On the peculiar properties of initiation and propagation of multifront detonation

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Abstract. At initiation an external source must initiate a chemical reaction in the combustible mixture and ensure its self-sustaining propagation over the whole volume of the mixture. Traditionally, the critical energy is considered as the measured value of successful initiation of a mixture, providing 100% excitation of the process – combustion or detonation. Actually, the initiation of combustion or detonation process has a “threshold” nature (“yes” – “no”). The spatial–temporal law of initiator energy allocation has a noticeable effect on the critical initiation energy. The instability of the system of equations of gas dynamics and chemical kinetics is manifested in the multifront nature of the detonation front and the cellular structure of the trajectories of the transverse waves. The important question is the moment and place of occurrence of such a structure, especially for expanding waves. Along with the kinetic–gasdynamic instability of the combustible mixture, a certain role is played by the instabilities introduced by the initiator: for example, the constrictive (sausage) electromagnetic instability of an electric conductor with a current, the Meshkov–Richtmeier instability at the expanding boundary of the liquid metal of the exploding wire, etc. In this report the existing problems of initiation and propagation of detonation are discussed based on modern data, including the results of the author's research on these issues.

1. Introduction
It is generally accepted that the meta-stability of the combustible mixture in the initial state is due to the presence of a potential barrier with a height equal to the activation energy. The chemical transformation of the initial mixture into reaction products is carried out under the condition that the certain amount of particles (“sufficient” for the subsequent development of a self–sustaining chemical reaction) overcomes this barrier.

The energy from an external source supplied to these particles to overcome the potential barrier height is the energy of initiation of the combustible mixture. The initiator passes to mixture the relay race of the reaction front propagation, it is important that the reaction does not die out. The “sufficient” value determined by the number of effective collisions of the particles of the mixture, even at present, is estimated to the accuracy of the order. Therefore, the calculation of the critical parameters of the mixture initiation is still a very complex problem.

The question of the critical ignition energies of the mixture (laminar or turbulent subsonic combustion) and initiation of detonation (supersonic propagation) is crucial not only from the point of view of scientific knowledge, but is also of great practical importance for assessing the explosion hazard of combustion systems. Problematic information about initiation and propagation of detonation wave (DW) are useful and for young investigators.
2. Methods of determination of the initiation energy

Traditionally the following initiators are used as external source for excitation in a reacting mixture of combustion and detonation waves: electric or laser spark (breakdown), exploding wire, explosive charge, flow of hot and active particles, thermal igniter, etc.

The effect of excitation of combustion or detonation processes in a combustible mixture is usually characterized by a “threshold” nature (“yes” – “no”) for any initiator – Figure 1. In the idealized model of a strong explosion for an inert medium (for example, [1–2]), the energy of the explosion is the determining parameter on which the propagation of a blast wave depends. By analogy, and for a combustible mixture, the minimum initiator energy, which provides 100% excitation of the combustion or detonation regime, is commonly called as the critical energy. The critical ignition energy $E_{\text{ign}}$ (at least, with spark ignition) traditionally acts as the main parameter of the fire hazard of a mixture. For initiator with the ideal space-time characteristics the critical initiation energy of detonation $E^*$ serves as a measure of the detonation hazard of mixtures: the smaller $E^*$, the more dangerous the mixture.

![Figure 1. The threshold nature of the initiation of detonation - traces of the damped blast wave at $E<E^*$ (left) and the initiation of multi-front detonation at $E \geq E^*$ (right photo).](image)

The formulation of the initiation condition on the basis of a single parameter – the critical energy $E^*$ – is very attractive. However, the multi-front structure of a real detonation wave (instead of its idealized one-dimensional model with a smooth front) does not allow to consider the problem of initiating the combustion and detonation waves completely solved because of questions about the influence of the spatial and temporal characteristics of the initiator on the excitation of the combustible mixture and the formation of self-sustaining waves (burning or detonation).

