Full Paper

Propellants, Explosives, Pyrotechnics

DOI: 10.1002/prep.201800142

Ignition of Organic Explosive Materials by a Copper Oxide Film Absorbing a Laser Pulse

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Abstract: This work present experimental results of PETN ignition and of numerical simulations of PETN, RDX, HMX, and TATB ignition by a copper oxide (CuO) film heated using a millisecond fiber laser YLS-150. It was established that the dynamic ignition threshold of organic explosive materials (EM) in a three-layer system (glass – CuO – EM) by a laser pulse has a minimum at the CuO film thickness of 12 μm. The density of the critical ignition energy of EM is increasing in the sequence of PETN, RDX, HMX, and TATB with PETN having the smallest and TATB having the highest density of the critical ignition energy.

Keywords: simulation · laser ignition · explosive · copper (II) oxide · PETN · RDX · HMX · TATB

1 Introduction

A number of works on experimental study and mathematical modeling of the initiation of condensed EM, heterogeneous pyrotechnic compounds, and solid rocket fuels by laser pulses with lengths from milli- to nanosecond, has increased over the last decade. First of all, it is explained by an increase of sensitivity of modern electronic equipment used for recording of fast processes occurring in condensed EM during the delay period, e.g., pulse conductivity and luminescence of a solid and explosion products. Secondly, the conductivity and luminescence data of a solid body are necessary for the development of mechanisms and mathematical models of the ignition of condensed EM. Finally, laser initiation of energetic materials is studied with the aim of creation of light detonators, effective and safe ignitors for future weapon systems and rocket engines [1–3].

Experimental study and numerical calculations of the PETN ignition by a CuO film absorbing a millisecond laser pulse are described in [4–5]. Systems of a CuO – PETN and a glass – CuO – PETN were considered. Recently, a mathematical modeling has been done to determine the ordinary dependence of the ignition threshold by a laser pulse [6] and detonation by impact [7] of organic EM.

The aim of this work is to perform a numerical simulation of the ignition of organic EM (PETN, RDX, HMX, and TATB) by a CuO film heated using a millisecond fiber laser YLS-150 in a three-layer system (glass – CuO – EM), and to determine the possibility of ignition of RDX, HMX, and TATB by the given laser.

The aim of this work is to determine the possibility of ignition of RDX, HMX, and TATB by a CuO film heated using a millisecond fiber laser YLS-150 in a three-layer system (glass – CuO – EM), and to measure dependence of the ignition threshold of organic EM (PETN, RDX, HMX, and TATB) by the given laser in the three-layer system using numerical calculations.

1.2 Description of the Numerical Calculations

Consider a three-layer heterogeneous system: glass – CuO – EM. For the given three-layer system, we can write a one-dimensional heat equation. For the glass ($h_1 \geq z \geq 0$)

$$\rho_1 c_1 \frac{\partial T_1}{\partial t} = \lambda_1 \frac{\partial^2 T_1}{\partial z^2} \tag{1}$$

for the CuO film ($h_2 \geq z \geq h_1$)

$$\rho_2 c_2 \frac{\partial T_2}{\partial t} = \lambda_2 \frac{\partial^2 T_2}{\partial z^2} + \alpha(1 - R_{12}) I_0(t) \exp(-\alpha(z - h_1)) \times \times \frac{[1 + R_{23}\exp(2\alpha(z - h_1 - h_2))]}{[1 - R_{12}R_{23}\exp(-2\alpha h_2)]} \tag{2}$$

for EM ($h_3 \geq z \geq h_2$)

$$\rho_3 [c_3 + H_f \delta(T_3 - T_f)] \frac{\partial T_3}{\partial t} = \lambda_3 \frac{\partial^2 T_3}{\partial z^2} + \rho_3 Q Z exp\left(-\frac{E}{RT_3}\right) \tag{3}$$

with initial and boundary conditions:

