Laser Initiation of PETN containing Nickel Inclusions

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Abstract. The spectral and kinetic characteristics of pentaerythritol tetranitrate (PETN) containing nickel nanoparticles glow initiated by laser pulses was studied with high temporal resolution. It was shown that glow which is chemiluminescence arises as a result of chemical reaction initiation. We suggest that the glow is concerned on excited nitrogen dioxide NO$_2$ luminescence. The reaction propagation leads to the explosion in the microsecond time range that is accompanied by thermal glow of the reaction products with temperature T=4300 K.

1. Introduction
Utilization of composite materials based on pentaerythritol tetranitrate (PETN) containing inclusions of metal nanoparticles in low concentrations allows one to diminish drastically the critical energy density of laser initiation of explosion [1-4]. This gives a principal way to application of such materials as explosives in detonators’ cups initiated with laser irradiation.

The dependencies of critical energy density of these explosives initiation with YAG-Nd$^{3+}$ laser ($\lambda=1064$ nm) operating in the Q-switch mode on the inclusions’ concentration with one typical size and on the size of Ni inclusion with definite concentration were studied in our works [3,5]. The minimal critical energy density corresponding to 50% explosion probability $H_{50}=0.7$ J/cm$^2$ was obtained for the samples containing 0.1 %wt nickel inclusions with average radius 160 nm.

It was experimentally and theoretically shown in [6,7] that the laser radiation is absorbed by inclusions. That leads to their heating up to evaluated temperatures. As many researchers believe the consequence is hot points formation where the self-sustaining chemical reaction propagates making the entire sample explode.

In order to give an insight into the primal chemical reactions, it is essential to apply the methods allowing one to observe the appearing products of PETN molecules decomposition with high temporal resolution starting from the laser pulse.

The present paper is devoted to the spectral and kinetic study of PETN polycrystals containing ultrafine Ni particles glow initiated with neodymium laser pulses.

2. Experimental section
The experimental samples were prepared from PETN powder with narrow size distribution. The typical grains size at the distribution maximum was $1 - 2$ μm. The dopant was the Ni powder with the size 160 nm at the distribution maximum. The Ni powder was synthetized by Roman Kolmykov at Kemerovo State University. The EDX analysis with spectrometer JEOL JED 2400 was used to estimate the nickel oxide content in the metal powder. Thus the nickel oxide weight percent was
determined as 12±1%. The procedure of powder mixing and samples pressing was thoroughly described in [8].

The samples were pressed in the hole with diameter 3 mm at the center of a copper plate with thickness 1 mm using a hydraulic press. This way the samples with typical density 1.7 g/cm$^3$ were obtained.

We used a YAG:Nd$^{3+}$ laser produced by Lotis that was operated in the Q-switch mode as an initiation source. The pulse duration fwhm was 14 ns, wavelength was 1064 nm and the energy density at the sample surface was 2 J/cm$^2$.

The main elements of registration systems were a polychromator and a streak camera based on an image intensifier tube (IIT). The sample’s glow arising during its irradiation with a laser pulse and explosion was directed to the spatial-temporal slit of the polychromator. The slit width was 0.1×0.2 mm determined the spectral and temporal resolution of the registering system. The spectral resolution achieved was 10 ns. The temporal resolution depends on the streak-camera scanning parameters. The polychromator decomposes the glow in the spectral range 350-750 nm. The spectral band width 0.2 mm falls onto the photocathode of the streak-camera. The streak-camera do the temporal scan of the spectrum. The glow on the output screen of the streak-camera is a matrix that vertical elements allows one to obtain the spectral dependencies of samples glow intensity at different time moments while horizontal ones give kinetics of glow intensity at a definite wavelength. This light matrix is read from the IIT screen with a PSD-matrix and transferred to a computer where the image numerical processing is carried out.

The spectral sensitivity of the optical registration system was calibrated with the standard lamp TRSh 2850-3000. The standard lamp has a definite color temperature with known emission coefficient thus its spectrum is readily calculable with the Plank equation. The calibration included the lamp spectrum registration and ration of measured and calculated intensity determination at the measurement conditions.

The spectrum restoration was performed by division of measured spectrum of the glow intensity with the sensitivity values at every wavelength at a definite time moment or at one wavelength at different time moments.

3. Results and discussion

The explosion glow intensity kinetics that is presented on the fig. 1 is almost identical in entire studied spectral range. The measurement with streak-camera using higher temporal resolution showed that the glow intensity rising in the first peak area (fig. 1) ceased after the laser pulse ending. Then one observes the intensity decreasing almost to background noise level during about 1 μs with following flash that takes 1.5 – 2.5 μs. The last flash of explosion glow is bimodal typically (fig. 1).

Fig. 1. The explosion glow intensity kinetics of PETN+Ni. 1 – $\lambda = 420$ nm; 2 – $\lambda = 600$ nm.  
Fig. 2. The glow spectrum of PETN+Ni at the end of the laser pulse.
The glow spectrum at the end of the laser pulse is shown on the fig. 2. A broad band with vibrational structure and maximum glow intensity at the wavelength 420 nm is observed on the spectrum. This glow could be associated with laser induced fluorescence, but it would demand the three-photon absorption at least, which is a highly improbable process. Moreover, at the laser pulse intensity used we have not notice any glow when the samples without inclusions were irradiated.

As it was shown in [6, 7], the laser irradiation is absorbed by metal inclusions mostly causing their heating. The estimated temperatures are higher than typical melting points of metals. At the same time a PETN layer is heated to the inclusion’s temperature. The layer with thickness could be estimated as

$$d = (\chi \cdot \tau_i)^{1/2}$$ (1)

where $\chi$ is thermal diffusivity of PETN, $\tau_i$ is pulse duration. Thus the PETN layer with thickness $d=40$ nm is heated to the temperatures above its melting point ($T_m=414.4$ K) or even PETN evaporation point on the pulse front. The high temperatures at the hot-spot makes the PETN molecules decompose in the heated layer surrounding the inclusion. The forming small molecules and radicals react chemically with each other and the reaction propagates. The measured glow spectrum shown on the fig. 2 could be assigned to the emission of an excited PETN fragment. We assume that this fragment is nitrogen dioxide NO$_2$.

The reaction development on the consequent stages causes the deep fragmentation of intermediates, heating of the reaction products and reaction propagation over entire volume of the sample ending with explosion. The spectra measured at the time moments 2, 3, and 4 (the time one may see on the fig. 1) are presented on the fig. 3. It is seen that at $t$ values about 0.5 $\mu$s the glow arises from the hot-spots still. At $t > 1.5$ the reaction propagates over entire sample that causes the explosion.

Fig. 3. The glow spectrum of PETN+Ni at . a – at the time moment 2 on the fig. 1; b - at the time moment 3 on the fig. 1; c – at the time moment 4 on the fig. 1.
The spectra shown on the fig. 3 are described satisfactorily in terms of Plank formula at the temperature 4300 K. The approximation results are shown as dotted lines on the fig. 3. Thus on the reaction propagation stage the thermal glow is predominant. We interpret this result as an argument in favour of thermal explosion mechanism.

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