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INSTABILITY OF COMBUSTION AND DETONATION OF GASES

K. I. SHCHELKIN

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CONTENTS

1. NORMAL COMBUSTION OF GASES

NORMAL or fundamental combustion is, following the nomenclature of V. A. Mikhel'son,[1,2] a plane flame propagating in a stationary gas or in a laminar flow by means of the thermal conductivity, which heats the unburnt gas, and by means of diffusion which supplies it with chemically active particles from the combusting layer. The propagation velocity of normal combustion in a previously prepared homogeneous combustible mixture (this velocity is also called the normal velocity of the flame) is determined both by the coefficients of thermal conductivity and diffusion and also by the chemical reaction rate at a temperature close to the combustion temperature.

The time of chemical reaction τ is from dimensional analysis equal to the time t necessary for a collision between the molecules multiplied by the mean number of collisions per act of chemical reaction:

$$\tau = tn.$$

The number n is obviously inversely proportional to the chemical reaction probability. Assuming the time between collisions to be equal to the ratio of the mean free path l to the mean velocity of the molecules c (the latter is close to the speed of sound), we find the chemical reaction time

$$\tau \approx \frac{l}{c} n. \tag{1}$$

The width of the zone of the chemical reaction is in order of magnitude equal to the product of the chemical reaction time and the speed of the flame:

$$\lambda = \tau u_n = l \frac{u_n}{c} n. \tag{2}$$

In addition, it follows from dimensional analysis that the width of the chemical reaction zone is also some fraction of the total width of the heated region in front of the flame propagating with a velocity u_n:

$$\lambda \sim \beta \frac{\varkappa}{u_n}. \tag{3}$$

Here κ/u_n is the width of the heated region (κ is the coefficient of thermal conductivity) and $\beta < 1$.

Comparing (2) and (3), and bearing in mind that the coefficient of thermal conductivity is

$$\varkappa = \frac{1}{3} lc,$$

we obtain for the normal velocity of the flame the relation

$$u_n = c \sqrt{\frac{\beta}{3n}} = \frac{c}{\sqrt{n}} \varphi, \tag{4}$$

where φ is a quantity smaller than unity.

The normal velocity of a flame is, as is seen from (4), proportional to the speed of sound, but considerably smaller than the latter, since the number of collisions n necessary for one act of chemical reaction is usually of the order of $10^4 - 10^5$.

With the aid of (4) and (2) one can find the width of the channel of the chemical reaction zone—the width of the flame front:

$$\lambda = l \sqrt{n}\, \varphi. \tag{5}$$

It is proportional to the mean free path, but much larger than the latter.

The complete theory of the propagation of normal combustion, now quite generally accepted, is that of Ya. B. Zel'dovich and D. A. Frank-Kamenetskiĭ.[3] It is not presented here, because for what follows the general physical facts on the normal flame presented above are sufficient.

The normal velocity of a flame is one of the main physico-chemical constants of volatile fuels. It increases with increasing initial temperature of the mixture and changes little with changing initial pressure. The normal velocity reaches a maximum in a mixture with a certain deficiency of air (oxygen) compared with a mixture in which there is just enough of

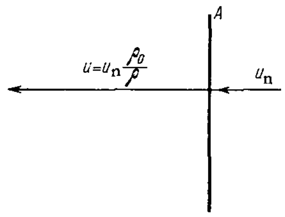


FIG. 1. Diagram of a normal flame.

it for complete combustion. For air mixtures of hydrocarbons the maximum normal velocity amounts to 0.3–0.4 m/sec. The maximum normal velocity in a hydrogen-air mixture is 2.67 m/sec. This is generally the maximum velocity recorded in air mixtures of combusting gases.

Oxygen mixtures of combusting gases and vapors for which the number of collisions necessary for one chemical reaction act is considerably smaller than for air mixtures have higher normal velocities (but still far from the speed of sound). Thus, the maximum normal velocity in a butane-oxygen mixture is 4.8 m/sec and in a hydrogen-oxygen mixture it is about 10 m/sec.

The width of the chemical reaction zone in a normal flame in air mixtures is of the order of several tenths millimeters, and in oxygen mixtures it is even less.

Let us imagine that the normal combustion front is stationary (Fig. 1). The unburnt gas enters into the combustion zone with a velocity u_n, the combustion products escape from it in accordance with the law of conservation of mass with a velocity

$$u = u\frac{\varrho_0}{\varrho}, \tag{6}$$

where ρ_0 and ρ are the densities of the unburnt gas and of the combustion products respectively.

By virtue of the conservation laws of mass, momentum, and energy there appears on the combustion front a pressure drop whose magnitude for the case $u_n \ll c_0$ is given by the expression [10]

$$p - p_0 = -p_0 q\left(\frac{u_n}{c_0}\right)^2 = -p_0 q M^2. \tag{7}$$

Here c_0 is the speed of sound in the unburnt mixture, q is the ratio of the thermal effect of the combustion reaction to the initial internal energy of the gas

$$q = \gamma(\gamma - 1)\frac{Q}{c_0^2},$$

γ is the ratio of the heat capacities; M in formula (7) is the Mach number of a normal flame, i.e., the ratio of the normal speed to the speed of sound in the unburnt gas mixture.

The pressure drop on the front of a normal flame turns out to be very small. Thus, for example, for an air mixture of pentane (C_5H_{12})

$$u_n = 0.35 \text{ m/sec}, \quad c_0 = 340 \text{ m/sec}, \quad M = 10^{-3}, \quad q = 7,$$

and the difference between the pressures on both sides of the zone of the normal flame is

$$p - p_0 = -p_0 q M^2 = -p_0 \cdot 7 \cdot 10^{-6} \approx -10^{-5} p_0.$$

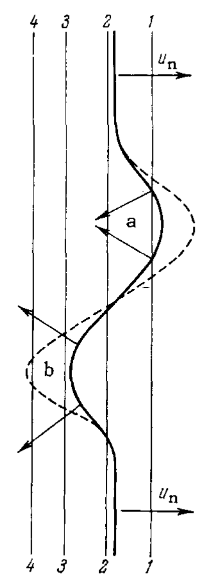
At atmospheric pressure the drop on the flame front is in this case about 0.1 mm of the water column. In oxygen mixtures it is approximately by one to two orders higher, but even in this case it remains very small.

2. INSTABILITY OF NORMAL COMBUSTION

The problem of the stability of normal combustion against infinitesimal perturbations, which is of fundamental significance for the theory of combustion, was posed and solved by L. D. Landau as early as 1944.[11] Making use of the considerable pressure drop on the front of a normal flame, L. D. Landau applied to the problem the exact equations of perturbation theory. Without carrying out the calculations which indicate the absolute instability of a plane front of normal combustion, we will indicate the physical reason for this phenomenon.

We superimpose on a normal combustion front (Fig. 2) a perturbation—we distort the front as indicated by the wavy line—and follow its behavior. When the gas crosses a portion of the flame front which makes an angle with the approaching flow, the tangential component of the speed of the gas remains unchanged, and the normal component increases in proportion to the ratio of the densities ρ_0/ρ. The gas stream which crosses the combustion surface at an angle will be deflected as indicated on Fig. 3. As a result a flow will occur behind the wavy front of the flame whose flow lines are schematically indicated in Fig. 2. The flow lines converge behind the convex portions of the flame (in zones a) and diverge behind the concave portions (in zones b). In the 4—4 plane

FIG. 2. The initial perturbation of the normal front (solid line) is increased (dashed line), because in the combustion products for the cross sections 1–1 and 2–2 in region a $p_1 > p_2$, and for the cross sections 3–3 and 4–4 in region b $p_3 < p_4$.



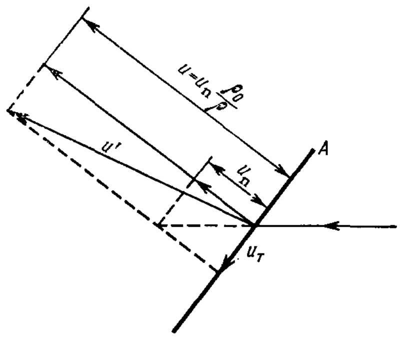


FIG. 3. Deflection of a gas flow by the front of a flame.

behind the flame front the pressure is constant throughout, but a certain nonuniform distribution of speed remains: the speed of the combustion products behind the convex portions of the front is somewhat higher than behind the concave portions.

The gas behind the convex portions (in zones a) is under conditions similar to those in a converging nozzle (confuser) and behind the concave portions (b zones) similar to those in a diverging nozzle (diffuser). For this reason the pressure is increased slightly in zones a, and decreased slightly in zones b compared with the pressure in the 4—4 plane and in the unburnt gas. As a result the initial perturbation represented by the wavy line increases, and the flame front is further distorted as indicated by the dashed line.

On Fig. 2 the front has no width. Actually in considering the wavelength of the perturbation one must take into account the finite dimensions of the combustion zone (Fig. 4). The wavelength of the perturbation and consequently also the linear dimension of the surface of the combustion front should, if we follow the qualitative instability mechanism described above, exceed the width of the combustion zone, more precisely the zone of heating. At the instant when the dimension of the flame R exceeds the width of this zone, i.e., the instant at which the flame front itself will appear, the initial perturbations will, by virtue of the absolute nature of the instability, begin to grow.

The condition for the appearance of the instability can be written in the form

$$R > \frac{\varkappa}{u_n}$$

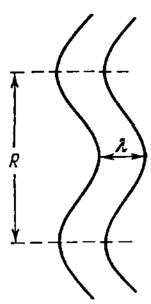


FIG. 4. R is the length of the perturbation wave, and λ is the width of the flame front.

or

$$\mathrm{Re}_n = \frac{R u_n}{\varkappa} > 1. \qquad (8)$$

When the dimensionless criterion (8), reminiscent in its form of the Reynolds criterion, exceeds a quantity of the order of unity, the plane zone of the normal combustion ceases to exist, the flame becomes self-turbulent, and turbulent combustion appears which follows laws other than the laws of normal combustion.

