Features of initiating the light-sensitive explosive composites for safe blasting of borehole charges in coal mines

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Abstract. The purpose of paper is to study physical and chemical patterns for starting detonation in the explosive charges by means of laser pulse radiation. Studies of the physical and chemical properties of the mechanism for stimulating the detonation of explosives by pulse radiation of an optical quantum generator have been carried out. Methodology of experimental and theoretical studies as well as mathematical modeling, involving gas-dynamics equations, has been applied. Basic research results as for studying sensitivity of the explosives being initiated by pulse light radiation have been analyzed. Numerical modeling was performed taking into consideration the real process of igniting the explosive by infrared laser radiation. The proposed mathematical model makes it possible to study the peculiarities of initiating the explosive transformation of bursting explosives by means of short light pulses. Tetranitropentaerytrite (PETN) was used to show that the process is determined completely by the parameters which characterize radiation intensity and absorption properties of the explosive. Depending on these parameters values, initiation processes may be implemented qualitatively - either on the surface or inside the explosive. In the latter case, the release of chemical energy results in the formation of so-called "chemical" pressure peak. With the use of lead azide, it has been shown experimentally that the initial temperature does not affect the increase in explosive sensitivity even in case when laser radiation takes place in the nanosecond pulse mode. Experimental results are applied while developing light-sensitive composites with the preset explosive and physical-chemical properties. The determined patterns were used in the development of the lightsensitive explosive composites for blasting agents of explosive charges.

1 Introduction

Currently, blasting is applied in terms of almost all coal and shale mines of Ukraine. Blasting energy helps recover about 10 - 15% of coal; 50 - 60% of mine workings are driven by means of blasting as well. In terms of shale mines, blasting operations are used to extract more than 60% of coal and to drive almost all the development workings.

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Considerable share of blasting operations is accounted for driving development and permanent mine workings as well as extraction of bituminous shale, which stipulates particular importance of the industrial safety rules in coal mines being dangerous in explosions of methane-air mixtures and coal dust.

According to the studies [1, 2], uncontrollable explosions during re-drilling or mechanical effect upon the unexploded borehole charges are one of the main causes of injuries and accidents in mines in the process of blasting operations. Within the period of 1976 – 2005, Injuries due to such explosions in Ukrainian mines accounted from from 30 to 45% of total injury rate recorded during blasting operations. It was determined that poor quality of electric detonators was the main reason of the unexploded borehole charges. Experts made the same conclusions while analyzing the results after studying the reasons of borehole charge burning and explosion of methane-air mixture, i.e. defective electric detonator results in the rise of temperature and pressure which effect is enough for uncontrollable explosion of the borehole explosive charge. 30% of the explosions of methane-air mixture is caused by the burning of borehole charges of the permissible explosive [2, 3].

Efficiency of any blasting operations depends mostly upon the system of the explosive initiation. First of all, that efficiency means ability of the system to provide high level of safety while preparing and performing blasting operations as well as absolute nonavailability of the reaction to the effect of external electromagnetic blasts. Initiation techniques including primary explosives have considerable effect upon the safety level. Thus, development of the primary explosives being sensitive to mechanical and thermal fields similarly to the secondary explosives (e.g. pentaerythritetetranitrate (TEN) is the most efficient and practically the only way to solve the problem.

Such issues as safe use of blasting agents, their reliability and operational precision as well as their environmental, technological, and economic efficiency are becoming more and more important in the process of wide-scale blasting of borehole or downhole explosive charges. There will be enough to mention organization of the operations during underground coal mining, open-pit iron-ore mining and building material extraction [4, 5]. It becomes clear that the solution of those problems is connected immediately with the solution of a key problem – development of the technology and synthesis of innovative priming explosives [6-8] which properties correspond to the requirements of modern technologies, and systems of their initiation [9].

Analysis of the results of experimental studies carried out for the recent 35 years in terms of safe and reliable operation of electric detonators has demonstrated that the problem is in the structure, the priming explosive and in the way of the priming explosive initiation. Substitution of the priming explosive for practically safe light-sensitive composite along with the use of laser initiation of the explosive charges is the most efficient solution of that problem.