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$$T_1(z,\ 0) = \ T_2(z,\ 0) = T_3(z,\ 0) = T_0, \tag{4}$$

$$\frac{\partial T_1(0,\ t)}{\partial z} = \frac{\partial T_3(h_1 + h_2 + h_3, t)}{\partial z} = 0, \tag{5}$$

$$\lambda_1 \frac{\partial T_1(h_1,\ t)}{\partial z} = \lambda_2 \frac{\partial T_2(h_2,\ t)}{\partial z},\ T_1(h_1,\ 0) = \ T_2(h_2,\ 0), \tag{6}$$

$$\lambda_2 \frac{\partial T_2(h_1 + h_2,\ t)}{\partial z} = \lambda_3 \frac{\partial T_3(h_1 + h_2,\ t)}{\partial z},$$

$$T_2(h_1 + h_2,\ t) = \ T_3(h_1 + h_2,\ t). \tag{7}$$

Where $h_1,\ T_1$ - the thickness and the temperature of the glass plate; $h_2,\ T_2$ – the thickness and the temperature of the CuO film, $h_3,\ T_3$ – the thickness and the temperature of EM; T_0 – the initial temperature of the three-layer system glass – CuO – EM; $\lambda_1,\ c_1$ – the thermal conductivity coefficient and the specific heat capacity of the glass plate; $\lambda_2,\ c_2$ – the thermal conductivity coefficient and the specific heat capacity of the CuO film; $\lambda_3,\ c_3$ – the thermal conductivity coefficient and the specific heat capacity of the EM; $\rho_1,\ \rho_2,\ \rho_3$ – the densities of the glass, CuO, and EM respectively; $R_{12},\ R_{23}$ – the reflection coefficients of the light flux from the glass – CuO boundary and CuO – EM respectively; $H_f,\ T_f$ – heat of fusion and the melting point of EM; $Q,\ Z,\ E$ – the heat of reaction per unit mass, the frequency factor, and the activation energy of thermal decomposition of EM; R – the universal gas constant; $\delta(T_3 - T_f)$ – the delta function.

The EM heat conduction Eq. (3) contains the delta function due to the fact that at the phase transition temperature $T = T_f$, the energy as a function of temperature have a discontinuity at the value of H_f [8]. The delta function, according to [8], is given by

$$\delta(T_3 - \ T_f) = \frac{\partial \vartheta(T_3 - T_f)}{\partial T},$$

where ϑ – the Heaviside function defined by the expression

$$\vartheta = \begin{cases} 1,\ T_3 - T_f \geq 0 \\ 0,\ T_3 - T_f < 0 \end{cases}.$$

Approach described in [4] allows us to take the fusion into account to solve the heat equation of EM.

The dependency of the light flux intensity from time is given in the form of a rectangular pulse

$$I(t) = W_{max}/\tau_i = I_0,\ \tau_i \geq t \geq 0,\ I(t) = 0,\ at\ t > \tau_i, \tag{8}$$

where $W_{max} = 72.2 \cdot 10^4\ J/m^2$ – the energy density of the laser pulse; $\tau_i = 20 \cdot 10^{-3}\ s$ – the duration of the laser pulse [6]. Thus, the rise time of the laser pulse energy density is linear $W = \ I_0 t$ at $\tau_i \geq t \geq 0$.

The absorption of the transmitted light flux in the glass plate and EM was neglected due to the small absorption coefficients of the glass and organic EM in this spectral region. According to [9], the secondary organic EM at the wavelength of the first harmonic of a pulsed neodymium glass laser ($\lambda = 1.06\ \mu m$) have high transparency.

The external heat dissipation in this formulation of the ignition problem of EM was neglected due to the fact that the duration of the laser pulse and the delay time of the ignition are much shorter that the characteristic time of the heat dissipation. In addition, it was assumed that the thermal conductivity and heat capacity of the melted and solid EM differ insignificantly. The burnout was also not taken into account as according to [10], for EM with a large thermal effect, a large activation energy, and a short thermal initiating pulse in the region of the ignition threshold the burnout can be neglected. In [11–13], one-dimensional problems of EM ignition with nanosecond electron pulses were solved, in which burnout was taken into account. Calculations have shown that burning of EM is insignificant and does not affect the threshold of the thermal initiation. For example, for an exponential distribution of the absorbed energy of a nanosecond electron pulse, it is shown that at the end of the pulse the degree of decomposition on the surface is $3 \cdot 10^{-4}\ \%$, and by the time of the explosion ~2.5 % [13].