The instability of a normal flame was first observed experimentally by Zel'dovich and Rozlovskiĭ.[12] They ignited a mixture of 56.6% hydrogen, 41.1% oxygen, and 2.0% carbon disulfide inside a steel bomb 150 mm in diameter and having a window for photography. Whereas at initial atmospheric pressure the flame reached the walls of the bomb without change in the nature of the propagation, on increasing the pressure the velocity increased abruptly at some distance from the walls and the combustion changed into detonation. The change occurred at a value of Re_n of (8) of $2 - 5 \times 10^5$. Zel'dovich and Rozlovskiĭ assume that the transition from combustion to detonation is due to the instability of normal combustion. The unexpectedly large value of the criterion (10^5 instead of 1) indicates a greater stability of normal combustion than predicted theoretically. The flame is stable with respect to perturbations which exceed in their length by 4—5 orders the width of the combustion zone, becoming unstable only with respect to relatively long perturbations. The instability turned out not to be absolute.

In the experiments described the action of the chamber walls on the flame front was not excluded. Weak perturbations (including weak shock waves) reflected by the walls of the chamber and meeting the flame may interact with it. It cannot be stated in advance whether they stabilize or increase the instability of the flame.

The effect of the chamber walls was practically completely excluded in the experiments of Rakipova, Troshin, and the author of this article; these experiments were carried out with acetylene-oxygen mixtures enclosed in soap bubbles and ignited at the center with a red-hot loop.[13] Photographs of the spherical flame through a narrow slit in a screen separating the soap bubble from the camera indicate a gradual increase of the velocity of propagation of the flame, beginning at the instant when criterion (8) reaches a value $3 - 6 \times 10^4$. In [13] distortions of the flame front were recorded, indicating that the front was self-turbulent and appearing after Re_n of (8) increased to the same value of $3 - 6 \times 10^4$.

Subsequently the appearance and growth of instability was studied by the Toepler method.[14] For criteria (8) exceeding 10^4 for acetylene-oxygen mixtures and 0.7×10^4 for mixtures of Saratov (natural) gas with oxygen, there appeared on the front perturbations of increasing intensity—wave-like inhomogeneities.

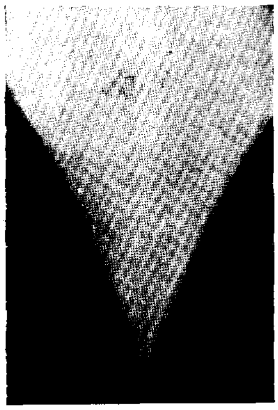


FIG. 5. Photograph of the acceleration of a flame in a mixture of 33% C_2H_2 and 67% O_2 in a soap bubble.

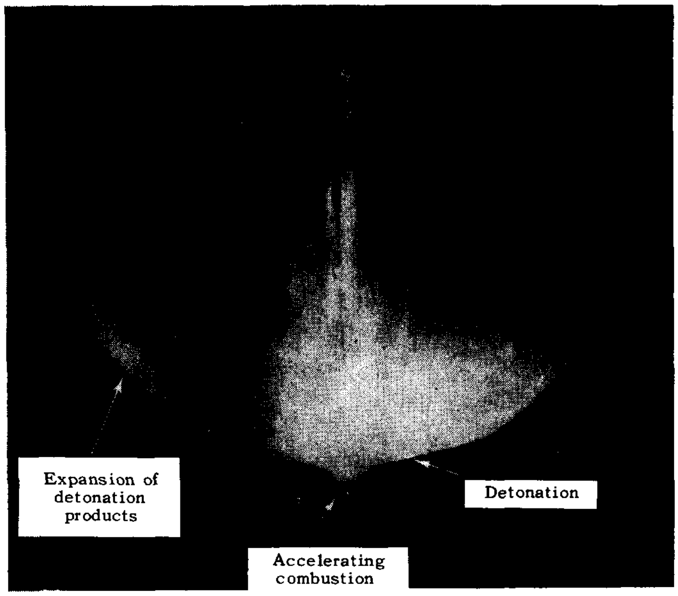


FIG. 6. Transition of a slow spherical flame into a spherical detonation in a mixture of 22% C_2H_2 and 78% O_2 enclosed in a thin rubber sphere.

Figure 5 shows the progressive acceleration of a spherical flame in an acetylene-oxygen mixture indicating its instability. Figure 6 shows the transition of slow combustion into spherical detonation in a thin rubber sphere due to the instability of normal combustion.

Self-turbulence was also observed by a group of authors directed by Yu. Kh. Shaulov.[15] The results of this work coincide with those described above.

The stability of a normal flame with respect to relatively short-wavelength perturbations observed experimentally and contradicting theory requires a consideration of stabilizing effects not taken into account in Landau's theory.

One of the stabilizing factors (which was pointed out in the first experimental work of Zel'dovich and Rozlovskiĭ[12]) may turn out to be the effect of the curvature on the thermal flux, increasing the velocity of normal combustion at the concave surface and decreasing it at the convex surface. Thus the top of the Bunsen flame is always rounded off because the velocity of the concave flame is always larger than the velocity of a plane flame. For this reason perturbations on a plane flame are evened out: the flame propagates faster in the retarded (concave) sections and more slowly than for a plane flame in the convex sections. The effect of the curvature on the velocity of the flame can stabilize combustion with respect to perturbations approximately by an order longer than the zone of heating. This increases the criterion (8) to approximately 10. However an increase by one order of magnitude does not remove the difficulty. The discrepancy between the experimental and theoretical values of criterion (8) remains larger by about three orders of magnitude.

The effect of viscosity on the flow of gas stabilizes combustion, the effect being stronger for short-wavelength perturbations than for long-wavelength ones. However, the discrepancy between the theory and experiment by several orders is difficult to ascribe to such a weak stabilizing effect.

In Landau's work the following conditions were assumed on the boundary dividing the burnt and unburnt gas:

$$v_{1x} - \frac{\partial \xi}{\partial t} = v_{2x} - \frac{\partial \xi}{\partial t} = 0.$$

Here v_{1x} is the perturbation of the velocity of the unburnt gas along the x axis, perpendicular to the perturbed front, v_{2x} is the perturbation of the velocity of combustion products along the same axis, ξ is the displacement of the combustion front.

These conditions correspond to a constant velocity of propagation of the flame along the x axis. Under this condition the flame shifts (is translated) from the equilibrium position in correspondence with the change in the rate of the gas flow, i.e., with its perturbation. On increasing the velocity the flame shifts towards the burnt gas, and on decreasing it it shifts towards the unburnt gas. At the same time each point on the front shifts independently of its neighboring points.

Actually, however, the flame front does not represent an independent inflammation of gas particles, the flame propagates in all directions (over the unburnt gas) with a constant normal velocity. For this reason, strictly speaking, one must set on the boundary the constant velocity of the flame in a direction perpendicular to the normal front.

Normal propagation has a stabilizing action. Combustion "consumes" the gas more rapidly in front of the concave sections of the combustion zone than in front of the convex sections. Let us consider the distortion of the flame front F unsupported (unhindered) by an external reason (Fig. 7). Applying Huygens' principle to combustion one can readily convince oneself that the given perturbation decreases spontaneously. The amplitude of the perturbation decreases particu-

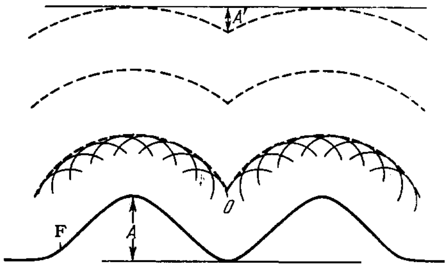


FIG. 7. Decrease in the amplitude of the initial distortion of the flame from A to A′ because of the equalizing action of the combustion propagation.

FIG. 8. Spherical unaccelerated flame with slowly attenuating perturbations on its surface in a mixture of 10% Saratov (natural) gas and 90% air. The light spot at the end of the dark arrow is the center of the sphere.

larly rapidly at the instant of the appearance of a discontinuity O on the surface of the front. With time the rate of decrease of the amplitude decreases and approaches zero asymptotically. The very slow smoothing out of the perturbation during the last stage leads incidentally to an interesting phenomenon. One can often observe on a spherical flame a structure reminiscent of the surface of a volley ball. Perturbations on the surface of a flame when it is stable decrease rapidly during the initial stage of smoothing out and very slowly during the last stage. For this reason the traces of the perturbations remain on the surface of the flame for a long time, giving it the form shown in Fig. 8.

In order to be convincing, the above qualitative discussion about the stabilizing effect of the propagation of combustion requires quantitative estimates. A rigorous solution of the problem is thus of considerable interest.