Before the late 1980s, there were all prerequisites to develop maximally safe, reliable, and high-precision, economically and technologically efficient systems to initiate explosive charges by laser radiation. A series of techniques has been developed for shock initiation in the explosive charges with the help of zircon crystals, metallic foil and wool elements as well ascarbon nanotubes which explode under the effect of massive laser monopulse [10, 11]. However, that way turned to be not prospective to solve the problem of laser initiation in general. It was required to develop such primary light-sensitive high-energy compositions, which would allow applying laser systems during any blasting operations, and differ with high flexibility and ability to meet new requirements from the viewpoint of innovative technologies. In other words, there was a need in the innovative system of initiation with considerable physical and technical potential.

Owing to that fact, studies of regularities in the interaction of explosives and laser pulse radiation are one of the most topical tendencies in the field of chemistry and physics of explosion. In particular, analysis of optical properties of explosives is of high importance to understand a shock initiation mechanism. In this context, topicality of the understanding of initiation process physics as well as fundamental knowledge are important basis to develop new class of primary explosives characterized by abnormally high light-sensitivity.

2 Analysis of recent developments

Among the recent publications, attention should be paid to several results obtained while studying laser initiation of explosive mixtures with particles of metal and other inert materials [12 - 16]. In particular, paper [12] represents calculation of the dependence of absorption efficiency coefficients upon the sizes of aluminium nanoparticles in TEN matrix. Values of critical density of the energy of TEN laser initiation with aluminium have been measured experimentally. Paper [10] shows that in terms of "hexogen - nickel nanoparticles" mixture, minimum density of the initiation energy corresponds to the particles with the diameter of 40 - 45 nm. Authors of paper [14] have defined that the content of 0.1% Al additive in TEN reduces sensitivity to the shock comparing to the sample without such additive. That helps consider such high-energy mixed composition as a prospective material for optical detonators. One more paper worth examining deals with the explosive composites in which explosives with added nanodiamonds of detonation synthesis are used as the filling material for a polymer matrix [10]. For the recent 5 years, a number of theoretical papers have been written where the authors propose following ideas: applying calculations to evaluate parameters of the interaction of laser radiation and explosives; calculating critical densities of ignition energy with the use of simulation modeling; predicting possibilities in principle to control the threshold of energy materials initiation by adding light diffusors into the explosive etc. [15]. However, such a way to study properties and characteristics of primary explosives may be wrong. Thus, initiation of explosives transparent for laser radiation requires compulsory consideration of optical effects being rather topical for each explosive.

One of the effects of such explosive composite structure is the explosion delay which time depends upon the dimensions and concentration of nanodiamond powder. The concept, combining the abovementioned ones [10 - 16] and other similar papers, is based on practical ideas: topical practical task is to develop reliable primary light-sensitive explosives with the addition of different-material diffusors. That is one way. Another way to develop light-sensitive explosives is based on the idea of composite explosives – polymer matrix saturated with explosive microparticles [10].

Studies of the ignition mechanism involved measuring of the absorption spectrum of some brisant explosives in terms of specially prepared samples. In this context, silver azide was used in the form of thin transparent plates with the thickness of 0.1 - 0.01 mm. The measurement results have demonstrated that in terms of room temperature, that substance is active to absorb light with the wavelength being less than $\lambda = 0.35$ mcm. If $\lambda = 0.3$ mcm, then coefficient of absorption is about 10^5 m^{-1} with sharp growth in terms of further decrease in the wave length. If $\lambda > 0.385$ mcm up to the closest infrared spectrum area, absorption is insignificant. Similar results have been also obtained for other explosives such as thallium and lead azides, fullminating silver, lead styphnate, and TEN [16].

Data on the measurement of spectrum coefficient of explosive absorption have become a basis for a thermal ignition mechanism. The essence of the mechanism is as follows: being absorbed in a thin layer of the explosive (thickness is $10^{-5} - 10^{-6}$ m), light of blue and ultraviolet portion of the spectrum results in its heating sufficient for the initiation of chemical reaction. Experiments with the lead styphnate ignition by means of flashover light

indicate the important role of short-wave radiation for the explosive initiation [10]. Conclusion as for the essential role of a thin surface layer of the substance in the process is confirmed by a series of other results. For instance, paper [17] states that lead azide initiation does not depend upon the material pureness, dispersion, and sample density. Moreover, the paper highlights the effect of dispersion of the substance decomposition products upon the ignition parameters being characteristic for considerable indices of explosive absorption.