In the numerical solution of the system of equations (1)–(3) with initial and boundary conditions (4)–(7), implicit difference schemes were used. The difference equations were solved by the sweep method [14]. In this case, the three-layer glass – CuO – EM system was represented as a continuous medium, in which the thermophysical, optical, and chemical properties depend on the coordinate. The Arrhenius nonlinearity was linearized at each time step by means of the Frank-Kamenetsky transformation

$$exp\left(-\frac{E}{RT_{3i}}\right) \approx exp\left(-\frac{E}{R\check{T}_{3i}}\right) exp\left(\frac{E\Delta \mathrm{T}_{3i}}{R\check{T}_{3i}^2}\right),$$

where $\Delta T_{3i} = T_{3i} - \check{T}_{3i},\ T_{3i},\ \check{T}_{3i}$ – the temperatures of the current and the previous time step in the i-th cell of the difference scheme along $z$coordinate. Then, taking into account that $\Delta T_{3i} = T_{3i} - \check{T}_{3i} \ll 1$, the exponential was expanded in a series up to terms of the first order of smallness

$$exp\left(-\frac{E}{R\check{T}_{3i}}\right) exp\left(\frac{E\Delta T_{3i}}{R\check{T}_{3i}^2}\right) \approx \left(1 + \frac{ET_{3i}}{R\check{T}_{3i}^2} - \frac{E}{R\check{T}_{3i}}\right) \exp\left(-\frac{E}{R\check{T}_{3i}}\right).$$

When solving the system of heat conduction equations, the time and coordinate steps were constant and did not depend on the thickness of the CuO film and were respectively equal to $h_t = 10^{-7}\ s$ and $h_z = 0.04\ \mu m$. With these values, the error in the law of energy conservation did not exceed 3 %. The thickness of the glass plate and PETN was ~1 mm. The characteristic warm-up time for glass and

EM with the 1 mm thickness is much shorter than the time of the initiation delay of EM. To reduce the time and the error of calculations in the numerical simulation, the values of the thicknesses of the glass plate and EM were chosen in a way that, at the time of initiation, the temperatures on the boundaries of the three-layer system did not change, i.e., $T_1(0,t) = T_3(h_1 + h_2 + h_3,\ t) = T_0$.

The numerical solution of the heat conduction equations for glass and CuO was carried out with the thermophysical parameters and densities given in Table 1. The specific heat capacity of the CuO film was taken from [17] at 450 K. The coefficient of thermal conductivity of the CuO film was taken from [16], as for compressed powder. The numerical solution of the heat equation for secondary explosives was carried out using the kinetic and thermophysical parameters given in Table 2.

Table 1. Thermophyiscal parameters of materiales.

Thermophysical parameters	$\lambda,\ W/(m \cdot K)$	$c,\ J/(kg \cdot K)$	$\rho,\ kg/m^3$
Glass	1.05	$8.0 \cdot 10^2$	$2.6 \cdot 10^3$
CuO	1.013	$6.0 \cdot 10^2$	$6.45 \cdot 10^3$

Table 2. Kinetic and thermal parameters of organic explosives.

EM	PETN	RDX	HMX	TATB
$E,\ kJ/mol$	196.6	197.3	220.8	250.9
Z, s^{-1}	$6.3 \cdot 10^{19}$	$2.02 \cdot 10^{18}$	$5.0 \cdot 10^{19}$	$3.18 \cdot 10^{19}$
$Q,\ MJ/kg$	1.26	2.1	2.1	2.51
$c,\ J/(Kg \cdot K)$	1255.2	1020	1250	1000
$\lambda,\ W/(m \cdot K)$	0.2508	0.105	0.293	0.418
$\rho,\ kg/m^3$	$1.77 \cdot 10^3$	$1.82 \cdot 10^3$	$1.9 \cdot 10^3$	$1.93 \cdot 10^3$
$H_f,\ KJ/kg$	193	235.5	192.46	270
$T_f,\ K$	413	476	558	623

The absorption coefficient of the CuO film at the wavelength of the ytterbium fiber laser was measured experimentally using a Shimadzu UV-3600 spectrophotometer with $\alpha = 1.75 \cdot 10^5\ m^{-1}$ [5]. Coefficients of reflection from the interface $R_{12} = 0.53,\ R_{23} = 0.52$ [5].