Recently the assumption was advanced (see [10], p. 187) about the existence of another stabilizing effect. In a spherical flame, so long as it is small in dimension, perturbations can average over the surface of the front and can stabilize combustion. The time needed for propagation of the perturbations over the combustion products from one section of the sphere to another (opposite) section is of the order of R/c_{com} where c_{com} is the speed of sound in the combustion products. The time characteristic for combustion (the time during which the flame propagates over a distance equal to the thickness of the flame zone) is of the order of λ/u_n. Hence the condition for the smoothing out of perturbations in the burnt gas can be written in the form

$$\frac{R}{c_{com}} > \frac{\lambda}{u_n}, \quad \text{or} \quad R u_n > \lambda c_{com}.$$

Dividing both sides by the thermal conductivity, we obtain a criterion for the absence of smoothing out of perturbations

$$\frac{R u_n}{\varkappa} > \frac{\lambda c_{com}}{\varkappa}, \quad \text{or} \quad \mathrm{Re}_{fl} > \frac{c_{com}}{u_n}. \tag{9}$$

Substituting numbers, we find the ratio c_{com}/u_n for oxygen mixtures to be equal to 10^2—10^3, and for air mixtures 10^3—10^4. The numbers agree almost precisely with the experimental value of criterion (8). More exact agreement should not be expected when the two criteria (8) and (9) are correct only to within the order of magnitude.

It is not impossible that the smoothing out of perturbations over the combustion products will turn out to be the strongest stabilizing factor absorbing all the other stabilizing effects: the action of the combustion propagation, the effect of the viscosity, and others. Thus, to verify the theory and for elucidation of the true meaning of criterion (8) experiments are essential in which smoothing out would be excluded.

One must mention two papers in which stable combustion modes were obtained.[16,17] It is, however, difficult to compare their results with the experiments cited above, since the theoretical problem was solved under the assumption of an induction mode. The stability was attained under a more complicated form of conditions on the combustion surface. The experimentally observed transition from stable combustion with short-wavelength perturbations to instability with long-wavelength perturbations was not confirmed by the investigations.

One of the factors stabilizing combustion is, as was noted above, the dependence of the velocity of propagation of the flame on the curvature of the front. The thermal flux into the unburnt gas increases near concave sections of the front and decreases near the convex sections. Therefore the velocity of the flame increases around concavities and decreases near convexities on the flame front, and this indeed stabilizes the flame when the wavelength of the perturbation is equal to or exceeds approximately by an order of magnitude the width of the combustion zone.

However, all that has been said is correct, as was first noted by Zel'dovich,[6] only when the diffusion coefficient of the combusting material and of the oxidizer is approximately equal to the coefficient of thermal conductivity of the mixture. The situation changes radically if the diffusion coefficient of the component which determines the process (of which there is little) exceeds the coefficient of the thermal conductivity of the mixture. In this case so-called diffusion combustion takes place at the combustion boundary: the front of the flame is at rest (or almost at rest), the combusting component flows from the unburnt gas by virtue of the diffusion. Here the velocity of the flame near convexities of the front increases compared to the velocity in a plane front, and near concavities it decreases. The effect of the front distortion on the velocity of the flame changes sign compared with the case when the diffusion coefficients and the thermal conductivities are close to each other. The point is that in diffusion combustion the supply of the combusting component (of which there is not enough) is greater near the convex sections of the front (the diffusion to the front is from a larger volume) and smaller in the concave sections compared with a plane flame. Therefore at points where random convexities have appeared the flame propagates more rapidly, it extends forward. The combustion moves over the unburnt mixture in the form of separate bell-shaped or spherical bubbles. This type of instability was first observed in pure form by V. I. Kokochashvili in a mixture of hydrogen with bromine (35–40% H_2 + 65–70% Br) at a pressure of 200 mm Hg in downward propagation of a flame.[6]

No continuous combustion front is produced in this type of instability, because the gas between the spheres (bubbles) becomes strongly hydrogen deficient. The propagation limit of the flame turns out to be broader than the propagation limit with a continuous front. By virtue of the concentration of the combustion in different sections (diffusion collects the combusting material from the surrounding space) the flame propagates at compositions which are outside the propagation limits of a continuous front.

As a result of instability the lower limit of propagation of a flame in mixtures containing hydrogen is lower when the flame moves upward compared with the case of downward propagation. The bubbles or spheres are pulled up by convection. In downward motion there is no convection and the propagation possibilities decrease on this account. Diffusion combustion and the instability connected with it is not only observed in hydrogen-containing mixtures. In mixtures containing a large amount of heavy phlegmatizer, whose molecular weight is considerably larger than the molecular weight of oxygen and of the combusting materials (carbon tetrachloride, CCl_4, in a mixture of carbon or methane with oxygen), the diffusion coefficient of the combusting component (or the oxidizer) is considerably higher than the thermal conductivity of the mixture. In such mixtures diffusion combustion and the instability of the plane flame connected with it are also observed.

The very subtle phenomena at the limits of propagation of the flame in phlegmatized mixtures and in mixtures containing hydrogen, discovered and investigated by V. I. Kokochashvili and Ya. B. Zel'dovich,[6] may turn out to be essential for the techniques of safe explosions.

3. DETONATION

Detonation is the name given to combustion propagating in broad tubes with a constant ultrasound velocity completely determined for each combustible mixture. For example, the detonation velocity in a hydrogen-oxygen mixture (the composition in each case is stoichiometric) is 2800 m/sec, of methane with oxygen—2320 m/sec, and of pentane (C_5H_{12}) with air—1710 m/sec. Detonation combustion is always accompanied by a strong increase in the pressure and by a considerable increase in the density of combustion products compared with the initial combusting mixture. The combustion products move in the detonation wave in the same direction as the detonation. In contrast to detonation, slow combustion, for example normal combustion which was discussed above, is accompanied by a decrease in the pressure and density in the combustion zone, the products of the combustion move in it in a direction opposite to that of the front of the flame.

The gas-thermodynamic theory of detonation was developed already at the end of the last and at the beginning of this century. In the plane case one obtains from the laws of conservation of mass, momentum, and energy (and the equation of state) for the gas crossing the combustion front (four equations with five unknowns—the velocity of the wave D, the velocity of the combustion products w, the pressure p, the density ρ, and the temperature in the wave) expressions for the velocity of the wave and the velocity of the combustion products in the laboratory system of coordinates

$$D=\sqrt{\frac{(p-p_0)\,\varrho}{(\varrho-\varrho_0)\,\varrho_0}}\,, \tag{10}$$

$$W=\sqrt{\frac{(p-p_0)\,(\varrho-\varrho_0)}{\varrho\varrho_0}}\,. \tag{11}$$

Eliminating the velocities (of the wave and of the combustion products) and the temperature, one finds a connection between the pressure and the density of the combustion products—the Hugoniot equation

$$\frac{p}{p_0}=\frac{\varkappa-\frac{\varrho_0}{\varrho}+\frac{2\gamma Q}{c_0^2}}{\varkappa\frac{\varrho_0}{\varrho}-1}\,. \tag{12}$$

Here, as in (10) and (11), p_0 and ρ_0 are the pressure and density of the initial gas, and $\kappa = (\gamma + 1)/(\gamma - 1)$.

In Fig. 9 we present a graph of the Hugoniot equation. The upper branch of the curve corresponds to detonation ($p > p_0$). The conservation laws allow for each mixture (given p_0, ρ_0, and Q) an infinite number of detonation velocities corresponding to any pressure on the upper branch (above the point E) of the Hugoniot curve. Detonations whose pressures lie above the point B are called strong, those whose pressures lie below are called weak. The point B describes the Chapman-Jouguet (C.—J.) detonation. This detonation and only it, as was first shown by D. Chapman (1899) and E. Jouguet (1904), corresponds to the experimen-

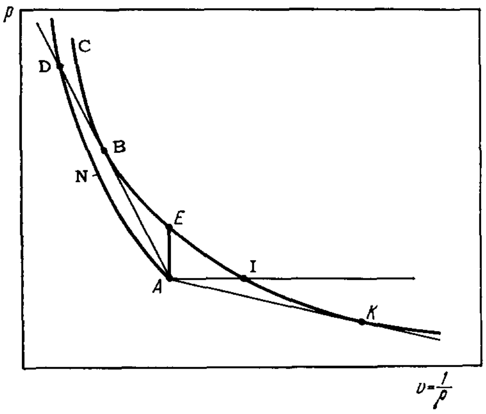


FIG. 9. Hugoniot curve. N – for shock waves, C – for combustion waves.

tally observed spontaneously propagating detonation having a completely determined velocity for each mixture. The selection rule for the unique value of the detonation coincides with the rule for determining the point of tangency B of the straight line AD (it is called the Michelson line) with the Hugoniot curve. At the point of tangency B the detonation velocity relative to the velocity of the combustion products is precisely equal to the speed of sound in them. For this reason the C.—J. detonation propagates without attenuation and with constant velocity. The rarefaction waves, as well as the weak compression waves, appearing behind the combustion front, which move always with the speed of sound, do not overtake the front and it propagates indefinitely without attenuating or becoming stronger.

A strong detonation will not propagate spontaneously with constant velocity, it will be weakened by rarefactions whose velocity (the speed of sound) is always larger than the velocity of the front relative to the combustion products. A strong detonation propagates without attenuation if, for example, a piston moves behind it with the velocity of the combustion products. No rarefaction waves appear in this case behind the detonation front.

A weak detonation—all the points on the section B—E—is possible if there exists an external source of inflammation which ignites the gas with a velocity larger than the velocity of C.—J. detonation. A limitingly fast weak detonation (the velocity of propagation is infinite, and on Fig. 9 the point E corresponds to such a detonation) can be produced in a mixture which is liable to ignite under illumination if one simultaneously illuminates the entire tube by an intense source. Inflammation at constant volume is also a limitingly fast weak detonation.

The section E—I of the Hugoniot curve has no physical meaning, it corresponds to imaginary velocities of the propagation of combustion.

The lower branch of the Hugoniot curve (all points below I) refer to deflagration—propagation of combustion with constant velocity smaller than the speed of sound. In deflagration combustion is accompanied by an expansion of the combustion products which like in all expansion waves move in a direction opposite to the motion of the front. In detonation combustion is accompanied by a contraction of the gas and the combustion products move in the direction of the propagation of the combustion wave.

