to the speed of sound in an unperturbed gas should be noted. In this region the streamtube shape cannot be considered in any approximation as independent of D, if a correct picture of the flow is to be obtained. This is so because a streamtube which has passed through the maximum pressure of the sonic perturbation front does not expand at all. In solids this is due to the natural strength of the material, and in liquids and gases to constraints imposed in experiments by the containers. Moreover, under certain conditions the dynamic resistance of a liquid can have a similar effect. In an infinitely weak wave the sonic point is reached for infinitely low expansion existing behind the leading edge, i.e., as we see from (1.3) infinitely small heat release so that a chemical reaction at the sonic point does not result in lateral scattering of material. Thus, in the region of reduced pressure we have for the chemical peak of a weak detonation wave $ds/dx \leq 0$, while in the case of an irreversible chemical reaction $d\alpha/dx$ is positive. Hence, if heat conduction and friction at the walls are neglected, at the weak wave limit we have

$$f(D) > 0. \tag{1.7}$$

If conditions (1.4) and (1.5) are fulfilled simultaneously

$$f(D) = 0, \tag{1.8}$$

and, conversely, if condition (1.8) is fulfilled, from the definition of $f(D)$ and from (1.3) it follows that (1.4) and (1.5) are valid at the point $x_0$. Consequently, Eq. (1.8) defines the parameter D of steady-state detonation modes, whose number is equal to the number of positive real roots D of Eq. (1.8).

We shall clarify the relation between $f(D)$ and the stability of steady-state modes. Let there be a certain steady-state mode $D_i$. To investigate its stability we assume that owing to random perturbation the shock-wave intensity is increased by a small increment $\delta D > 0$, and that the state of the gas in the interval $0 \leq x \leq x_0$ has changed correspondingly, so that in this interval Eq. (1.3) is fulfilled, while in the remaining region $x > x_0$, the state of gas at the instant of this perturbation has not changed. The new value of $D = D_i + \delta D$ will not, generally, be a root of Eq. (1.8), i.e.,

$$f(D_i + \delta D) \neq 0. \tag{1.9}$$

This means that only one of conditions (1.4) and (1.5) is fulfilled at point $x_0$, and that $x_0$ is equal to either $x_1$ or $x_2$.

If $x_0 = x_1$, $x_0$ is the Jouguet point at which with condition (1.9) fulfilled we have

$$\psi > 0. \tag{1.10}$$

From (1.4) and (1.5) it also follows that

$$f(D_i + \delta D) > 0. \tag{1.11}$$

Inequality (1.10) means that at the Jouguet point is released at a higher rate than that required to sustain steady-state mode $D_i + \delta D$, as a result of which the wave will be intensified [1]. (In this case a steady-state solution at the point $x_0$ contradicts irreversibility of the chemical reaction. A detailed gasdynamic pattern of the intensification of perturbations is not considered here). Thus, in the case of (1.11) the perturbation of a steady-state mode is intensified.

If with condition (1.9) fulfilled, we have $x_0 = x_2$, relation (1.5) is valid at point $x_0$, but $c > u$, hence

$$f(D_i + \delta D) < 0. \tag{1.12}$$

We see from Eq. (1.3) that in this case the material is being compressed ($d\rho/dx > 0$) in the subsonic part of the region $x > x_0$. Hence from the first ex-

pression of (1.1) and from the condition of streamtube expansion it follows that $u^2$ decreases with increasing x at a rate more rapid than $1/\rho^2$. Therefore, for a steady-state flow to become sonic or supersonic with increasing x an even more rapid rate of decrease of $c^2$ is always necessary, so that in the adiabatic process $\partial(\rho^2c^2)/\partial\rho < 0$. Yet for all materials capable of propagating compression shock waves $\partial(\rho^2c^2)/\partial\rho < 0$. This derivative is of the same sign as the second adiabatic derivative of $1/\rho$ (used in the theory of shock waves) with respect to pressure.