Studies of that problem in Ukraine started its considerable development along with the use of lasers while researching brisant explosives ignition [18 - 25]. Since the beginning of 1990s, studies have been carried out by scientists of Dnipropetrovsk Mining Institute both with the colleagues from the Russian Federation [26 - 28] or Poland [29 - 31] and individually [32 - 34].

Experimental studies of the initiation of detonation of light-sensitive energy-saturated composites and brisant explosives were performed involving nanosecond (impulse duration is $\tau = (2 - 5) \cdot 10^{-8}$ s), microsecond ($\tau = (0.5 - 0.7) \cdot 10^{-6}$ s), and millisecond ($\tau \sim 10^{-3}$ s)pulses of neodymium and ruby lasers [18 – 23]. While analyzing the obtained results, attention should be paid to the issues relating to the problems of mechanical ignition of the explosives and problem of practical implication of the laser initiation method. Sensitivity to the action of laser pulse is the most important property of brisant explosives.

Despite the fact that the majority of the considered explosives demonstrate low absorption of the radiation of neodymium (wave length is $\lambda = 1.06$ mcm) and ruby ($\lambda = 1.69$ mcm) lasers, values of critical densities of the ignition energy, obtained in the series of studies, have appeared to be equal or less than the corresponding energies, measured whileinitiating continuous spectrum by light sources (Table 1). As it has been expected, the research results show that sensitivity to the laser impulse action of the initiated explosivesis much higher than the one the secondary explosives. In other words, widely accepted series of the explosive sensitivity correlates in general with the sensitivity to laser action.

Table demonstrates that the explosive sensitivity depends upon the laser impulse duration. All the analyzed substances have made it possible to note that lengthening of the impulse duration results in the increased initiation threshold [26 - 29, 32]. Unfortunately, that issue has not studied enough. Only paper [28] shows the performed measurement of threshold density of lead azide initiation energy in terms of smooth changes in pulse duration within the range of 7.5 - 100 ns. The experiment has demonstrated that the lengthened duration of the action results in the increased ignition threshold. Range of energy density being 50 - 300 J/m² corresponds to the mentioned range of changes in the action duration.

The developed powerful laser systems have widened considerably the range of explosives potentially sensitive to the laser radiation effect. Studies have shown that phlegmatized TEN may be initiated by neodymium laser pulse with the duration of 4 ns and energy density of $10^7 - 10^8$ J/m² [I7]. That paper proposes following initiation mechanism: as a result of material ablation in solid substance, shock wave is excited with its further transformation into detonation one. The research results have proved the previously drawn conclusion [18] on the fact that laser energy losses for substance evaporation and dispersion are the powerful source of energy losses in the process of laser initiation of brisant explosives.

Research methodologies. The paper applies methodology of experimental and theoretical studies as well as mathematical modeling with the use of gas-dynamics equations.

Explosive name	Length of radiation wave λ , mcm	Impulse duration $ au$, s	Threshold energy density E_{kr} , J/cm ²	Source of information
Silver azide	1.06	10-3	2.1	
Cadmium azide	1.06	5·10 ⁻³	0.015	[18]
Cadmium azide	1.06	10 ⁻³	4.0	
Fulminatingmercury	1.06	5.10-8	1.0	
Lead styphnate	1.06	5.10-8	0.4	
Lead styphnate	1.06	10-3	2.2	
Lead azide	1.06	10 ⁻³	0.8	[19]
Lead azide	1.06	3.10-8	$3.8 \cdot 10^{-8}$	
Lead azide	1.06	6·10 ⁻⁷	$3.0 \cdot 10^{-2}$	[23]
Lead azide	1.06	10-7	0,8	[21]
β – lead azide	0.69	8·10 ⁻⁸	$1.5 \cdot 10^{-3}$	[22]
TEN	0.69	4·10 ⁻⁹	$4 \cdot 10^{-2}$	
TEN	0.69	$4 \cdot 10^{-9}$	10-3	[23]
TEN	0.69	3.10-8	$1.3 \cdot 10^{-2}$	
TEN	0.69	3.10-8	$5 \cdot 10^{3}$	
Octogen	0.69	3.10-8	15	
Hexogen	0.69	3.10-8	30	
Pyroxyline	1.06	3.10-8	60	
$0.45Si + 0.55P_3O_4$	1.06	10-3	10.6	
	1.06	10-3	2.9	[26]
0.25Al+0.75KClO ₄	1.06	10-3	6.6	[30]
	1.06	2.10^{-8}	5.0	

Table 1. Critical densities of the ignition energy for certain initiated explosives.