The velocity of deflagration, and also the velocity of the combustion products is calculated from the same formulas as detonation. However, for deflagration there are no selection rules distinguishing some point on the Hugoniot curve, for example the point of tangency K—the C.—J. deflagration. Thermodynamically arbitrary velocities of deflagration are possible, but only on the section of the weak deflagration I—K is its magnitude determined by the physico-chemical properties of the mixture. Strong deflagrations (section below the point K) are impractical from thermodynamic considerations, although they are allowed by the conservation laws.

An example of weak deflagration is the normal velocity of a flame, the slowest of all actually occurring deflagrations. The state of the combustion products in it lies extremely close to the point I—combustion at constant pressure.

Thus far we were discussing the gas-dynamical description of detonation. The physico-chemical aspect of the phenomenon was first approached by Zel'dovich.[32,8,6] He drew attention to the fact that the gas in a detonation wave does not ignite instantaneously. After its compression some time is needed for the development of the chemical reaction. Consequently, a shock wave should always move before the combustion front in a detonation wave with the same velocity as the front. Thus detonation is a complex consisting of a shock wave and a combustion zone.

The velocity of the wave is proportional [as is seen from (10)] to the square root of the tangent of the angle of inclination of the straight line AD to the abscissa axis of Fig. 10. From the condition of the equality of the velocities of the shock wave and the combustion zone it follows that the state of the shock-compressed, but as yet unreacted, gas and the state of the combustion products should lie on the same straight line. The pressure in the shock wave is thus determined by the point D, and that of the combustion products by the point B. It is interesting that the point B describes detonation if the initial state is taken to be the point A and deflagration if the initial point is D—a compressed gas. Thus the complex constituting the detonation consists of a shock wave and the Chapman—Jouguet deflagration moving along on it.

The length (depth) distribution of the pressure in the complex is shown schematically on Fig. 11. The length of the "little square" is determined by the induction period of the inflammation reaction. The shape of the curve on the section N—B on which the pressure

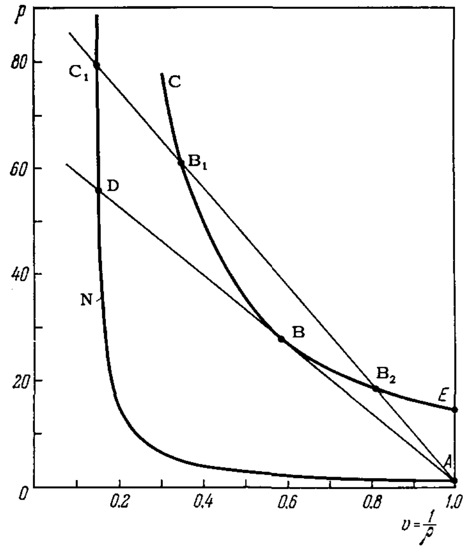


FIG. 10. Detonation portion of the Hugoniot curve.

drops from C to B depends on the course of the chemical reaction. Usually the reaction rate depends on the temperature exponentially with a large value of the exponent. Therefore the pressure distribution in the shock wave-deflagration complex is often shown under the assumption that after the induction period the pressure has altogether not changed, and after its completion the mixture is instantaneously combusted (Fig. 12).

The detonation shown in Figs. 11 and 12 is usually called the Zel'dovich-Neumann detonation. J. Neumann (USA)[19] proposed it two years after Zel'dovich. A year later Döring also proposed it.[40] The works of Neumann and Döring were for a long time unknown in the Soviet Union, as were the works of Ya. B. Zel'dovich abroad. For almost twenty years the Zel'dovich-Neumann model of detonation was considered fundamental although not the only one. As early as 1926 Campbell and Woodhead[20] in England discovered spinning detonation. They showed that in a smooth tube with mixture compositions with a large excess or deficiency of the combusting component the combustion zone is clearly not planar. Combustion in the

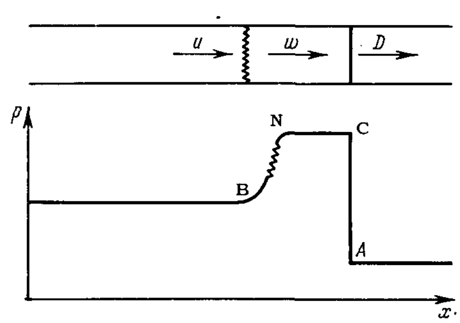


FIG. 11. Schematic distribution of the pressure in detonation considered as a complex consisting of a shock wave and an inflammation zone.

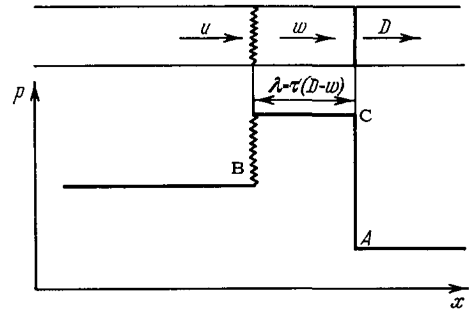


FIG. 12. Extremely simplified diagram of detonation.

form of a nucleus propagates in the forward direction with the velocity of the detonation and simultaneously rotates about the tube axis. From the nucleus the flame propagates at some distance behind the front over the entire cross section of the tube. A photograph of spin detonation on a moving film is shown in Fig. 13. For comparison, a "normal" (not spinning) detonation is shown on Fig. 14. The velocity of the spin detonation along the tube axis (the velocity of the process as a whole), disregarding the clearly three-dimensional structure of its front, coincides accurately with the velocity of detonation in the same mixture calculated according to the classical one-dimensional theory.

Spinning detonation which does not fit into the Zel'dovich-Neumann model was considered for many years an exceptional phenomenon characteristic only of certain gas mixtures, for example, of carbon monoxide with oxygen. Then Kh. A. Rakipova, Ya. K. Troshin, and the author of this article showed (for references see [10]) that detonational spin occurs always and in all mixtures near the propagation limits of detonation, by whichever method they were attained: by decreasing the diameter of the tube, by decreasing

FIG. 13. Photograph of spin detonation in a mixture of 20% H_2 and 80% air on a moving film. The detonation is propagating from left to right and the film is moving from top to bottom.

FIG. 14. Photograph of "normal" detonation in a mixture of $2H_2 + O_2$ on a moving film. The detonation moves from left to right, and the film from the top down.

the initial pressure of the mixture, or by changing the concentration of the combusting component.

After a long and very difficult search the author of this article[21] and Zel'dovich succeeded in establishing that the spin is of gasdynamical nature, and constitutes an inclined overdriven detonation propagating with constant velocity in a spiral along the tube walls. The combustion in the spinning detonation, as it turned out, begins behind the front NP of a triple Mach configuration (see Fig. 24a). The wave NV of the same configuration constitutes a direct shock front whose characteristics can be calculated from the Zel'dovich-Neumann theory.

But this did not exhaust the capabilities of detonations to prepare surprises for the investigators. B. V. Voĭtsekhovskiĭ and his co-workers,[23] after careful investigation of the structure of the front of a spinning detonation and measuring the pressure in its various zones, observed (at least under certain conditions) a structure of the spin of considerably greater complexity than was previously assumed. In particular they recorded considerably higher pressures than those calculated according to the old scheme (see Fig. 24a). They recorded an inclination angle of 75° between the combustion zone in the spin nucleus and the tube. Here a contradiction also appeared with the theory, since according to the theory this angle cannot be larger than 45°. B. V. Voĭtsekhovskiĭ expressed the assumption that the inflammation of the gas occurs in the spin behind the transverse wave NK and not NP, as was assumed according to the theory. But gasdynamic calculations did not confirm this assumption. Then in order to obtain correspondence between the experiment and the calculations, a new structure of the spin was assumed in Voĭtsekhovskiĭ's laboratory. Roughly speaking, the nucleus of the spin consists according to this hypothesis of two triple configurations. In addition to the configuration shown in Fig. 24a, in spin in a compressed gas 2 another identical configuration is propagated, and combustion occurs in two inclined waves: in the "old" wave NP and in the "transverse" wave traveling in the gas 2 (it is not shown on Fig. 24a). A more appreciable portion of the mass of the gas burns in the "transverse" wave of the second configuration. In addition one can also find evidence for the first hypothesis of Voĭtsekhovskiĭ on inflammation in the wave NK (see Fig. 24a). If the gas is very close to the limit of propagation of a detonation and the inflammation time in the wave NP is sufficiently large, and the wave NP itself is stable against distortions (this type of stability will be discussed below), then inflammation in the boundary layer behind the wave NK cannot be excluded following two retardations of the gas: one behind the wave NV, and the other behind the front NK. In this instance the temperature in the boundary layer behind the wave NK is considerably higher than behind the front NP even after retardation of the gas in the boundary layer behind this front. Such a scheme is more logical. A natural place is immediately found for the phenomenon discovered by Voĭtsekhovskiĭ and his laboratory co-workers. It should be emphasized that the calculation of the triple configuration with retardation in the boundary layer is not trivial. The conception above is based more on numerical estimates than on a rigorous internally consistent calculation. The usual trivial gasdynamic calculation of inflammation behind the transverse wave NK leads to contradiction with the conservation laws; for this reason indeed a more complex gasdynamic scheme of the spin was worked out containing some contradictions. If the just stated conceptions in favor of the initial hypothesis of Voĭtsekhovskiĭ with allowance for the retardation of the gas in the boundary layer were confirmed, then the spin structure discovered in his laboratory would occupy a natural place among other quite diverse detonation structures. The contradictions on which it is not here the place to dwell, would be resolved most successfully. However, let us return to the primary subject of this article—the instability of combustion and detonation.