Thus for $D = D_i + \delta D$ steady-state flow is subsonic throughout and does not satisfy the condition for vacuum at $x = \infty$. In a perturbed mode the transition to vacuum occurs in the non-steady-state rarefaction wave which in propagating through the subsonic flow, overtakes the front, and weakens it. Such perturbed motion is a supercompressed detonation wave [4]. In other words, the generated perturbation $\delta D > 0$ is attenuated when condition (1.12) is fulfilled. It can be shown in a similar manner that a perturbation of opposite sign ($\delta D > 0$) is intensified in the case of (1.12) and is attenutated in the case of (1.11).

The stability condition obtained is expressed in a more concise form (unique for any sign of $\delta D$) as follows. A detonation wave propagating at the velocity $D_i$ is stable (as regards transition into another steady-state mode) with respect to small perturbations, if

$$(df/dD) \pm < 0, \quad D = D_i, \tag{1.13}$$

and it is unstable when the sign of inequality (1.13) is reversed.

The two derivatives at the points $D_i$ are of the same sign. In fact, if the function $f(D)$ with a break at the point $D_i$ changes its sign when passing through that point, the derivatives $f'_-$ and $f'_+$ will be of the same sign. If, however, $f(D)$ does not change its sign, $x_0$ is one and the same differentiable function of D ($x_1$ or $x_2$) on both sides of $D_i$, and, consequently, the function $f(D)$ does not have a break at the point $D_i$. In the following analysis it is not the absolute values of $f(D)$ for all D that are important, but rather its continuity, its vanishing at points (1.8), and the sign of its derivatives at these points. The selection of a function with such properties is nonunique. The function $\varkappa f(D)$, where $\varkappa$ is any continuous piecewise differentiable positive function of D, can be taken as f(D). The derivative of $\varkappa f(D)$ can be made unique at the points $D_i$ by a suitable selection of $\varkappa$. No distinction will, therefore, be made in the following between $f'_-$ and $f'_+$, and $f(D)$ will be represented in all diagrams by a smooth curve.

The fact that criterion (1.13) was derived in the analysis of a special kind of perturbations is immaterial and does not limit the generality of (1.13) (see §3 of [1]).

We shall prove two statements defining the properties of solutions of (1.8).

1) Let at the points $D_i$ satisfying (1.8)

$$df/dD \neq 0. \tag{1.14}$$

We also assume that the thermodynamic functions of the reacting material do not have singularities leading to breaks of the detonation adiabatic curve. We prove that the maximum $D_{max}$ and the minimum $D_{min}$ roots of Eq. (1.8) describe modes which are stable as regards transition to other modes (1.8). The proof of stability of the mode $D_{max}$ is derived by contradiction, as was done in [1]. Let us assume that the solution $D_{max}$ is unstable, i.e., that at the point $D_i$ the converse of inequality (1.13) holds. This means that for small positive increments of $\delta D$

$$f(D) > 0. \tag{1.15}$$

Yet $d\alpha/dx$ remains finite with unlimited increase of D, while $c^2ds/sdx$ increases infinitely. (This statement is meaningless, if $ds = 0$ is assumed a priori an unbounded medium). Passage to the limit of an unbounded medium can be achieved with a nonunique definition of the function $f(D)$ and by our multiplying the latter by $sdx/ds$. However, this is unnecessary, since the stability of $D_{max}$ is an unbounded medium was proved in [1]). Hence it follows that for rather large $D > D_{max}$ the inequality sign in (1.15) is reversed (a detonation wave as $D \to \infty$ necessarily becomes supercompressed). By virtue of the continuity of $f(D)$ this feature proves the existence of a solution of (1.8) for $D > D_{max}$. This contradicts initial conditon and proves the stability of the mode $D_{max}$.

The stability of the mode $D_{min}$ is proved in a similar manner, if property (1.7) of the function $f(D)$ for small D is taken into account.

2) An opposite kind of stability corresponds to the two adjacent roots $D_i$ and $D_{i+1}$ of Eq. (1.8) with limitation (1.14), i.e., if the mode at the point $D_i$ is unstable (stable) it is stable (unstable) at the point $D_{i+1}$. This property of solutions of (1.8) follows immediately from criterion (1.13), and from the continuity and single-valuedness of the function $f(D)$ (Fig. 1).