Objective of the paper is to carry out studies of physicochemical peculiarities of the mechanism to stimulate explosives by means of pulse radiation of optical quantum generator. Standard methods to test phlegmatisedsilver azide as for its sensitive it to external physical and mechanical effects have been applied.

Objective of the paper is to study regularities of the initiation of explosive detonation and to analyze dependence of the ignition delay upon the density of laser radiation energy.

3 Statement of the basic material

Semi-space $x \ge 0$ is occupied with TEN on which radiation of constant intensity q falls. Semi-space x < 0 is vacuum. While initiating, coefficients of absorption K and reflection A are considered to be constant.

Energy is absorbed according to the exponential law. Movement of chemically active medium being compressed is described by equations of gas dynamics:

$$\frac{\partial u}{\partial t} + \frac{\partial p}{\partial t} = 0, \ \frac{\partial V}{\partial t} - \frac{\partial u}{\partial m} = 0;$$
$$\frac{\partial}{\partial t} \left(e \frac{u^2}{2} \right) + \frac{\partial}{\partial m} (pu) = q(I - A) \alpha \exp(\alpha m) + QL;$$

chemical kinetic:

$$\frac{\partial a}{\partial t} = -L(a, e);$$

and state equation:

$$P = p(v, e).$$

In this context, e, u, p, v, a are in terna lenergy, velocity, pressure, specific volume, and

concentration of the explosive espectively; $\alpha = K_V$ is mass coefficient of absorption; *t* is time; *m* masscoordinate; *Q* is TEN calorific capacity.

Expression for L in kinetic equation was applied in Arrhenius form:

$$L = az \exp(-E_a C_v / R \cdot e_T),$$

where z is pre-exponential factor; E_a is activation energy; R is gas constant; e_T is thermal constituent of internal energy.

The calculations involved semi-empirical equation of state [29] describing properties of TEN and decay products:

$$p = \frac{p_0 c_0^2}{n-k} (y^n - y^k) + \Gamma(y, e_T) \rho e_T$$

In this context $G = \left(1.5 + \frac{\partial y}{I + by}\right) \left(\frac{I + \mu e_T}{I + \varepsilon e_T}\right)$ is Gruneisen coefficient; p is explosive

density; p_0 is initial density; c_0 is sound velocity under normal conditions; c_V is thermal capacity; $y = p/p_0$ is compression; n = 6.21, k = 3.40, d = 5.35, b = I, $\mu = 2.136 \cdot 10^{-3}$ kg/kJ, $\varepsilon = 6.4 \cdot 10^{-3}$ kg/kJ are empiric constants obtained from experimental data on TEN shock compression and additional data on TEN physicochemical properties.

Calculation of the phase disintegration was performed according to Maxwell rule. Phase mixture was assumed to be balanced being described by a one-velocity model.

As a result of the introduction of dimensionless variable, following parameters of the problem, which were taken as constant ones, have been obtained

$$\lambda_1 = \frac{Q_u}{c_0^2}, \ \lambda_2 = \frac{Q}{c_0^2}, \ \lambda_3 = zt_l = \frac{Q}{c_0^2}$$

while parameters

$$\delta = \frac{q(I-A)\alpha t_q}{Q_U}$$
 and $\tau = t_q / t_G$

experienced their changes correspondingly to q, b, α changes; Q_u is evaporation heat; t_q is laser pulse duration, $t_G = K^{-1}c_0^{-1}$ is characteristic time of the propagation of sound excitation within the substance layer of a unit optical thickness.

