The regulation of secondary explosives sensitivity to laser influence

B P Aduev, D R Nurmukhametov, A A Zvekov and I Yu Liskov

Institution of Coal Chemistry and Materials Science, 650000, Sovetsky av., 18, Kemerovo, Russia.

E-mail: lesinko-iumx@yandex.ru

Abstract. The explosive decomposition of pentaerythritol tetranitrate (PETN) pressed pellets containing metal nanoparticles initiated by laser pulses was studied. The critical energy density values, which are the ones that make the sample to explode with 50% probability, were experimentally determined for PETN pellets containing nickel nanoparticles with three average radii initiated by the radiation of 1\textsuperscript{st} harmonics of neodymium laser. It was shown that the critical energy density for PETN-Ni samples anti-correlates with the linear absorption coefficient determined with the optoacoustic approach. The dependence of the critical energy density of PETN\textsuperscript{1}N samples containing aluminum nanoparticles on their weight percent concentration was measured using the influence of 1\textsuperscript{st} and 2\textsuperscript{nd} harmonics of the laser. It was determined that the minimum critical energy density for both wavelengths coincide while the minimum abscissae, providing almost equal linear absorption coefficient, values are different. The variation of dopants’ nanoparticles’ radius and concentration makes it possible to regulate the sensitivity of PETN to laser influence.

1. Introduction

There have been many works recently concerned on the regulation of energetic materials sensitivity to laser pulses influence. The practical aspect of the problem is linked to the development of new detonator types and explosives initiated by laser irradiation showing fewer drawbacks than contemporary detonators of other types. Such kind of detonators is essential, for instance, for application in military and special techniques [1]. In order to design the composition of new materials it is necessary to do a research into the mechanism of explosives initiation by laser pulses as it is studied incompletely. It is evident that the explosive’s initiation is coupled with the laser pulse energy consumption by the material. In our paper [2] the effectiveness of light absorption by metal nanoparticles of different radii in pentaerythritol tetranitrate (PETN) were calculated at different wavelength. This calculation results needs to be verified experimentally.

The aims of the present work are: (i) experimental measurement of linear absorption coefficient $k_{\text{eff}}$ of the laser irradiation for different radii of nickel nanoparticles and the link between $k_{\text{eff}}$ and the laser initiation threshold determination; (ii) Experimental verification of the explosion initiation threshold dependence on the concentration of aluminum nanoparticles with fixed size under the influence of first and second harmonics of neodymium laser.

1 To whom any correspondence should be addressed.
2. Experimental section
The samples were prepared from PETN powder with narrow size distribution, the average size was 1-2 μm. The three type of nickel powder were used as the additives, the particles’ size at the size distribution maximum was 280 nm (Ni(1)), 160 nm (Ni(2)), and 130 nm (Ni(3)). We also used the aluminum powder with the size in the distribution maximum 100-120 nm. The samples were pressed in a copper plate with thickness 1 mm having a round hole with diameter 3 mm at its centre. The samples’ density was ρ=1.77±0.03 g/cm³.

A YAG:Nd³⁺ laser operated in Q-switch mode was used as the initiation radiation source. The main (1064 nm) and the second (532 nm) harmonics of the laser were applied in the present study. The pulse duration fwhm was τ≈14 ns for the first and 12 ns for the second harmonics of the laser. The intensity distribution over the beam’s radius was nearly rectangular. The radiation was focused unto the sample surface with a lens. The beam diameter on the sample was 2.5 mm. The pulse energy was varied with a set of neutral light filters that attenuated the light intensity.

The dependencies of explosion probability on the pulse energy density were measured as follows. The samples were set on an aluminum plate. The samples’ surface was protected with an optical glass plate 1 mm thick that partly resisted the gas-dynamic response of the high-pressure zone forming after the irradiation. The explosion event was fixed if the laser influence was accompanied by a sharp sound, the experimental cell destruction and leaving of an imprint on the aluminum plate with the diameter equal to the sample’s one. The experimental explosion probability was plotted against the initiation energy density then. The initiation threshold was determined as the energy density at the explosion probability level 50%.

In another experimental series the similar samples were used in the absorption coefficient measurements with optoacoustic approach [3]. In that case the samples were set in the acoustic contact with a piezo detector made of CTS-19 ceramics. The irradiated surface was not protected with a glass plate. The pulse energy was 5 mJ which was enough for the measurable optoacoustic signals stimulation and was significantly less than samples’ destruction threshold. The signal of piezo detector was fixed with an oscilloscope LeCroy WJ332A.

3. Results and discussion
The influence of nickel nanoparticles’ size on the initiation threshold of PETN – Ni composite in the conditions of 1st harmonics of the neodymium laser irradiation was studied. The nanoparticles’ weight percent concentration was 0.1%. The critical energy densities of the laser pulse were measured for three samples containing nanoparticles with different sizes Ni(1), Ni(2), and Ni(3). The corresponding dependence is plotted on the fig. 1 (curve 1). One is able to notice that the optimal size of the nanoparticle exists which leads to the maximum sensitivity to the laser irradiation.
Fig. 1. The experimental dependencies of the critical laser pulse energy of PETN-Ni composite initiation $H_{cr}$ (1) and the effective absorption coefficient $k_{\text{eff}}$ (2) on the nanoparticles’ size at the size distribution maximum $d_{\text{max}}$.

Fig. 2. The critical laser pulse energy density of PETN-Ni composite initiation $H_{cr}$ dependence on the effective absorption coefficient $k_{\text{eff}}$ measured with optoacoustic method.