2 Results and Discussion

The results of the numerical calculations of the system of heat equation Eq. (1–3) with the initial and boundary conditions Eq. (4–7) for PETN, RDX, HMX, and TATB are shown in Figures 1–6. Calculations have shown that the ignition of PETN, RDX, HMX, and TATB occurs near the interface of the light-absorbing CuO film – EM as shown in Figures 1–4. The ignition of EM by a hot "wall" occurs before the end of the laser pulse, i.e., in the dynamic heating mode. As the absorbed energy in the CuO film increases, the nature of the

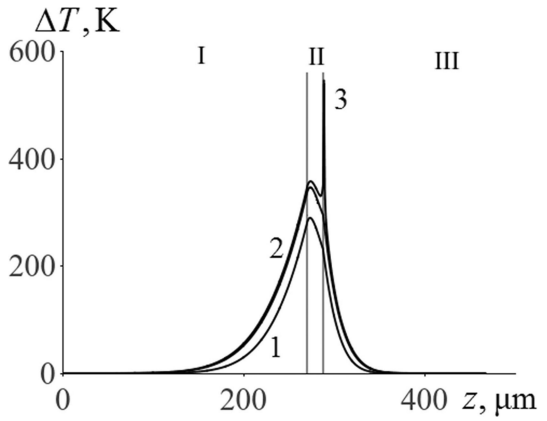

Figure 1. The temperature distribution profile in the glass plate (I), CuO film (II), and PETN (III) at the film thick-ness of $h_2 = 18\ \mu m$ and time steps $t = 3.0\ ms$ (1), $t = 4.0\ ms$ (2), and $t = 4.17\ ms$ (3).

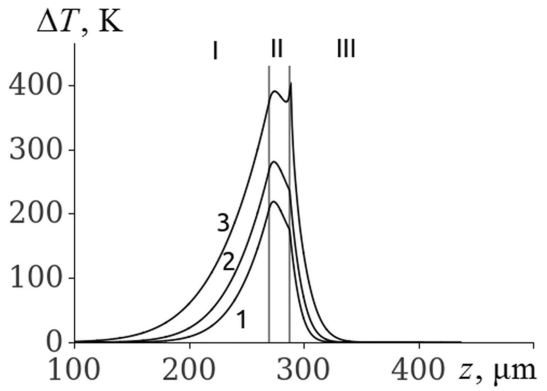

Figure 2. The temperature distribution profile in the glass plate (I), CuO film (II), and PETN (III) at the film thick-ness of $h_2 = 18\ \mu m$ and time steps $t = 1.76\ ms$ (1), $t = 2.57\ ms$ (2), and $t = 4.23\ ms$ (3).

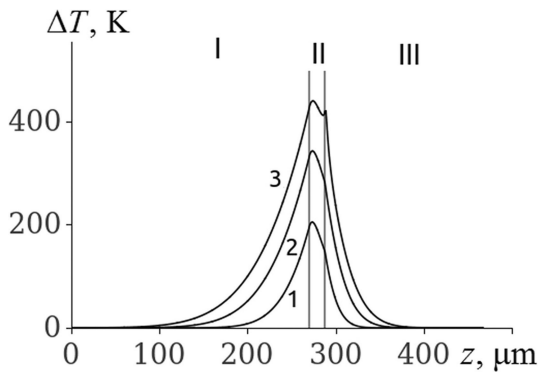

Figure 3. The temperature distribution profile in the glass plate (I), CuO film (II), and PETN (III) at the film thick-ness of $h_2 = 18\ \mu m$ and time steps $t = 2.1\ ms$ (1), $t = 4.6\ ms$ (2), and $t = 6.027\ ms$ (3).