Indeed, each mixture under given conditions (pressure, temperature, composition) can be characterized by some characteristic spatial and temporal scales (for example, the sizes of the induction zone or reaction zone $r^*$ and the corresponding induction or reaction times $t^*$). At the same time, under these conditions, the explosive mixture absorbs the certain amount of energy $E_\nu$ from the initiator during a finite period of time $t_0$ in the finite space area $V_0$:

$$E_\nu = \int_{V_0} \int_{t_0} \varepsilon(t, V) \cdot dt \cdot dV = \eta E_0$$  \hspace{2cm} (1)

$\varepsilon(r,t)$ – the function describing the space-time law of the input energy, $\nu$ – the dimension of the task ($\nu=1,2,3$ for flat, cylindrical and spherical symmetry, respectively). It must be mentioned, that $E_\nu$ is only part $\eta$ of the energy $E_0$ originally stored in the initiator. For various initiators the function $\varepsilon(r,t)$ has its own specific form, and in the general case it is a complex function of the characteristic scales of the initiator and the mixture (more precisely, of their ratio).

For example, from electrodynamics (for example, [3]), the equation $q=q_0 \cdot e^{-\delta t} \cdot \cos(\omega t+\varphi_0)$ of damped oscillations for RLC-circuit is well known for case of discharge of capacitor charged previously up to voltage $U_0=q_0/C$ - Figure 2. Here $\omega_0=1/LC$ is the fundamental frequency of oscillation, $\delta=0.5R/L$ –
the attenuation coefficient, and the frequency of damped oscillations is determined by the next formula \( \omega^2 = \omega_0^2 - \delta^2 \) (with \( \omega^2 > 0 \)). It should be emphasized that this solution is valid only in the case of the constancy of the parameters of all elements of the circuit. If we consider that the reactive and inductive resistances change due to heating at discharge, then the task becomes much more complicated.

If an exploding wire is used as an initiator instead of an electrical discharge, then in addition to the change in the resistance of the wire and heating when a pulsed current is passed through it, the phase state of the wire should be taken into account when the melting temperature is reached. From solid state the wire turns into a liquid state, a continuous medium turns as a result into a drip cloud with a sharp decrease in conductivity. This leads to a fundamental change in the discharge characteristics – Figure 3: O corresponds to the beginning of the initiating pulse, A – wire melting, B – liquid metal dispersion, CD is current pause with minimum value, D is discharge by pairs metal.

The influence of the temporal and spatial factors of input and transfer of energy from the initiator to the combustible mixture and the characteristics of the excitation and development of a chemical reaction in a mixture are extremely important in the tasks of initiation and, above all, for the correct experimental determination of the critical energy and also its optimization.

Many characteristic spatial and temporal parameters are required to describe the gas-dynamic and kinetic features of the initiation of combustible mixtures. Because of their uncertainty the problem of initiating combustion and detonation is solved now only with sufficiently strong simplifications.

3. Typical instabilities
It should be noted that only solitary articles can be founded where such aspects take into account as the change in resistance of the exploding wire at passing a pulsed current, the development of instability on the expanding boundary of the molten metal and its crushing into droplets, the electromagnetic instability of a plasma cloud, the formation of a blast wave… In general, non-reactive (inert) media were analyzed.

In the case of reacting mixtures, the instability of the gas-dynamic complex from the head shock wave (SW) and the following flame front is added, theoretically analyzed in the works of many researchers (for example, [4]). When a subsonic combustion mode is ignited, it is necessary to take into account the turbulization of the combustion front and the mixture itself, which requires additional characteristic parameters of the turbulent flow...
Some of the above mentioned processes can be seen in Fig. 4, where the schlieren-frames of the explosion dynamics of a wire are shown for case when high-voltage pulse from a charged capacitor fed to wire. One can see the development of instabilities of different nature (electromagnetic, hydrodynamic, ...), the initial stage of the formation of a blast wave.

4. Features of wave propagation during initiation
From Figure 4 it can be seen that at the initial stage the shock front practically indistinguishable with the boundary of the plasma cloud, and the separation of the shock wave (SW) from the plasma is fixed in subsequent frames (SW - light line).

Figure 5a shows a streak-record photo of the formation of a blast wave in an inert medium: the boundary of the expanding plasma cloud at the initial stage, its expansion to the maximum size and the subsequent attenuation of the luminous zone. Reduction of the plasma cloud is clearly visible. Such dynamics are typical at explosive processes: firstly the expansion of the plasma cloud, the stopping of the cloud boundary and the return movement to the point of initiation. A light oblique line on Figure 5a is the trajectory of a shock wave. SW-velocity in an inert medium gradually decreases with distance from the point of initiation.