If $\tau \gg 1$, then the shock release wave will cover the absorbing layer completely within the operating time of optical quantum generator. In this case, thermal energy increase within the absorbing volume is limited by energy losses for gas-dynamic motion of the material. If $\tau \gg 1$, then the shock release wave will propagate over the explosive layer of optical thickness being far less than a unit. Thus, rapid energy growth is possible within the internal areas of the explosive. In terms of the calculations, τ parameter was taken as equal to 1 - 10. Absorption coefficients of some explosives measured in papers [13, 15, 34] fall into the range of $10^4 - 10^5 \text{m}^{-1}$ which corresponds to the indicated range of τ change.

In terms of $\delta = 3 - 10$ range, we can expect that coefficient of explosive absorption while initiating will be approximately constant.

4 Discussion of the obtained results

Fig. 1 represents dependences of TEN initiation time upon τ that corresponds to different values of δ parameter, i.e. 5, 7, 9. It is clear that if δ is constant, then initiation grows proportionally to δ increase, i.e. growth of the coefficient of explosive absorption in terms of unchangeable velocity of energy introduction results in the growth of "gas-dynamic losses" which was expected. Besides, curves slope to the abscissa axis decreases along with δ increase. It means that the effect of the absorption coefficient (substance dispersion) in this case is of less importance. Thus, growing intensity of radiation causes the increase in energy efficiency of radiation.

Fig. 2 shows temporal development of TEN initiation process, if $\delta = 5$ and $\tau = 1$. Density of the absorbed energy of light radiation E_l and coefficient of mass absorption α corresponding to data δ and τ are equal to $19 \cdot 10^4 \text{J/m}^2$ and 7.59 m²/kg.

At t = 10.8 ns moment of time, dependence of heat energy upon optical thickness is equal to exponential except the surface area where curve slope to axis *m* decreases slightly. That is connected with the internal energy losses for evaporation and substance motion. Further heating results in more rapid energy growth within the substance internal areas and initiation of chemical reaction inside the explosive. In this case, initiation takes place at t = 20.6 ns moment of time within $m \approx 0.15$ plane.

Light energy absorption is accompanied by the formation of the pressure peak with exponential leading edge. Fig. 2 represents the motion of that excitation into the substance depth which propagation velocity is close to normal sound velocity. Development of chemical reaction results in "chemical" pressure peak (curve I).





Fig. 1. Dependence of TEN initiation time upon δ and τ parameters.

Fig. 2. Evolution of thermal energy within TEN surface layer during laser initiation: $\delta = 5$, $\tau = 1$; time t (ns): 1 - 20.6; 2 - 10.8.

Figs. 3 – 5 show changes of thermal energy and pressure respectively in terms of other τ value and $E_l = 2.17 \cdot 10^4 \text{ J/m}^2$, $\alpha = 68.3 \text{ m}^2/\text{kg}$. We can see that up to 10 ns moment of time, the explosive does not practically evaporate; that is why, dependence of thermal energy upon optical thickness is close to exponential.

Since $\tau \gg 1$, shock release waves, propagating from the boundaries of substance being heated, are able to reduce pressure within the area of light action up to that moment (Fig. 3, curve 4). It results in the increased energy losses per gas-dynamic motion and, consequently, in further growth of thermal energy and pressure (see curves 3). Since the velocity of energy addition on the explosive surface is maximal, internal energy grows a little faster than within the surrounding layers of the substance. Up to the moment of light

pulse (t = 30 ns), energy density obtained on the surface is enough for further self-acceleration of chemical reaction (curves 1, Fig. 3). It is natural that in this case there is no "chemical" pressure peak. Excitation motion, generated as a result of laser energy absorption, is impossible to observe in Fig. 3 as up to t = 10 ns moment it has propagated at the distance exceeding the mentioned ones.





Fig. 3. Evolution of thermal energy within TEN surface layer during laser initiation: $\delta = 5$, $\tau = 9$, time t (ns): 1 - 31.4, 2 - 30, 3 - 20, 4 - 10.

Fig. 4. Evolution of thermal energy within TEN surface layer during laser initiation: $\delta = 5$, $\tau = 9$, time *t* (ns): 1 - 31.4, 2 - 30, 3 - 20, 4 - 10.