Even the first investigators of the spin detonation noted that on departing from the limiting mixture composition for the propagation of detonation the number of spin nuclei increases, and the spin becomes multiple. Then the detonation becomes "normal" and planar. In 1957-1959[24,25] Yu. N. Denisov and Ya. K. Troshin undertook an investigation of multiple and "normal" detonation by means allowing them to observe in the detonation front inhomogeneities considerably smaller in size than they could observe previously. It turned out that not only spinning detonation (simple and multiple), but also "normal" detonation has actually a three-dimensional structure. In connection with the first experiments of Denisov and Troshin the author of this article undertook to investigate the stability of a planar detonation in the Zel'dovich-Neumann model. The detonation, as a physical analysis showed, turned out to be unstable.[26]

4. INSTABILITY OF A PLANAR GASEOUS DETONATION WITH RESPECT TO DISTORTION OF THE COMBUSTION FRONT

Instability in detonation differs in principle from instability in normal combustion. In normal combustion instability can appear as a result of the distortion of the gas flow on crossing the combustion front, or as a result of a difference in the coefficients of diffusion and thermal conductivity. On the other hand, instability in detonation is connected with the exponential temperature dependence of the chemical reaction time. A small increase (decrease) in the temperature of the unburnt gas in detonation leads to a sharp decrease (increase) in the time from the initial compression in the wave to the instant of inflammation of the gas in it. Both the instability of detonation with respect to distortion of the inflammation front, considered in this section, and the instability with respect to one-dimensional perturbations to which the next section is devoted are related to this phenomenon.

Figure 15 shows the cross section of the complex which constitutes the detonation. Here VV is the shock compression front, BKLKB is the inflammation zone,

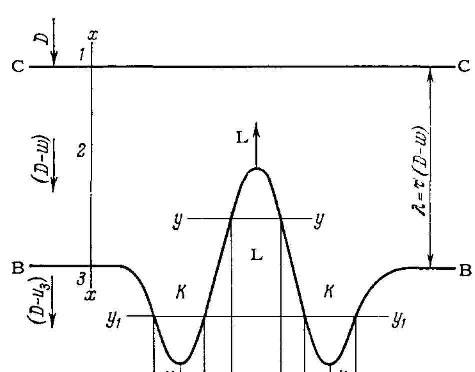


FIG. 15. Diagram of a small perturbation of the inflammation zone in the detonation KLK (considerably exaggerated on the figure). Below the sections indicate the decomposition of the perturbation perpendicular to the motion of the detonation.

distorted by the perturbation KLK which has been strongly exaggerated in Fig. 15 for purposes of illustration. Actually of course the initial perturbation of the plane front must be infinitesimally small. The distortion KLK can appear, for example, as the result of a small inhomogeneity in the composition of the mixture leading to a decrease in the period of induction of inflammation in the zone L and to its increase in the region K. Above the line BKLKB the gas is compressed to a pressure of p_C, below it to p_B (cf. Fig. 12). In the plane wave the pressure drop from p_C to p_B, as shown in Fig. 12, is retained for an arbitrarily long time: it satisfies the conservation laws and the condition of the equality of the velocities of the shock wave front and of the combustion zone. However in a nonplanar wave the pressure jump is unstable in the transverse direction, right and left in Fig. 15. It is not in equilibrium and disintegrates, as is shown in the cross sections below the BKLKB line. The pressure in the K region decreases, whereas in the L region it increases. This is a most important detail and we will therefore repeat this statement again in other words. The pressure drop from p_C to p_B along the x axis is stable. However, the perturbation KLK produces unstable pressure breaks (jumps) perpendicular to the direction of motion of the detonation front. Nothing prevents these jumps from disintegrating. Moreover, they must disintegrate like an arbitrary pressure discontinuity (differential) produced in the gas by any method.

Thus in the zones L the pressure of the reaction products is increased, the detonation in the region L turns out to be overdriven, and the Jouguet condition is violated. In front, from the zone L to the shock front CC, a compression wave propagates whose induction time decreases in this region, the gas is inflamed more quickly and the point L approaches the front CC. In the KK zones the pressure decreases as a result of expansion, the gas is cooled, the induction period is increased, and the points KK move away even further from the shock front CC. In all, the disturbance KLK, having appeared, is strengthened. This indeed constitutes the instability of the complex consisting of a shock wave and the planar inflammation zone that follows it.

Since the speed of sound is always larger in the gas compressed by the shock wave than the velocity of the front with respect to the compressed gas, perturbations of the combustion zone give rise to perturbation of the shock front CC. The shock wave (one readily sees) is unstable when the perturbations of the combustion zone are of a dimension on the order of or larger than the width of the inflammation zone (λ in Fig. 12). Perturbations of smaller extension than the zone width are averaged (the pressure in them is equalized) before they reach the shock front.

The described qualitative mechanism of instability allows one to find a quantitative criterion for its appearance. The activation energy E of the inflammation reaction will enter into it. Clearly, the more the period of the inflammation reaction depends on the temperature (the higher the activation energy), the less stable the detonation. The changes in the pressure in the expansion and compression zones (K and L) which lead respectively to cooling and heating of the unburnt gas have the larger an effect on the chemical reaction time (the position of the points K and L in Fig. 15), the stronger the reaction rate depends on the temperature.

For a derivation of the criterion it is essential to allow for one more circumstance. The pressure in the zone K, of dimension λ, is equalized (the zone relaxes) in a time on the order of $\lambda/2c_2$ (here c_2 is the speed of sound in the shock-compressed gas of the zone K). The time during which the shock-compressed gas remains in zone K of dimension λ is of the order of

$$\frac{\lambda}{D-W}.$$

But in the shock wave $c_{com} > D - W$ always, so that the inequality $2c_2 > D - W$ is even more correct in it; therefore,

$$\frac{\lambda}{2c_2} < \frac{\lambda}{D-W}.$$

The time of the equalization of the pressure in the transverse direction is always less than the time during which the perturbation remains in the detonation front (less than the lifetime of the perturbation). For this reason the period of the induction always "has time" to take on a value corresponding to the pressure which has decreased or increased as a result of the disintegration of the transverse discontinuity. More accurately, the induction period has time to assume a value corresponding to that temperature which will be established after the compression of the gas in the region L and its expansion in the region K. Therefore one does not have to consider the time evolution of the disturbance, and it is sufficient to consider only the change in the chemical reaction rate as a function of the temperature change caused by the compression or expansion of the gas in the initial perturbation.

As a result one can formulate the following quantitative criterion for the loss of stability; if the adiabatic expansion of the gas from the zone K into the region L will on decreasing the temperature of the gas increase the delay in the inflammation by a quantity of the order of the delay itself or larger, then an arbitrary initial perturbation (distortion) of the flame front will increase and the planar detonation will lose its stability. Neglecting the dependence of the reaction time on the pressure (density), we obtain the instability criterion of a planar detonation

$$\left.\frac{d\tau}{dT}\right|_{T_C}(T-T_C) \gtrsim \tau, \qquad (13)$$

where τ is the chemical reaction time, and T is the temperature of the unburnt gas in the perturbation zone after the expansion.

The time of the chemical reaction—a quantity inversely proportional to its rate—is proportional to

$$\tau \sim \exp\left(\frac{E}{RT}\right). \qquad (14)$$

From (13) and (14) we obtain the criterion

$$\frac{E}{RT_C}\left(1-\frac{T}{T_C}\right) \geqslant 1. \qquad (15)$$

Near the point K the gas, whose pressure until the perturbation was p_C, expands adiabatically up to a pressure p_B. Therefore (15) can be rewritten as

$$\frac{E}{RT_C}\left[1-\left(\frac{p_B}{p_C}\right)^{\frac{\gamma-1}{\gamma}}\right] \geqslant 1. \qquad (16)$$

True, estimates of criterion (16) for various mixtures are not very precise, because the activation energy E at high temperatures is not known exactly, but they indicate an instability of the planar detonation practically in all gas mixtures in which it is observed.

The physical analysis carried out above gave rise to somewhat more thorough mathematical investigations of instability of planar detonation (R. M. Zaĭdel',[27] J. J. Erpenbeck,[28] and Pukhnachev[29]). The instability criterion (17) (see below) obtained by R. M. Zaĭdel' differs from (16). Nonetheless, in his work as in that of others instability of planar detonation is observed: in the main the results coincided with the results of the physical analysis.

Zaĭdel' and Pukhnachev arrive among others at the conclusion that detonation is stable with respect to perturbations whose wavelength is small compared with the width of the reaction zone. This type of stable detonation can, as they assume, take place in narrow tubes whose diameter is considerably smaller than the width of the reaction zone. The occurence of stability is clear from the above. Perturbations on the flame front are not imparted to the shock front: the pressure in the waves traveling away from them is averaged out over the cross section of the tube before it reaches the shock front.