From the properties defined in 1) and 2) together with the same nonessential limitation (1.14) follows that: a) Eq. (1.8) with property (1.7) taken into account has an odd number of solutions; b) is the solution of (1.8) is unique, it is stable; c) if there are three solutions, the solutions with maximum and minimum $D_i$ are stable, while the third solution is unstable; if there are five solutions in all, three of these are stable.

These properties of solutions of (1.8) formally coincide in accuracy with the properties of steady-state modes in an unbounded medium in the presence of nonmonotonic heat release [1]. However, here these properties, and in particular those of $D_{min}$, are of a different physical nature (see the discussion in §3).

§2. Let us consider quantitatively two simple models of detonation with one and two different rates of heat release, and with lateral scattering taken into consideration. We assume, in [22], that there is little streamtube expansion in the steady-state subsonic

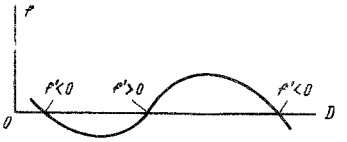


Fig. 1.

region. In this approximation, which will be used from formula (2.9) onward, the streamtube radius r, equal prior to expansion to the charge radius $r_0$, is proportional to x

$$r / r_0 = 1 + ax / d, \tag{2.1}$$

$$d \equiv 2r_0,\ a = \text{const},\ ax_0 / d \ll 1. \tag{2.2}$$

(It is clear a priori that inequality (2.2) is all the better fulfilled the larger is d. It can be proved by means of the solutions to be derived later, that it is permissible to use (2.2) for d decreasing down to the critical diameter.) Furthermore, we assume that the adiabatic exponent $\gamma$ of the gas is constant.

We begin by considering the case of a single irreversible chemical reaction of the monomolecular kind, whose rate is defined by the equation

$$d\alpha / dx = L(D)(1 - \alpha),$$
$$\alpha(0) = 0,\ 1 \geqslant \alpha \geqslant 0. \tag{2.3}$$

This equation, weakly dependent on D, is obtained in an approximation to within the multiplier if the constant of the reaction rate depends only on the shock-wave intensity. The assumption L = L(D) corresponds to the known concept of induction time and of subsequent fast reaction, as well as to the approximate expression for the reaction time in terms of the temperature of the shock-wave front [15, 17].

Solution of (2.3) yields

$$\alpha = 1 - \exp(-Lx),\ d\alpha / dx = L \exp(-Lx). \tag{2.4}$$

The adiabatic derivative $(\partial P/\partial\alpha)\rho$ which enters into (1.3) is determined from the relation

$$dH = VdP,\ V \equiv 1 / \rho,$$
$$H = \gamma PV / (\gamma - 1) + q(1 - \alpha). \tag{2.5}$$

Here H is the enthalpy. We thus obtain

$$(\partial P / \partial\alpha)_\rho = (\gamma - 1)\, q / V. \tag{2.6}$$

Substituting (2.4), (2.6), and $c^2 = \gamma PV$ into Eq. (1.3), and expressing u in accordance with (1.1) in terms of jV/s, we find

$$\left(\gamma PV - j^2 \frac{V^2}{s^2}\right) \frac{dV}{V\,dx} =$$
$$= (\gamma - 1)\, qL \exp(-Lx) - j^2 \frac{V^2}{s^3} \frac{ds}{dx}. \tag{2.7}$$

For Eq. (2.7) to be closed with respect to V in (2.7) we must still express P in terms of V and s = = s(x). With Eqs. (1.1) and (2.5) we find

$$\frac{dP}{dV} = -\frac{j^2}{s^2} + \frac{j^2 V}{s^3} \frac{ds}{dV}. \tag{2.8}$$