Above, the attractiveness of using a single parameter — critical energy — as the main parameter of initiation was noted. But such an approach is justified under rather harsh conditions for the remaining parameters and, first of all, for the space-time characteristics of the initiator. For example, a strong explosion model assumes an instantaneous release of energy ($t_0=0$) in a region of zero size ($r_0=0$), while the explosion pressure is assumed to be so high that the initial pressure $P_0$ of the medium can be
neglected [1-2]. Figure 5b shows the conversion of the blast wave trajectory from Fig. 5a from the point of view of an idealized model of a strong cylindrical explosion, when the law of motion of the blast wave is determined by the instantaneous energy of the initiator. It can be seen that the experimental points correspond to the ideal model of a strong explosion (inclined straight line Y=A+BX, but not passing through the origin of coordinates) only when the “delay” in the time of energy release by the initiator is taken into account - X=-A/B.

Graphs with a similar behavior are also characteristic of the case of spherical symmetry (point initiation of a spherical blast wave), as well as for initiators in the form of a laser spark and explosive charge. This indicates that at the initial stage of initiation almost all commonly used initiators do not satisfy the conditions of instantaneous release of their energy. In addition, different types of instability break up the ideal one-dimensional form of a blast wave (plane, filament and point for a plane, cylindrical, and spherical symmetry case).

It can be noted that the recalculation of experimental trajectories from the point of view of a strong explosion model (see Figure 5b) makes it relatively easy to determine the useful energy of a source (exciting a given explosion wave) by the slope of the straight line constructed in the corresponding coordinates: t'\textsuperscript{2}=f(t\textsuperscript{2}).

If the same source with already known energy is used to initiate a combustible mixture, then the critical energy for detonation initiation in the mixture can be determined.

Figure 6 shows a photo scan of initiation of detonation by an exploding wire in a combustible mixture, which is analogous to the inert mixture in Figure 5a. In this case, the initiator transmits the “relay” to the energy release of the mixture, and the mixture either “picks up” the further propagation of the blast wave (at E≥E*) and accelerates it to the detonation wave (DW), or because of the inconsistency of the energy release, the blast wave from the source gradually falls out (as in Figure 1a). The dependences of the velocity of the blast wave as it moves away from the wire are shown in Fig. 7: the shock wave (SW) decays when the energies initiator and mixture are incoordinated.
Figure 7. Dependences of the wave velocity on its radius for cases of DW—initiation by initiators of various energies.

Figure 9. The number of disturbances on the wave front with distance from the initiator at initiating a multifrontal DW.

(E<E*) – dashed line, or the energy release of the mixture “pushes” the damped blast wave and puts it into self-sustaining detonation mode. This is clearly seen in Figure 6: in the expanding wave, local foci of reaction arise, from which secondary micro-explosions develop. The blast waves from such microexplosions overtake the damped wave and are replaced by the wave damping to a new acceleration.

Figure 8. The dependence of the energy release of the mixture on the degree of DW—overdriven.

Figure 10. Idealized scheme of gas-dynamic and kinetic processes during detonation initiation.

Due to the instability of the gas-dynamic complex from the shock wave and the chemical reaction zone, the question of the moment of generation of the cellular structure of a real detonation front (which can be seen in Figure 1) is extremely important. Since the shock wave is resistant to small disturbances, and the shock wave in the reacting mixture is unstable, it is logical to assume that the appearance of the structure is associated with the moment when the energy release of the mixture becomes a positive value. Under the assumption of the equilibrium state of detonation products, this corresponds to a wave velocity when the equilibrium adiabat of detonation products intersects with the shock adiabat of the initial mixture. In fig. 8 for some typical mixtures a plot of the energy release in
overdriven DWs on the overdriven degree is presented, values are dimensionless on the Chapman – Jouget detonation parameters.

It should be noted that the zero-value of the energy release of the combustible mixture with strong recompression does not guarantee the absence of cellular structures of detonation caused by the instability of the system of gas-dynamic and kinetic equations. Instability in the reacting system can be introduced by the initiator, as discussed above with the example of an exploding wire. When using a spark discharge between the electrodes, the plasma discharge channel almost never looks like an ideal straight line due to its streamer nature. With a laser breakdown, the plasma cloud is almost always different from the ideal sphere. Geometric nonideality is inherent for other initiators, especially in the nearest zone. All of the above said confirms that only with a noticeable distance from the initiation region and large times, the time and spatial characteristics of the initiator can be neglected and the self-similar solution of the blast wave in an inert environment can be used. It should be noted that a solution with an instantaneous reaction at the wave front and a constant wave propagation velocity is self-similar for a reacting mixture.