According to the data published for the resent 10 years, TEN initiation in terms of much higher radiation flow density is of the same character as in the examples analyzed before. In terms of minor values, there is no self-acceleration of chemical reaction. For instance, if $\delta = 4$, initiation takes place, if $\tau \le 3$. If $\tau > 3$, "gas-dynamic losses" terminate the development of chemical reaction irrespective of the fact that density of absorption energy during the laser operation exceeds evaporation heat by 4 times. If $\delta = 4$ and $\tau = 4$ up to t = 30 ns moment of time (end of the pulse action), maximum density within $m \sim 0.28$ plane is obtained. Further, thermal energy and pressure of surface layer decrease. Drop in energy density within the area of optical thickness m > 0.6 is of slower character than in the surface layer. It is connected with the fact that energy reserve is not sufficient for further substance evaporation and dispersion.



Fig. 5. Evolution of thermal energy within TEN surface layer during laser initiation: $\delta = 5$, $\tau = 1$, time t (ns): 1 - 20.6; 2 - 10.8.

Basing upon the practice of the development of the first optical initiation system in the world [38, 39], dependence of the initiation threshold upon the dimensions of radiated zone (size effect) should be taken into consideration. Character of the known dependences tells about the availability of boundary radiation spot which decrease results in the fact that threshold density of the initiation energy starts its growth and initiation energy drops. When

laser beam radius tends to zero, initiation energy tends to a finite value. In such a way, for instance, energy of the initiation of pressed (by $2 \cdot 10^9$ Pa pressure) lead azide by laser beam of \emptyset 40 mcm is 6 mkJ; it experiences practically no changes in terms of further diameter decrease [25]. That effect is explained by the conditions of lighting within the explosive volume being typical diffusively dispersing medium with dense packing of diffusers [37, 38].

A problem concerning dependence of the initiation delay upon the value of energy density of laser radiation is of high importance. That problem was studied by A. Chernay in his thesis and in papers [40 - 41] while analyzing peculiarities of lead azide initiation by 30 ns monopulse. It has been shown that in terms of energy density increase, initiation delay decreases and in the contest of three-time or four-time exceedance of critical value, it becomes less than the laser pulse duration. That statement is rather important from the practical viewpoint as it indicates the fact that laser action may be used to obtain high accuracy of the explosive charge operation [29, 30, 43 – 45]. During the action of neodymium laser monopulse, time of phlegm atized silver azide delay was recorded within the limits of 0.17 - 0.22 mcs. Energy of laser radiation exceeded the threshold energy of lead azide ignition.

Tests of the samples of phlegmatized silver azide and TEN according to the standard methodologies as for their sensitivity to mechanical, thermal, and electric actions have shown that none of the obtained indices of silver azide exceeds the results obtained during TEN testing. Thus, on the one hand, phlegmatized silver azide is the priming explosive; on the other hand, in terms of its sensitivity to external actions, safety rate of phlegmatized silver azide is similar to the brisant explosives. Consequently, the obtained results make it possible to state that light-sensitive explosive – phlegmatized silver azide – is the explosive which may be used for the initiating agents and which determines the overall safety of the blasting operations.

5 Conclusions

Numerical modeling has been performed involving physical parameters of the explosive with pulse laser radiation on neodymium glass with 1.06 mcm wave length. Mathematical model makes it possible to study peculiarities of the initiation of explosive transformation of brisant explosives by means of short light pulses. In terms of TEN, it has been demonstrated that the process is completely determined by the parameters characterizing radiation intensity and absorption properties of the explosive. Depending upon the values of those parameters, qualitatively different initiation processes may be implemented – either on the surface or inside the substance. In the latter case, emission of chemical energy results in the formation of so-called "chemical" pressure peak. Factors influencing the explosive initiation by laser radiation often include initial temperature of the explosive sample. According to the data of certain studies, dependence of the explosive sensitivity to laser radiation upon the initial temperature is known. There were experimental observations of insignificant increase in the sensitivity of lead azide samples in terms of initial temperature growing by 420 K, if laser in free generation mode is used (pulse is up to 1 ms). However, in terms of giant mode ($\tau = 30$ ns), effect of the sample heating temperature upon the sensitivity was not defined.

According to all the obtained indices while testing the explosives for their sensitivity, phlegmatized silver azide is the example of one of the way to obtain relatively safe priming light-sensitive explosives. To improve considerably the safety rate of blasting operations in mines, it is recommended to apply light-sensitive priming explosive and laser systems of initiation respectively.

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