We solve (2.8) approximately by assuming the rate of variation of s to be lower than that of V, by integrating by parts the second term in the right-hand side of (2.8), and by taking the function of s out of the integral at the upper limit of V. Neglecting the counterpressure $P_1$ and integrating (2.8), we obtain

$$P = j^2 \left(V_1 - \frac{V}{s^2}\right) + \frac{V_1}{2} \frac{\gamma - 1}{\gamma + 1} j^2 \left(\frac{1}{s^2} - 1\right), \tag{2.9}$$

where $V_1$ is the specific volume of the initial material

Substitution of (2.9) into (2.7) yields

$$j^2 \left[\gamma V_1 - (\gamma + 1) \frac{V}{s^2} + \frac{\gamma V_1}{2} \frac{\gamma - 1}{\gamma + 1} \left(\frac{1}{s^2} - 1\right)\right] \frac{dV}{dx} =$$
$$= q(\gamma - 1) L \exp(-Lx) - \frac{j^2 V^2}{s^3} \frac{ds}{dx}. \tag{2.10}$$

Equating the expression in brackets in (2.10) to zero, we obtain at the Jouguet point

$$V = \frac{\gamma V_1}{\gamma + 1} s^2 \left[1 + \frac{\gamma - 1}{2(\gamma + 1)} \left(\frac{1}{s^2} - 1\right)\right] \approx \frac{\gamma V_1}{\gamma + 1} s^2. \tag{2.11}$$

Substituting this value of V into the right-hand side of (2.10) and equating this side, in accordance with (1.5), to zero, we find

$$(\gamma - 1)\, qL \exp(-Lx) =$$
$$= \frac{\gamma^2}{(\gamma + 1)^2} D^2 s \frac{ds}{dx} = \frac{2\gamma^2}{(\gamma + 1)^2} \frac{aD^2 s^{3/2}}{d}. \tag{2.12}$$

The second equation relating D and x is derived from (2.10) and from the Chapman-Jouguet condition (1.4) written in the form $\gamma PV = (jV)^2/s^2$. Eliminating PV from this by means of the Bernoulli equation

$$j^2 V^2 / 2s^2 + \gamma PV / (\gamma - 1) - \alpha q = {}^1/_2 D^2 \tag{2.13}$$

and using (2.11) and (2.4), we find

$$D^2 [1 + \gamma^2 (s^2 - 1)] =$$
$$= 2(\gamma^2 - 1)\, q [1 - \exp(-Lx)]. \tag{2.14}$$

The transcendental equations (2.12) and (2.14) represent Eq. (1.8) in a parametric form (with x as parameter) for the steady-state detonation modes $D_j$.

To simplify the calculations we use the inequality in (2.2) and substitute unity for s in (2.12) and (2.14). With these simplifications we obtain from (2.12) and (2.14).

$$\frac{D^2}{2(\gamma^2 - 1) q} = \frac{1}{1 + k},\quad k \equiv \frac{4a\gamma^2}{(\gamma + 1) Ld}. \tag{2.15}$$

If $d = \infty$, Eq. (2.15) yields the known relationship between D and q for detonation in an unbounded gaseous medium. The number of solutions of Eq. (2.15) can vary depending on the diameter d and the form of the function L(D).

It can be shown that a function of the kind (1.6), which defines stability of the mode is for the model considered here, of the form

$$f_1(D) = 1 / (1 + k) - D^2 / 2q(\gamma^2 - 1). \tag{2.16}$$

Let us consider the case in which the dependence of L on the temperature of the shock-wave front is subject to a law similar to that of Arrhenius

$$L = A \exp(-\mu E / RT), \tag{2.17}$$

where A is a constant preexponential factor, E is the specific activation energy, $\mu$ is the molecular weight, and R is the gas constant. Equation (2.15) is now transformed to

$$y = 1 + \lambda \exp(my) \equiv 1 + k$$
$$\left(y = \frac{2(\gamma^2-1)q}{D^2},\quad \lambda = \frac{4a\gamma^2}{Ad(\gamma+1)},\quad m = \frac{\gamma E}{(\gamma^2-1)(\gamma-1)q}\right). \qquad (2.18)$$

Expression (2.18) was derived with consideration of relationship $RT = (\gamma - 1)\mu D/2\gamma$ for a strong shock wave in gases.