5. ONE-DIMENSIONAL INSTABILITY OF DETONATION

Detonation in narrow tubes, stable with respect to distortions of the combustion zone, turns out to be unstable with respect to one-dimensional perturbations, as R. M. Zaĭdel' and Ya. B. Zel'dovich have shown.[30]

Detonation in the Zel'dovich-Neumann model is clearly a Jouguet deflagration propagating in the shock-compressed gas—the shock wave (Fig. 16a). In the steady-state regime the compressed gas is inflamed after a time τ after entering the shock front. This time corresponds to the velocity of propagation of combustion, equal to the flow velocity of unburnt gas, and is opposite to it in sign: the combustion front B remains stationary with respect to the shock-compression front AC. Let now the chemical reaction time be randomly shortened for any reason by some

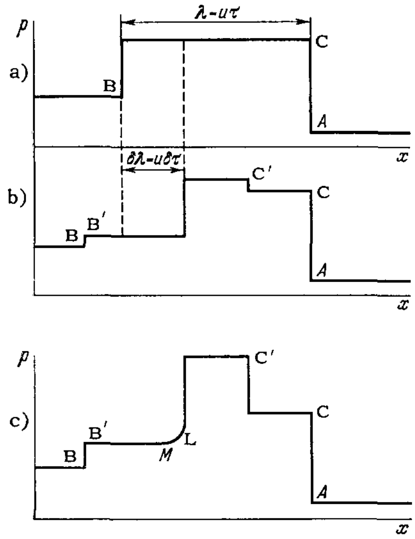


FIG. 16. The appearance of two shock waves C′ and B′ on shortening the period of the inflammation delay on the detonation front by $\delta\tau$.

small amount. The flame front will now move toward the shock front, and a velocity component of the flame will now appear, directed toward the forward boundary of the shock wave. The conditions on the "unburnt gas—combustion products" boundary will cease satisfying the Jouguet condition. The new discontinuity will fall apart into two shock waves, one (C′) will travel in the shock-compressed gas and the other (B′) in the combustion products (Fig. 16b). If the increase in velocity is relatively large, then a rarefaction wave LM will appear in the considered instance (Fig. 16c). However, as long as the perturbation is small, the wave LM can be neglected.

The complex consisting of a combustion zone and a shock wave propagating in front of it is called a double nonstationary discontinuity. The wave velocity, the velocity of the gas, and its state in all zones of double nonstationary discontinuity can be readily calculated.[10]

The shock wave C′ which appeared as a result of the decrease of the chemical reaction time heats the gas additionally; this results in further decrease of the chemical reaction time, a new shock wave C′ travels in the gas which decreases the chemical reaction time even more, and so on. The perturbation (acceleration of the combustion) having appeared, will progressively increase. When the perturbation reaches the initial shock front AC, a rarefaction wave is propagated back in the compressed gas, the pressure on the front drops sharply, and the detonation may be attenuated as a result of the very strong dependence of the reaction time on the temperature (pressure of the shock-compressed gas).

The authors of [30] obtain for one-dimensional perturbations an instability criterion of detonation which coincides with the condition derived by Zaĭdel'[27] for loss of stability of a planar detonation with respect to distortions of the combustion front

$$\frac{E}{RT_B} > \frac{(\varkappa+3)(\varkappa+1+\sqrt{\varkappa+1})}{2(\varkappa+2)}. \tag{17}$$

Here T is the temperature of the compressed, unburnt gas (in zone C of Fig. 16a), $\kappa = (\gamma + 1)/(\gamma - 1)$, and γ is the ratio of the heat capacities.

Criterion (17) suffers from the deficiency that it depends neither on the ratio of the speed of sound in the burnt and unburnt gas, nor on the thermal effect of the combustion, on which, all other conditions being equal, the intensity of the shock wave traveling in the unburnt gas depends.

In [31] a different, quantitative solution, presented below, is obtained for the one-dimensional instability problem.

In a Chapman-Jouguet detonation the inflammation zone is at rest with respect to the shock front. Therefore the flame velocity is equal and opposite in sign to the flow velocity of the unburnt gas:

$$u = -u_{\text{com}}.$$

The distance from the shock wave to the inflammation zone is

$$\lambda = u\tau,$$

where τ is the inflammation delay.

In an unperturbed wave the volume of gas per unit of front surface burnt after an arbitrary period of time t, is

$$V = ut.$$

If the inflammation delay time is accidentally shortened in the time t by δt, then the flame front will approach the shock front by a distance of

$$\Delta\lambda = u\delta\tau$$

and the volume of gas burnt during this time will be

$$V' = ut + u\delta\tau.$$

The ratio of the gas volumes burnt during the same time intervals is equal to the ratio of the propagation velocities of the flame:

$$\frac{V'}{V} = \frac{u'}{u} = 1 + \frac{\delta\tau}{t}.$$

Since the time interval t is arbitrary, we will take it to be τ. Then, taking into account the signs, we can write

$$\Delta u = \frac{u'-u}{u} = -\frac{\delta\tau}{\tau}. \tag{18}$$

In writing (18), we must bear in mind that Δu is the dimensionless increase in the flame velocity averaged over the time interval τ. If in the following time interval τ the induction period will become shorter by a further $\delta\tau$, then the flame front will come closer to the shock front by an additional amount $u'\delta\tau$. In other words, in order for the new flame velocity u′ to be retained, the inflammation delay must become continuously shorter. If, on the other hand, it will, after being once shortened by $\delta\tau$, remain $\tau - \delta\tau$, then the combustion zone will settle at a new distance $\lambda - u\delta\tau$ from the shock front. The flame will again be at rest relative to the shock front.

The dimensionless pressure drop in the shock wave traveling in the unburnt gas and due to a small increase in the flame velocity will be determined in accordance with [10] (p. 221) by the expression

$$\Delta p = \frac{p'-p}{p} = \frac{c}{c+c_{com}} qM\Delta u. \quad (19)$$

Here p' is the pressure in the shock wave, p is the pressure in the gas, in which the shock wave is traveling, c and c_{com} are the respective speeds of sound in the unburnt and burnt gas, q is the ratio of the thermal effect of combustion to the internal energy of the unburnt gas, and M is the ratio of the velocity of the flame to the speed of sound in the unburnt gas.

The subsequent fate of an initial perturbation (in the time $\delta\tau$ and length $u\delta\tau$) depends on the reverse effect of the shock wave (19) upon the inflammation delay period, and correspondingly on the flame velocity given by expression (18). If the shock wave will in a period no longer than τ shorten the delay by a quantity exceeding $\delta\tau$, then the initial perturbation will grow. If, on the other hand, the shock wave shortens the inflammation delay by less than $\delta\tau$, then the initial perturbation will decrease. Consequently, to explain the condition for growth of an initial perturbation, one must find the dependence of the change of the inflammation delay (the relative change in the flame velocity) on the relative pressure drop in the shock wave.

The chemical reaction time depends on the temperature as

$$\tau \approx A \exp\left(\frac{E}{RT}\right). \quad (20)$$

Therefore,

$$\tau + \delta\tau = A \exp\left\{\frac{T}{R(T+\delta T)}\right\},$$

or

$$\frac{\delta\tau}{\tau} = -\frac{E\delta T}{RT^2}. \quad (21)$$

Assuming an adiabatic compression in the shock wave, we obtain

$$-\frac{\delta\tau}{\tau} = \Delta u = \frac{E}{RT}\left[1 - \left(\frac{p'}{p}\right)^{\frac{\gamma-1}{\gamma}}\right]. \quad (22)$$

Expanding (22) in a series and confining oneself to the linear term, we find

$$\Delta u = \frac{\gamma-1}{\gamma}\frac{E}{RT}\frac{\delta p}{p} = \frac{\gamma-1}{\gamma}\frac{E}{RT}\Delta p. \quad (23)$$

Making use of the instability condition [[10], p. 229, formula (31.2)]

$$\left.\frac{d\Delta p}{d\Delta u}\right|_{(19)} > \left.\frac{d\Delta p}{d\Delta u}\right|_{(23)},$$

we arrive at the criterion for the incipience of a one-dimensional detonation instability in the form

$$\frac{\gamma-1}{\gamma}\frac{E}{RT_B}\frac{1}{1+\frac{c_{fl}}{c}} qM > 1. \quad (24)$$

The criterion (24) is practically always larger than unity. A plane wave is therefore also unstable with respect to one-dimensional perturbations. If it can be observed, then only in special cases when the activation energy at high temperatures will turn out to be so small that criteria (16) and (24) will become less than unity [the inequalities (16) and (24) will not be fulfilled]. So far it is impossible to say whether such cases occur in reality.

In homogeneous gas mixtures detonation therefore does not propagate as a plane wave, but becomes either a pulsating or a spinning detonation. In this connection the old approach to the detonation limits, in which the attenuation of a planar detonation is considered loses its validity. The propagation limits of a detonation are now determined, as will be seen below, by the conditions for the propagation of detonation spin.

6. PULSATING DETONATION

Inhomogeneities in the detonation front, which was previously considered planar, first observed by Yu. N. Denisov and Ya. K. Troshin [24,25] by a so-called trace method and also by B. V. Voĭtsekhovskiĭ, B. E. Kotov, V. V. Mitrofanov, and I. E. Topchiyan [23] by a photographic method, have been investigated in detail in Soviet and foreign laboratories. [33,34,35,39,10,23] Of the foreign investigations a particularly interesting one is that of D. White [39] who studied the structure of a detonation in the mixture $2H_2 + O_2 + 2CO$ by an interference method. White invariably observed a strong turbulence behind the reaction zone. This is an additional convincing and independent proof of the three-dimensional structure of the detonation front. Pulsating detonation, as the detonation with numerous inhomogeneities in its front was called, can now be considered an independent form of combustion: it is a plane wave in the Zel'dovich-Neumann model, which has lost its stability. Spinning detonation turned out to be a limiting case of pulsating detonation, appearing when there is only one inhomogeneity in the tube cross section.