The absorption coefficient was measured with the optoacoustic approach using the same samples. The temporary acoustic signal curve registered by a piezo detector corresponds to the distribution of the thermal sources in the medium generated by the laser pulse, which is the foundation of the optoacoustic method [4]. The ascending exponential part of the experimental signal was used for the time-constant determination:

$$\tau_c = (k_{\text{eff}}c_0)^{-1}$$

where $\tau_c$ is the character time of the signal rise corresponding its $e$ times augmentation, $k_{\text{eff}}$ is linear absorption coefficient of light concerned on its absorption by nanoparticles, and $c_0$ is sound velocity in the sample. The measured $c_0$ value was $2500\pm200$ m/s in the experimental samples. The determined values of $\tau_c$ and $c_0$ were utilized to calculated $k_{\text{eff}}$ using equation (1). The resulting effective absorption coefficients are shown on the fig. 1 (curve 2). It is seen that the dependence $k_{\text{eff}}(d)$ is a non-monotonic function. The effective absorption coefficient one is able to write as

$$k_{\text{eff}} = \sigma n = Q_{\text{abs}} \sigma_g n$$

where $n$ is concentration, $\sigma$ is absorption cross section, $\sigma_g$ is geometric cross section, and $Q_{\text{abs}}(d)$ is efficiency of light absorption by metal nanoparticles in the PETN matrix that is a function of nanoparticles’ material, size and light wave length according to the calculation results [2] performed in terms of Mie theory. The dependence $Q_{\text{abs}}(d)$ is non-monotonic that correlates with the results depicted on the fig. 1.

Fig. 2 presents the same results as a $H_{cr}$ dependence on $k_{\text{eff}}$. The augmentation of linear absorption coefficient makes the critical energy density decrease providing that the nanoparticles’ concentration is constant.
A research into the influence of the nanoparticles' concentration on the critical energy density of PETN–Al composites was carried out. The composites' explosion was initiated with 1st and 2nd harmonics of the neodymium laser. As it was done in the previous cases the explosion probability dependence on the initiation energy density were measured. The nanoparticles' weight percent was in the range 0.025 – 1 %. The results are presented on the fig. 3. The both curves obtained at the wave lengths 1064 and 532 nm shows the minimum critical energy density values \( H_{cr} = 0.7 \text{ J/cm}^2 \) coinciding inside the experimental precision. However, the minimum point for 1st harmonics is observed at 0.2 weight % of nanoparticles while for 2nd – at 0.1 %. For that reason we can assume that the same value of \( H_{cr} \) means that

\[
k_{\text{eff.1}} = k_{\text{eff.2}}
\]  

(3)

The absorption effectiveness of the aluminum nanoparticles in PETN calculated at the wave lengths 1064 and 532 nm in [2] are shown on the fig. 4. The minimum points positions' give:

\[
n_1 = 2n_2.
\]

So, taking into account equations (2) and (3), an equation is expected

\[
\frac{Q_{abs1}}{Q_{abs2}} = 2.
\]

(4)

According to the fig. 4, the equality in formula (4) is provided for the nanoparticles’ diameter 120 nm, which matches the maximum position of the size distribution of the aluminum nanoparticles.

The following experiment was carried out in order to understand the dependence of \( H_{cr} \) on the nanoparticles’ weight percent concentration. A dependence of the optoacoustic signal amplitude, which is proportional to the pressure amplitude, was measured at constant pulse energy density using 1st harmonics of the neodymium laser. The resulting curve is plotted on the fig. 5. The pressure increases with the weight percent concentration of nanoparticles increase until the optimum value 0.2%. The consequent weight percent concentration increasing makes the pressure amplitude decrease.

The effect could be qualitatively explained as follows. According to [4], if the condition

\[
k_{\text{eff}} \tau < 1
\]

(5)

where \( \tau \) is laser pulse duration, is fulfilled, the instantaneous mode of laser excitation arises. The radiation energy consumed by the nanoparticles is converted into heat. The consequent temperature rise generates thermal stress and pressure increasing. The pressure jump appearing in the radiation
absorption layer propagates inside the sample as an acoustic pulse and is measured by a piezo detector. As long as the condition (5) is fulfilled, the pressure measured increases with the nanoparticles’ concentration increasing. The critical initiation energy density decreases in the same range (figs. 3 and 5). When an opposite condition \( k_{\text{eff}}c_0 \tau_i > 1 \) is met, the quasi-stationary mode of excitation evolves [4]. The surface layer having width \( x = k_{\text{eff}}^{-1} \), where the pulse energy is absorbed, has enough time to expand during the laser pulse in the case. As a consequence, the pressure amplitude diminishes and the critical energy density \( H_{\text{cr}} \) increases (figs. 3 and 5).

![Graph showing optoacoustic signal amplitude as a function of Al nanoparticles weight percent in the sample.](image)

Fig. 5. The optoacoustic signal amplitude as a function of Al nanoparticles weight percent in the sample.

In the direction of condition (5) meeting range the measurement of the effective absorption coefficient \( k_{\text{eff}} \) for optimal weight percent 0.2% of aluminum nanoparticles in the case of 1\textsuperscript{st} harmonics laser excitation was done with the optoacoustic approach. The value obtained was \( k_{\text{eff}} = 240 \text{ cm}^{-1} \). It means that \( k_{\text{eff}}c_0 \tau_i = 0.7 \). Thus the optimal concentration of nanoparticles (and corresponding \( k_{\text{eff}} \) value) matches the approximate boundary between the instantaneous and quasi-stationary modes.

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