The dependence of $1/(1 + k)$ on $1/y$ for $m = 2$ and $\lambda = 0.01$, 0.024, and 0.06 is shown in Fig. 2, where steady-state detonation modes are indicated by • and ×. Modes indicated by • are stable, while those indicated by × are unstable. The dashed extension of curves shown in Fig. 2 was constructed with the general property of (1.7) taken into account.

With increasing $\lambda$, i.e., with a decreasing charge diameter, the adjacent upper points converge, and for $\lambda \equiv \lambda_k = 0.024$ they merge into a single point (Fig. 2). The critical diameter $d_k$ of the charge corresponds to $\lambda_k$. There is only one solution when $d < d_k$ (for a weak detonation wave). Its physical meaning is discussed in §3.

The value of $\lambda_k$ is determined by the condition of tangency of the lines $1/y$ and $(1 + k)$. If the diameter for the mode $D_i$ is equal to the critical diameter, not only the function $f(D)$ but also its derivative vanish at the point $D_i$. This is precisely the exceptional case in which (1.14) is not fulfilled.

From the tangency condition it follows that $m\lambda_k \times \exp(1 + m) = 1$. A stable mode with a minimum $D_i$ exists when $\lambda > \lambda$ min, i.e., in a range of charge diameters not exceeding a certain critical value of $d_{max}$. The quantity $\lambda_{min}$ is defined by the condition of tangency of the straight line $1/y$ to the dashed curve (Fig. 2). However, this quantity cannot be calculated for the model considered here.

We now turn to the case of two exothermic reactions occurring at different rates. This case merits special consideration, since it is directly related to the well-known phenomenon of a low rate of detonation. Let the chemical reaction kinetics be defined, similarly to (2.3), by equations

$$d\alpha / dx = L_1(1 - \alpha),$$
$$d\beta / dx = L_2(1 - \beta),\quad \alpha(0) = \beta(0) = 0.$$

The functions $L_1$ and $L_2$ are dependent on D. We denote the heats of the irreversible chemical reactions by $q_1$ and $q_2$, respectively.

The most interesting case is that of a fast reaction characterized by low heat release. Such a relationship between the rate and the heat of a reaction occurs, for example, when the first reaction develops in the inhomogeneities of the material, while the second does so throughout the volume of the compressed material. Accordingly, we assume

$$L_1 \gg L_2,\quad q_1 \ll q_2. \qquad (2.19)$$

Generally speaking, in the case of two reactions the system of equations of the type (2.12) and (2.14) does not reduce to an equation of the type (2.15). However, if conditions (2.19) and $L_1q_1 \gg L_2q_2$ are fulfilled (it is sufficient for this inequality to be fulfilled for $D^2 \ll 2(\gamma^2 - 1)q_1$), a single equation

$$\frac{1}{y} = \frac{Q}{1+k_1} + \frac{1-Q}{1+k_2} \equiv z,$$
$$k_i \equiv \frac{4a\gamma^2}{(\gamma+1)dL_i},\quad Q \equiv \frac{q_1}{q_1+q_2},\quad (i = 1,2) \qquad (2.20)$$

is obtained for D.

The function $f(D)$, is similar to (2.16), of the form

$$f_2(D) = z - 1/y.$$

The results of calculation of the left-hand and right-hand sides of Eq. (2.20) are shown in Fig. 3 for the case of an exponential dependence of $L_1$ and $L_2$ similar to (2.17) and (2.18) with the same relationship between the activation energy $E_i$ and the reaction heat $q_i$.

$$k_1 = \lambda \exp(Qmy),\quad k_2 = \lambda \exp[(1 - Q)my)]$$

for $Q = 0.2$, $m = 3$, and $\lambda = 10^{-4}$, $5 \cdot 10^{-4}$, $5 \cdot 10^{-3}$, $10^{-2}$, and $2 \cdot 10^{-2}$. In addition to the modes existing in the case of a single reaction (Fig. 2) in the interval $5 \cdot 10^{-3} > \lambda > 5 \cdot 10^{-4}$, i.e., within a certain range of charge diameters, there exists one more stable detonation mode.