Figure 9 presents a graph of the number of disturbances on the expanding front of the photograph in Figure 1. It can be seen that a large number of small-scale disturbances occur near the initiator when the blast wave exits from recompression (the speed graph is shown in Fig. 7) - this area is marked with index I. With a distance, the number of disturbances at the front even decreases, only the disturbances size increases. But in the region marked by index II, due to the additional energy release of the combustible mixture, the wave is again compressed and an almost spontaneous increase in the number of new disturbances is observed. Figure 9 confirms the importance of wave overcompression in the mechanism of the occurrence of disturbances on a diverging detonation front.

5. Evaluations of the initiation energy

Despite the multiparameter and complexity of the initiation task, it is extremely important for practical purposes to be able to estimate the critical excitation energy of the combustion and detonation waves. Therefore, in many countries, approximate models of initiation have been developed and applied for assessing the explosion hazard of combustible systems. A detailed analysis of the existing models of initiation was published in [5].

Figure 10 presents the scheme of the “relay” process of energy transfer from the initiating source to the energy release of the combustible mixture. Region 1-2 at the origin of coordinates is the motion of a blast wave as a result of the initiator action, at this stage the energy release of the mixture is much less than the initiator energy, and the reaction front is almost inseparable from the shock front. But the non–steady character of the blast wave gradually increases the ignition delay and the zone between the shock wave and the reaction front increases — line 1 for the shock wave and line 2 for the particle 2 ’’ that crosses the shock wave at some time t_i and then moves with “waiting for self-ignition” when its induction time expires at moment t_i. Almost from the moment t_i until it flashes at the moment t* the mixture accumulates in the induction zone.

After the mixture ignites at the moment of the flash t*, the following scenarios are possible: a) the mixture only ignites and the front of the laminar flame tears along the unreacted flow in induction zone. Due to the low flame velocity compared to the flow speed, the process of SW–damping will continued – line 6; b) turbulent combustion is ignited - line 5 with the same as in a) scenario; c) at the moment t* a detonation wave (DW) is excited in the induction zone, the front of which quickly overtakes the shock wave. At burning the combustible mixture accumulated in the induction zone a large amount of energy is released. After overtaking the shock wave and entering into the unperturbed combustible mixture, this wave forms the main detonation wave, the propagation of which is provided by the energy release of the mixture - line 4 in Figure 10.

The analysis of different models and results was published in [5]. In this paper it is appropriate to present a graph of the calculated critical initiation energies of detonation for different mixtures depending on the molar concentration of fuel in the mixture - Figure11.
A comparison of calculated and experimental data demonstrates that only a few models can be used for practical assessments of the explosion hazard of combustible systems — see details in [5]. As an example, Fig.12 shows a comparison of calculated and experimental values of the critical mass of high explosive initiator for hydrogen-air mixtures (the maximum amount of experimental data compared to other fuels) from the molar concentration of hydrogen in the mixture.

Conclusions
The spatial-temporal characteristics of initiator are very important for correct determination of the critical conditions for initiation of combustion and detonation processes in chemically active mixtures. Almost all commonly used initiators do not satisfy the conditions of instantaneous release of their energy at the initial stage of initiation.

Different types of instability of an initiator stimulate the kinetic-gasdynamic instability of reactive system and the cellular structure of multifront detonation wave.

Recalculation of experimental trajectories of blast wave from the point of view of a strong explosion model makes it relatively easy to determine the useful energy of an initiator by the slope of the straight line constructed in the corresponding coordinates: \( r^{\nu+2} = f(t^2) \).

If the same source with already known energy is used to initiate a combustible mixture, then the critical energy for detonation initiation in the mixture can be determined.

In spite of some approximated models for prediction of mixture hazard are known for practical estimations, the modern theory of initiation of detonation wave must be constructed with taking into account the individual spatial-temporal characteristics of the initiator and his typical instabilities.

References
[1] Sedov L I 1987 *Methods of similarity and dimension in mechanics* (Moscow: Nauka)
[2] Korobeynikov V P 1973 *Problems of the theory of a point explosion in gases* (Moscow: Nauka)
[3] Sivukhin D V 1983 *General course of physics. V.III. Electricity* (Moscow: Nauka)
[4] Shchelkin K I and Troshin Y K 1963 *Gas dynamics of combustion* (Moscow: USSR Academy of Sciences)
[5] Vasiliev A A 2015 *Combustion, Explosion and Shock Waves* 51 9–30