The structure of inhomogeneities in a pulsating detonation, as the experiments and gasdynamic calculations compared with them show, is the following. [10] From the overdriven portions (L, Fig. 15) compression waves travel to the shock front and give rise on its surface to two types of disturbances: a break—the intersection of two shock waves (Fig. 17a), and the intersection of a shock wave with an inclined detonation wave (Fig. 17b). Simplified diagrams of the corresponding breaks, the well-known triple Mach configurations which are amenable to exact gasdynamic cal-

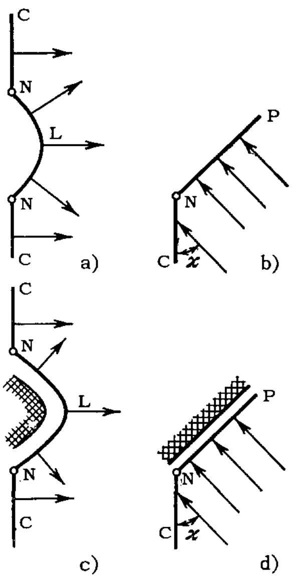


FIG. 17. Diagram of the deflection of the shock front of a detonation: a) – b) without combustion directly after the deflection; c) – d) with combustion.

culation, are shown on the right-hand side of Fig. 17. The line NP shows the "branches" or "Mach discs" of triple configurations. The angle κ differs appreciably for the configurations of both types. In the cases of intersection of a shock wave with a detonation wave (Fig. 17) it is appreciably smaller than in the intersection of two shock waves. The motion of the point N of Fig. 17 can be followed from the imprints on the side of the tube (Fig. 20) and one can thus measure the angle κ. Measurement and comparison with calculations of configurations of both types show that in the majority of cases perturbations of the type of 17a appear in pulsating detonation. There are many of them on the front. Colliding, they produce zones of increased pressure and temperature in which the gas is influenced (the diffuse zones at the points of intersection of the lines on Fig. 20).

Impinging on the plate perpendicular to the motion of the front (Fig. 18), the configurations leave on it imprints, one of which with the largest inhomogeneities is shown in Fig. 19. It is very curious that the mean

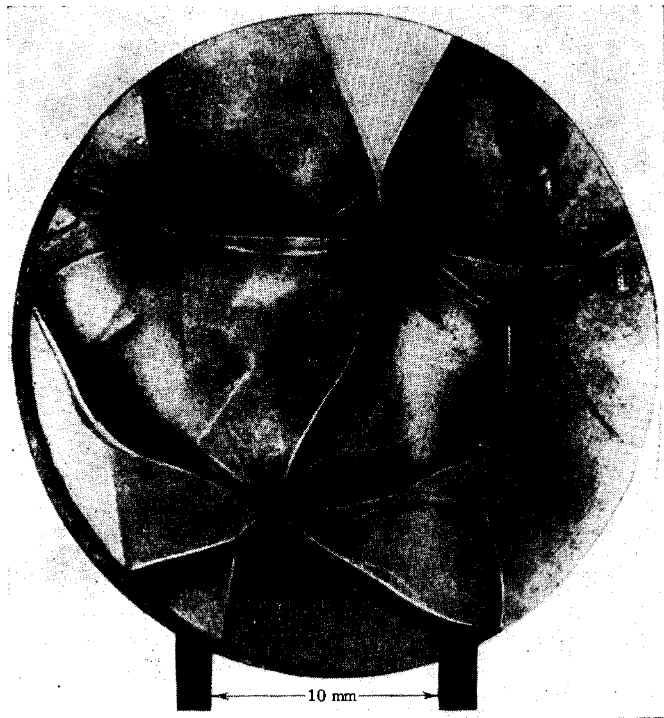


FIG. 19. Trace of the collision of a pulsating detonation with the end of a tube. The lines indicate the places where triple shock configurations met.

dimension of the cells similar to those shown in Fig. 19 is proportional to the chemical reaction time in the Zel'dovich-Neumann model. The model leaves a peculiar reminder of itself, and this is no accident.

Disregarding the instability, the existence of the pulsating detonation is based on this model. The instability does not destroy a detonation with a shock wave in front of the combustion zone, it only imparts to the wave a more complex three-dimensional structure.

Figure 21 shows two additional imprints of the pulsating detonation on a plate placed perpendicular to the motion of the wave front. These photographs, as those on Figs. 19 and 20, were taken by Ya. K. Troshin and Yu. N. Denisov with the aid of a simple, but at the same time sensitive, technique. To obtain clear imprints, the plates were covered by a special technique with a thin layer of soot.

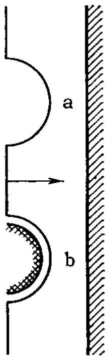
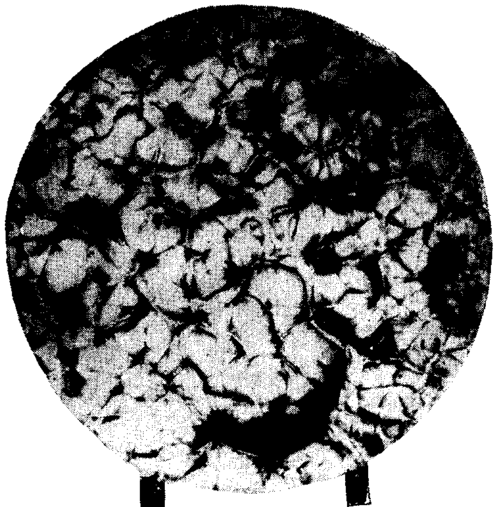
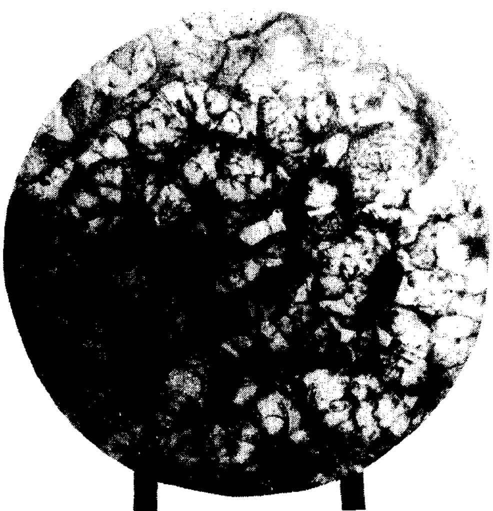


FIG. 18. Diagram of the collision of a detonation front with a solid wall.

FIG. 20. Imprint left by a pulsating detonation on the lateral surface of the tube.



a)



b)

FIG. 21. Imprints of a pulsating detonation incident on a sooty plate. a) A mixture of $2H_2 + O_2$, initial pressure 300 mm Hg; b) a mixture of $CH_4 + 2O_2$, pressure 800 mm Hg. Small-scale inhomogeneities (fine structure) are visible within some large-scale inhomogeneities.

The mean dimension of the inhomogeneities decreases with increasing initial pressure of the mixture. The method made it possible to record inhomogeneities down to 0.1 mm in size. The decrease in the dimension is accompanied by a certain increase in the velocity of detonation with increased pressure, leading to increased temperature of the shock-compressed gas and a decrease in the chemical reaction time.

Figure 22 shows the change in the detonation velocity and temperature of the shock-compressed gas in a $2H_2 + O_2$ mixture as a function of the initial pressure.

The dependence of the mean dimension of the inhomogeneity Δy on the chemical reaction time is expressed by a relation which is apparently correct up to a constant coefficient of the order of unity [34]:

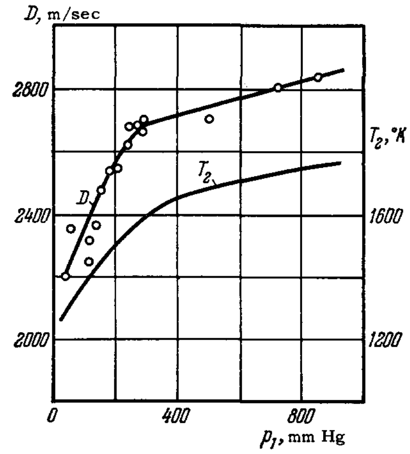


FIG. 22. Dependence of the detonation velocity and of the temperature T behind the shock front on the initial pressure of the $2H_2 + O_2$ mixture (tube diameter 16 mm).

$$\Delta y \approx \frac{(\gamma-1)\cdot 2\tau D}{(\gamma+1)\left(1-\sqrt{\frac{\gamma-1}{\gamma+1}}\right)} = \beta\tau D. \tag{25}$$

Using (25), one can determine the chemical reaction time in the Zel'dovich-Neumann model experimentally. This method was used in [36] to measure the temperature coefficient (the apparent activation energy) of the inflammation reaction of benzene with oxygen.

The activation energy in the temperature range shown in Fig. 23 turned out to be 37 kcal/mole.

Figure 21b shows an inhomogeneity structure more complex than Fig. 21a. Fine inhomogeneities, more numerous by an order of magnitude, fill some of the large cells. One might think that there appear in some instances on the detonation front inhomogeneities similar in structure to those of Fig. 18b: an intersection of a shock-wave with an inclined overdriven detonation takes place. The overdriven detonation existing within

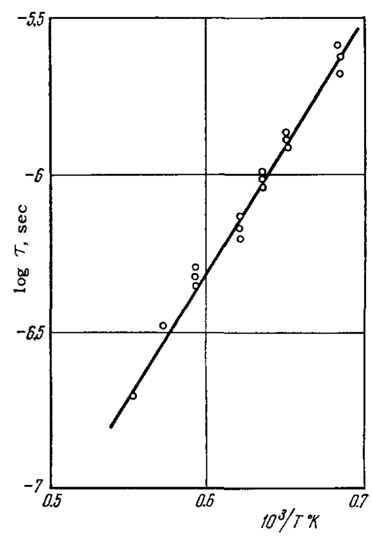


FIG. 23. Temperature dependence of the inflammation delay of a benzene-oxygen mixture in a detonation wave. The apparent activation energy under these conditions is 37 kcal/mole.

one large-scale cell may turn out to be unstable, regardless of the fact that the left-hand side of criterion (16) is always smaller for an overdriven detonation than for a Jouguet detonation.