§3. Depending on the charge diameter, one or three steady-state detonation modes can exist in the case of a single irreversible reaction. Three detonation modes (two stable, one unstable) exist within a certain range $\Delta$ of diameters bounded from above and below. Beyond the limits of $\Delta$ only one mode $D_{min}$ exists for smaller diameters. Similarly, there exists only one mode $D_{max}$ for larger diameters. Thus, the

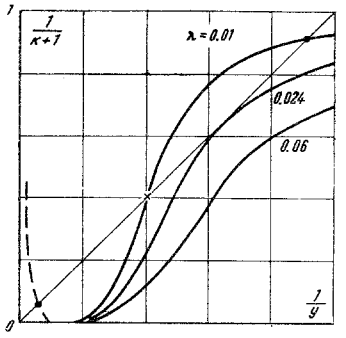

Fig. 2

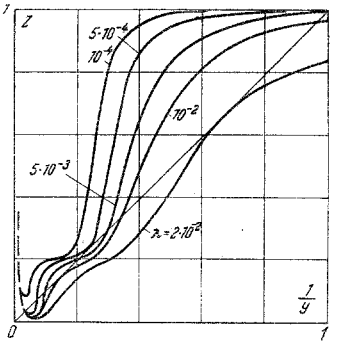

Fig. 3

modes $D_{min}$ and $D_{max}$ are limited by the critical diameters from above and below, respectively. At the limit $\Delta = 0$ both critical diameters coincide. If there are no other parameters, except d, which would make possible variation of the function $f(D)$, the probability of $\Delta = 0$ is nil.

The modes $D_{min}$ and $D_{max}$ are stable. The origin and stability of the mode $D_{min}$ are related to the natural strength of the material or to the presence of a container. In an unperturbed material the rate of $D_{min}$ is close to the speed of sound. If the material is homogeneous, the temperature of the shock wave initiating a chemical reaction in the $D_{min}$ mode is close to the temperature in front of the wave. It follows from this that with an explosive suitable for any extended storage the width of the reaction zone of the mode $D_{min}$ in a charge without a casing is very great, and because of this a steady-state mode can prove to be unattainable in laboratory conditons. Moreover, in a very slow reaction stability of the mode can be disturbed by heat conduction through a lateral surface of the charge [4].

If the charge is encased in a casing, an increase in the strength of the latter increases the rate of $D_{min}$, while even in a homogeneous medium the width of the reaction zone can decrease to dimensions acceptable under laboratory conditions. In this case it will be possible to detect the mode $D_{min}$, provided that the charge diameter has not yet exceeded its critical value.

Two detonation modes in a bounded medium were obtained by Evans [15]. Both modes (which we shall denote by $D_2$ and $D_3$ in the order of increasing rate) were derived in [15] by equating the width $\xi$ of the reaction zone, obtained from the theory of gasdynamics [14, 22, 26] to the function $\xi(D)$ applicable to the model of reaction kinetics considered in [15]. The modes $D_2$ and $D_3$ exist only when $d > d_k$ ($d_k$ is the critical diameter), and the rate of $D_2$ decreases with increasing d. It was assumed in [15] that in certain cases $D_2$ is related to the modes detected experimentally. Results approximately similar to those of [15] are cited in [14]. However, it will be seen readily that $D_2$ and $D_3$ in [15] are nothing else than two of the three "upper" modes occurring in a single irreversible chemical reaction within the interval $\Delta$ of diameters. Since in that case the mode $D_2$ is unstable, it cannot be detected as a steady-state one. With increasing d the mode $D_2$ vanishes together with the mode $D_{min}$, leaving only the mode $D_{max}$. The only mode which can correspond to a low detonation rate in a single chemical reaction is $D_{min}$. The magnitude of $D_{min}$, unlike $D_2$, does not decrease with increasing charge diameter. It can increase or remain nearly constant. These general statements are illustrated by calculations performed for a specific model of a chemical reaction (Fig. 2).

In the case of two chemical reactions occurring at two substantially different rates, two more modes appear together with the three considered here.