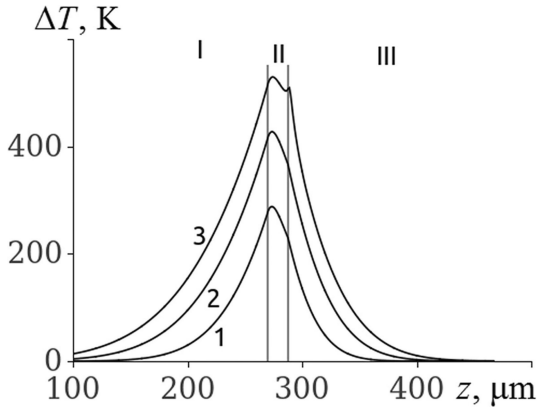

Figure 4. The temperature distribution profile in the glass plate (I), CuO film (II), and PETN (III) at the film thick-ness of $h_2 = 18\ \mu m$ and time steps $t = 3.02\ ms$ (1), $t = 5.84\ ms$ (2), and $t = 8.27\ ms$ (3).

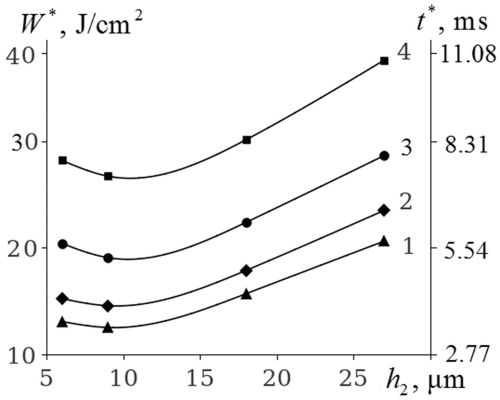

Figure 5. Dependence of the critical energy density and the dynamic delay time of ignition by a laser pulse on the thickness of the CuO film: 1-PETN; 2-RDX; 3-HMX; 4-TATB.

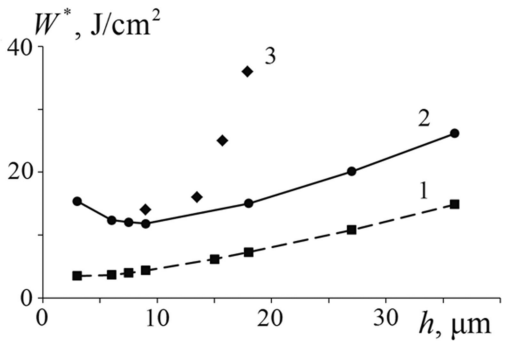

Figure 6. Dependence of the energy density of ignition PETN laser pulse on the thickness of the CuO film: 1 – two-layer system CuO – EM (calculation); 2 – three-layer system glass – CuO – EM (calculation); 3 – experiment [5].

temperature distribution over the depth of the three-layer system begins to change Figures 1–4: the temperature of the near-boundary layer of EM increases due to the exothermic reaction, and a significant amount of heat is transferred from the EM to the CuO film, see curves 3 in Figures 1–4. As can be seen from these figures, the amount of heat in the glass plate is noticeably larger than in EM.

In the experiment, the dynamic delay time t^* of the EM ignition is measured when a square-wave laser pulse is absorbed, which is then used to calculate the critical energy density. The dynamic delay time of the ignition of EM and the energy density of the initiating laser pulse W^* are related in the following way

$$W^* = t^* \cdot I_0.$$

Figures 1–4 show that the ignition delay time t of EM increases in the sequence: PETN, RDX, HMX and TATB, and varies accordingly from 4.17 *ms* (PETN) to 8.27 *ms* (TATB) at a CuO film thickness of 18 μm. In addition, the calculations have shown that the threshold energy density of ignition by a laser pulse W^* and the dynamic ignition delay time t^* of organic EM depend from the thickness of the CuO film in the three-layer system, increases in the sequence: PETN, RDX, HMX, TATB and have the minima as shown in Figure 5. The position of the minima on the dependencies $W^* = f(h_2)$ for EM data is approximately the same and is located at $h_2 \sim 12\ \mu m$.

Let us consider, at a qualitative level, the physics of the influence of the thickness of the CuO film on the dynamic ignition delay time. To do this, we determine the amount of heat Q_2 that is released in the CuO film when light is absorbed, taking into account the heat transferred to the glass plate and EM. Calculations have shown, as presented in Figures 1–4, that the temperature distribution profiles along the z coordinate at different time steps in both the glass plate and in EM at the boundaries with the CuO film are similar almost until the moment of ignition. Consequently, starting from the boundary conditions (6) and (7), we have

$$-\lambda_2 \frac{\partial T_2(h_1,t)}{\partial z} = -q_{12} \approx const, \quad -\lambda_2 \frac{\partial T_2(h_1+h_2,t)}{\partial z} = q_{23} \approx const, \tag{9}$$

where $-q_{21}$ – the heat flow to the glass plate from the CuO film; q_{23} – the heat flow into the EM from the CuO film.