In the Zel'dovich-Neumann model an overdriven detonation for an initial state A corresponds to (is identical with) a weak deflagration for the initial state c_1 (Fig. 10). Therefore, for an estimate of the instability limits of an overdriven detonation one can use the approximate criterion obtained for weak deflagration [10]:

$$(\gamma-1)^2 \frac{E}{R_T} \frac{Q}{c^2} M^2 > 1. \tag{26}$$

Here M is the ratio of the "propagation" velocity of the combustion with respect to the shock front to the speed of sound in the compressed gas. The reader is already familiar with the remaining quantities.

As a result of the higher temperature in the shock front of an overdriven detonation compared with a Jouguet detonation, the dimension of the cells on loss of stability turns out to be considerably smaller than in Jouguet detonations. Thus the fine structure appears.

A. N. Dremin, G. A. Adadurov, and O. K. Rozanov [37,38] observed an inhomogeneous structure in the detonation front in nitromethane (liquid) diluted with acetone. No pulsating detonation has been recorded in solid explosives. The inhomogeneities in the structure of explosives are practically always larger than those connected with the instability.

7. DETONATION LIMITS

Instability, however paradoxical this is, extends the possibilities of detonation propagation. In inclined waves (in the Mach "branch" NP, Fig. 17) the temperature is considerably higher in the direct shock wave CN. At the collision point of two inclined shock waves (of two Mach discs) the temperature is even higher. Such an instability, giving rise to perturbations of the shock front, produces on a relatively small area of the front points of particularly high temperature. Because of the exponential temperature dependence of the reaction time the mixture is inflamed in these considerably more rapidly than in the plane wave. From these hot points the combustion is then propagated over the entire tube cross section. Without hot points the inflammation at the detonation boundaries would occur very far behind the shock front, the losses to heat transfer and friction would lead to the attenuation of the detonation considerably sooner (for example, at a higher pressure, in a composition of higher fuel value on in a more rapidly burning composition) than in the presence of inhomogeneities.

In Tables I and II, made up of data from [10] (Sec. 4), we present the calculated pressure and temperature (dimensionless, relative to the initial gas) in the various zones of the triple Mach configuration (Fig. 24a and 24b). In zones 2 and 3 in each of the configurations the gas is compressed but not inflamed. Zone 4 is filled with combustion products. The temperature of the unburnt gas is always higher in zone 3; naturally, it is in this zone that the inflammation of the gas takes

Table I. Calculated pressure, density, and temperature in different zones of the triple Mach configuration in the case of detonation of a mixture of $2H_2 + O_2$, D = 2800 m/sec, p_1 = 760 mm Hg, γ = 1.4

Parameters	Zones on Fig. 21a			
	1	2	3	4
p/p_1	1	34.4	48.7	48.7
ρ/ρ_1	1	5.113	5.36	6.57
T/T_1	1	6.7	9.1	4.7

Table II. Calculated pressure, density, and temperature in a triple Mach configuration with an inclined detonation wave in the case of spin detonation in a mixture of $2H_2 + O_2$, D = 2200 m/sec, p_1 = 45 mm Hg

Parameters	Zones on Fig. 21b				
	1	2	3	4	5
p/p_1	1	20.2	37.8	34.3	34.3
ρ/ρ_1	1	4.7	5.2	3.2	6.6
T/T_1	1	4.5	7.3	10.6	5.2

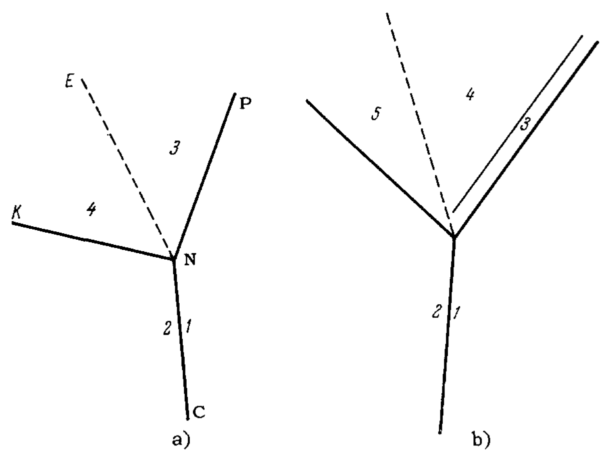


FIG. 24. Triple Mach configuration (break) in the case: a) of the intersection of two shock waves CN and NP, b) of the intersection of a shock wave with a detonation.

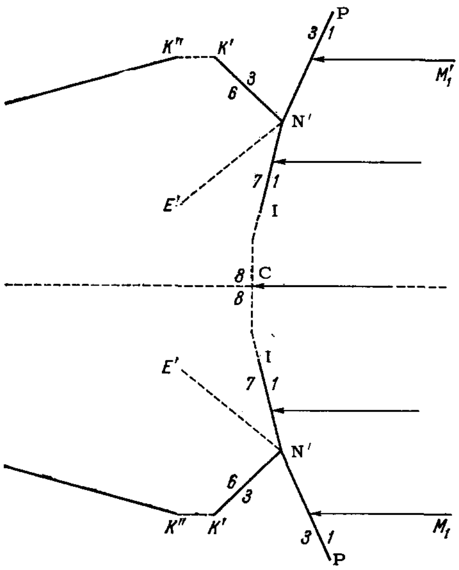


FIG. 25. The result of the collision of two shock configurations – two waves of the type NP (Fig. 24, a).

place. However, if in zone 3 the gas is inflamed sluggishly (the inflammation time is less than the time up to the collision of the configurations), two shock configurations collide. The reflection of the two NP waves from each other (Fig. 25) turns out to be irregular (Mach reflection). The calculated temperature in the various zones of the new configuration **resulting** from the collision is given in Table III. Two Mach configurations are taken as the initial configuration, one of which is presented in Table I. In the collision zone 8 and in the neighboring region 7 an even higher temperature is attained than in the inclined shock wave 3. The collision zones serve as good sources of ignition.

In view of the extremely strong temperature dependence of the chemical reaction time, the condition of the existence of triple configurations, assuming a rapid inflammation of the gas, coincides with the condition for the propagation of detonation.[10,34] Such an approach to the determination of the limits of existence of detonation differs radically from that which prevailed previously.[32]

The number of perturbations on the surface of a detonation front in a tube of diameter d with account of (25) turns out to be of the order of

$$\left(\frac{d}{\Delta y}\right)^2=\frac{1}{\beta^2}\left(\frac{d}{\tau D}\right)^2.$$

Hence the condition for the existence of one (spinning) perturbation can be written in the form

$$\frac{1}{\beta^2}\left(\frac{d}{\tau D}\right)^2=1, \quad (27)$$

and the condition for the attenuation of a detonation is

$$\frac{1}{\beta^2}\left(\frac{d}{\tau D}\right)^2<1. \quad (28)$$

Table III. Pressure, density, and temperature in the configuration of Fig. 25 resulting from the collision of two configurations of the type of Fig. 24a

Parameters	Zones in Fig. 25: 1	3	6	7	8
p/p_1	1	48.7	54.8	54.8	59
ϱ/ϱ_1	1	5.4	5.8	5.4	5.5
T/T_1	1	9.1	9.42	10.1	10.8

The chemical reaction time increases very rapidly on decreasing the detonation velocity. To the extent of the enrichment or depletion of the mixture of the burning component—the approach to the limit of the detonation propagation—the wave velocity decreases, the product τD increases, and the left-hand side of the criterion (28) decreases rapidly. The left-hand side of (28) decreases also as one approaches the limit because of a decrease in the pressure of the mixture or a decrease in the tube diameter. In this case losses to friction and heat transfer enter in the criterion implicitly. They decrease the wave velocity and thereby sharply increase the chemical reaction time.

The calculation of criterion (28) both from kinetic and thermochemical data is so far very unreliable: no accurate data are available on the kinetics of chemical reaction under detonation conditions, and the theoretical dependence of the detonation velocity on the initial pressure of the mixture and the tube diameter is unknown. Should this become necessary, the detonation limits are better determined from experiment. Relations (27) and (28), as is seen from the above, are only physically illustrative in nature.

To conclude, we return to the determination of the detonation instability. The instability was seen to be related to the strong (exponential) temperature dependence of the rate of the chemical reaction of the inflammation of the gas compressed and heated by the shock wave which precedes the inflammation zone. The shock wave was introduced by Ya. B. Zel'dovich in proposing the model of a detonation wave (which has come to be called the Zel'dovich-Neumann detonation) as the essential condition for the propagation of a strong and Chapman-Jouguet detonation. The instability leading to the appearance of a pulsating (and in the limit a spinning) detonation excludes the propagation of a planar shock wave before the combustion front. But a shock wave preceding the combustion zone—now in the form of quite complex three-dimensional intersecting and colliding shock configurations—has nevertheless remained a necessary condition for the propagation of a detonation (a strong or a Chapman-Jouguet detonation). In science fruitful ideas are often retained in some form, even under new conditions. Thus it was here, too. The idea of a shock wave inflaming the gas remained, although it underwent considerable change. The instability of the detonation is now considered a result of the inflammation of the gas in the shock wave. If there were no shock wave, there would be no instability!

It remains to say a few words about weak detonation. Its propagation is possible only in the presence of an external inflammation source. Inflammation in the shock wave before the combustion zone, as well as the appearance of the shock wave itself, makes the propagation of the weak wave impossible. There is consequently in weak detonation no fundamental cause which determines the instability. There is no self-inflammation of the gas with a sharp temperature dependence of the inflammation time. The weak detonation is for this reason always stable, at least against perturbations against which Chapman-Jouguet and strong detonations are unstable in the cases considered above.

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Translated by Z. Barnea