In this case the function $f(D)$ has four extrema; namely two minima and two maxima. The coincidence of any of these extrema with the point $D_i$ of the steady-state mode (1.8) can be achieved by our varying the parameter d on which f(D) is dependent. Four critical diameters correspond to the four such coincidences shown in Fig. 4.

If equality of any two of the critical diameters is excluded as improbable, it is possible to prove that with conditions (2.19) and $q_1L_1 \ll q_2L_2$ fulfilled there are five possibilities of emergence and disappearance of five modes for change in d. We number the $D_i$ in an increasing sequence by 1, 2, 3, 4, and 5 (Fig. 2). We denote the occurrence of a single first mode by 1 the first, second, and third modes by 1, 2, 3, and so on. The five possible sequences of emergence and disappearance of modes with increasing d will be denoted as follows: (a) 1, 123, 3, 345, 5,; (b) 1, 123, 12345, 345, 5,; (c) 1, 123,12345, 125, 5,; (d) 1, 145, 12345, 345, 5; (e) 1, 145, 12345, 125, 5,. Unstable modes are denoted here by even numbers (2 and 4), and commas correspond to critical diameters. The sequence (a), for example, indicates that within the range $D_5$ of diameters there is a single mode 1, in the range $d_1$–$d_2$ — three modes 123, in the range $d_2$–$d_3$–$d_4$–three modes 345, and in the range $d_4$–$\infty$—one mode 5. Cases of occurrence of modes 145, 12345, and 125 are shown in Fig. 5.

Not more than two steady-state modes, i.e., 1 and 3 and 5, can be detected in experiments with any charge diameter and the sequence (a). In the case of the four remaining sequences three steady-state modes, namely 1, 3, and 5 can be detected in a certain range of diameters, depending on the manner of initiation.

We note that for the sequence (a) mode 3 can in an experiment be erroneously taken for mode 5. The essential difference between these two is that while the rate of mode 5 increases smoothly to its asymptotic value with increasing d, that of mode 3 vanishes for $d = d_4$, and the rate of $D_3$ changes stepwise up to $-d_1$ In this context more detailed experimental investigations of detonation of explosive mixtures of the kind considered in [28] are of interest.

The functional relationships described are in qualitative agreement with experimental data on the low rate of detonation and on its dependence on the charge diameter [5, 8–12]. Mode 3 is probably detected as a mode with a low rate of detonation in many experiments with charges without casings. In experiments described in [12] modes of types 1, 3, and 5 were apparently achieved.

Data obtained by Cook [5] as regards the relation between the low rate of detonation in gelatins and the extent of saturation of them by air can be explained qualitatively on the basis of our analysis as follows. In the absence of gas bubbles the fast reaction phase is also absent; if the number of bubbles is very great, there is a fast reaction phase but almost all its heat is released. Lastly, with a small number of bubbles, conditions similar to (2.19) obtain, and in this case a steady-state mode at a low rate becomes possible.

Within the framework of our analysis all critical diameters, except the minimum one, can be derived

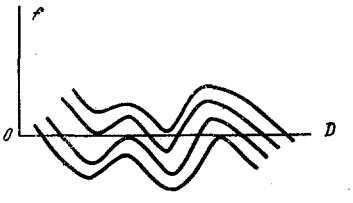


Fig. 4

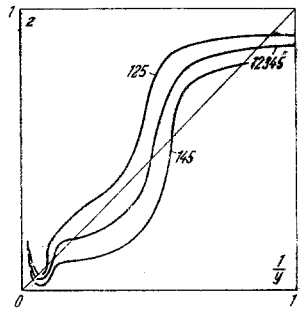

Fig. 5

for a given $a$ (or, generally, for a given shape of the streamtube) and a known reaction mechanism.

Allowance in a stationary mode for any weak dependence of the streamtube form on D leads to a small quantitative, but not qualitative, alteration of the results. It is not, however, excluded that certain interesting results, in particular for diameters close to critical ones, can be brought to light by a more rigorous solution of the system of equations of gasdynamics and chemical reaction kinetics.

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