The amount of heat in the CuO film is determined by the expression

$$Q_2 = \rho_2 c_2 \int_{h_1}^{h_1+h_2}\int_0^t \frac{\partial T_2}{\partial t} dtdz =$$
$$\int_{h_1}^{h_1+h_2}\int_0^t \left\{\lambda_2 \frac{\partial^2 T_2}{\partial z^2}\right\} dtdz +$$
$$\alpha(1-R_{12}) \int_{h_1}^{h_1+h_2}\int_0^t \{I_0(t)\exp[-\alpha(z-h_1)]$$
$$\times \frac{[1+R_{23}\exp(2\alpha(z-h_1-h_2))]}{[1-R_{12}R_{23}\exp(-2\alpha h_2)]}\Big\} dtdz. \quad (10)$$

Integrating Eq. (10) with the account that the laser pulse, according to Eq. (8), has a rectangular shape and taking into account the boundary conditions as given by Eq. (9), we obtain

$$Q_2 = -(q_{21}+q_{23})t + tI_0(1-R_{12})$$
$$\times \frac{[1-\exp(-\alpha h_2)]\left[1+R_{23}e^{-\alpha h_2}\right]}{\left[1-R_{12}R_{23}e^{-2\alpha h_2}\right]}. \quad (11)$$

We determine whether the function Q_2 has discontinuities dependgin on the thickness of the CuO film h_2. Taking the derivative of Eq. (11) with respect to h_2, we obtain that

$$\frac{\partial Q_2}{\partial h_2} = \alpha t I_0(1-R_{12})$$
$$\times \frac{1-R_{23}+2R_{23}(1-R_{12})e^{(-\alpha h_2)}+R_{12}R_{23}(1-R_{23})e^{-2\alpha h_2}}{\left[1-R_{12}R_{23}e^{(-2\alpha h_2)}\right]^2} > 0,$$

since $0 < R_{12},\ R_{23} < 1$, $e^{-\alpha h_2} > 0$, and $e^{-2\alpha h_2} > 0$.

Thus, the function Q_2 has no discontinuities and is a monotonically increasing function, as it should be from the point of view of physics. The greater the thickness of the CuO film, the less light energy leaves the film due to absorption. In the case of a thick film ($\alpha h_2 \gg 1$), expression (11) takes the form

$$Q_2 = -(q_{21}+q_{23})t + tI_0(1-R_{12}), \quad (12)$$

that is, the energy is absorbed in the near-surface layer.

For a thin film ($\alpha h_2 \ll 1$), taking into account the expansion of exponentials in a series up to terms of the first order of smallness, Eq. (11) takes the form

$$Q_2 = -(q_{21}+q_{23})t + tI_0(1-R_{12})\frac{\alpha h_2(1+R_{23})}{1-R_{21}R_{23}}. \quad (13)$$

As can be seen from Eq. (13), the amount of heat released in a thin CuO film during a fixed time period is proportional to its thickness. Thus, to heat a thin CuO film by a laser radiation to the ignition temperature of EM, more heat is needed, and therefore more energy, which is proportional to time.

To heat the CuO film, with a thickness of more than 12 μm, to a critical temperature more time is also needed, since they have a larger "cold part". These two effects lead to the appearance of the minima on the dependencies of the critical energy density (the dynamic delay time of ignition by the CuO film) of the PETN, RDX HMX, and TATB ignition (Figure 5). The CuO film thickness of 12 μm is optimal, since the critical ignition density of PETN, RDX, HMX, and TATB is minimal.

Comparing the results of calculations of the dependencies of the threshold ignition energies for RDX, HMX, and TATB on the thickness of the CuO film with the experimental and numerical results for PETN, it follows that the ignition of the RDX by a millisecond YLS-150 fiber laser is entirely possible for all the thicknesses of the CuO film. Ignition of HMX and TATB in a three-layer system is possible only for CuO films with a thickness of the order of 12 μm.

Figure 6 shows the results of experiments [5] on the PETN ignition by an ytterbium fiber laser (points 3) in a three-layer system: glass – CuO film – PETN and numerical simulation (curves 1 and 2). Curve 1 corresponds to numerical simulation of PETN ignition in a two-layer system (CuO film – PETN), curve 2 – in a three-layer system: glass – CuO film – PETN. Comparing these curves, one can see that the heat transfer into the glass significantly increases the threshold energy density of the PETN ignition by a millisecond laser pulse. In addition, it can be seen that for thicknesses of the CuO film of up to 14 μm, the results of numerical calculations for a three-layer system (curve 2) satisfactorily agree with the experiment. However, for a thickness greater than 14 μm, the discrepancy between the calculated curve and the experimental points begins to increase. What it can be connected with?

According to the experiment carried out in [5], upon irradiation of the CuO film, deposited on a glass plate, by a millisecond laser, it changes color (acquires a brown color). This can be only explained by the reduction reaction of the CuO film

$$CuO \rightarrow \frac{1}{2}Cu_2O + \frac{1}{4}O_2. \quad (I)$$

The reduction reaction of copper oxide is endothermic, and the copper oxide reduction reaction was observed, for example, in [25, 26], when CuO polycrystals and single crystals were irradiated with nitrogen ions and helium ions, respectively. The largest reduction of CuO polycrystals and single crystals was observed near the surface. In addition to the Cu_2O phase, a phase of metallic copper was observed.

It was shown in [27] that the nanostructured CuO films exhibit significantly lower thermal stability than the copper oxide crystal. The thermal reduction reaction (I) of the film begins at 430 K, which is 350 K less than for copper oxide crystal.

The CuO film in [5] was obtained from a compressed copper oxide powder. According to [28], the enthalpy of CuO (H_1) and Cu_2O (H_2) formation from components in standard conditions is equal to

$$H_1 = -159.753\ kJ/mol,\ H_2 = -175.560\ kJ/mol.$$

According to Hess's law, the change in enthalpy of reaction (I) will be equal to

$$\Delta H = \frac{1}{2}H_2 - H_1 = 72.0 kJ/mol.$$

Thus, the thermal effect of reaction (I) is negative and equal to

$$Q_I = -\Delta H = -72.0\ kJ/mol.$$

Therefore, for a better description of the experimental results shown in Figure 6, it is necessary to take into account the endothermic reaction (I) in the thermal conductivity Eq. (1) for a film. Taking into account the endothermic reaction will lead to an increase in the critical energy density of PETN ignition. In connection with the appearance of the Arrhenius nonlinearity in Eq. (1), the dependence on the film thickness will be more pronounced. Then, the system of equations (1–3) needs to be extended with equations of the degree of film reconstruction and the level of PETN conversion. To further improve the model, the frequency factor and the effective activation energy of the heterogeneous CuO reduction reaction are required.

3 Conclusions

The threshold energy density of the laser pulse (dynamic delay time) of the EM ignition in the three-layer system glass plate – CuO film – EM has a minimum, depending on the thickness of the CuO film. The appearance of a minimum is due to the fact that the amount of heat released in the thin CuO film during a fixed time is proportional to its thickness. Therefore, the thinner the copper oxide film, the more time it takes to heat it with laser radiation to the ignition temperature of the explosives. For films with a thickness of more than 12 μm, additional energy is required and, accordingly, the time for warming up its cold part to the ignition temperature during the time of the laser pulse.

The density of the critical ignition energy of EM by the laser pulse increases in the sequence of PETN, RDX, HMX, and TATB. The discrepancy between the numerical calculations and the experiment for PETN at the CuO film thicknesses of more than 14 μm is not due to the endothermic reduction of the copper oxide film.

The densities of the critical energy of the PETN and RDX ignition by a laser pulse are close. Initiation of the HMX and TATB by the millisecond fiber laser YLS-150 in glass – CuO – EM system is possible in the region of a minimum with the CuO film thickness of the order of 12 μm.

Acknowledgements

This work was supported by the Russian Science Foundation [grant number 18-13-00031].

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Received: May 3, 2018
Accepted: July 10, 2018
Published online: September 4, 2018

