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# Mechanism of hot spots formation in pentaerythritol tetranitrate under pulsed laser irradiation

**V I Oleshko[1], V P Tsipilyov[1], V E Zarko[2], A N Yakovlev and M A Mazur[1]**

[1]National Research Tomsk Polytechnic University, 634050, Tomsk, Russia
[2]Voevodsky Institute of Chemical Kinetics and Combustion, SB RAS, Novosibirsk

E-mail: oleshko@tpu.ru

**Abstract.** The spectral, kinetic, spatial, and amplitude characteristics of emissions of pressed samples of PETN (pentaerythritole tetranitrate) by the first harmonic of a neodymium laser (1064 nm, 12 ns) are studied. It is found that at the moment of irradiation of PETN by laser pulses, hot spots are formed on the surface of the samples. The energy density of laser radiation varies in the range of 0.5–3 J/cm$^2$. The characteristics of hot spots are determined by absorbing inhomogeneities and parameters of laser radiation. Multi-pulse excitation leads to the annealing of absorbing irregularities and increased threshold of optical breakdown. The received results are interpreted on the basis of ideas of the low-threshold optical breakdown as evolving within local (defective) regions of the PETN in the electric field of the light wave.

## 1. Introduction

Laser initiation of brisant explosives opens wide perspectives for the application of the explosion energy in various technological fields and blasting technologies. These technologies have been intensely studied over the last few years. However, the initial stages of PETN explosive decomposition triggered by pulsed laser radiation (LR) are still not studied. Recently, several alternative models of laser initiation of pure (without introduced additives) PETN specimens have been suggested [1–6]. The thermal spot model is now widely used, which states the impact of power laser radiation on an explosive to be accompanied by heating of local micro-regions (defects) termed as hot spots (HS), which leads to rapid chemical transformations. However, currently, there are no reliable experimental results validating this model.

The present work is aimed at elaborating immediate experimental evidence of hot spots formation in pure PETN samples under the impact of the first harmonic of neodymium laser and determining their formation mechanism.

## 2. Methods

The studied specimens of PETN ($C_5H_8N_4O_{12}$), sugar ($C_{12}H_{22}O_{11}$) and MgO were compacted in a steel mold under pressure P = 1.2·10$^8$ Pa into 6-mm tablets with the density of about 1.6 g/cm$^3$. The reasons to study sugar were its inertness and physicochemical properties being close to those of PETN. The mechanism of laser initiation of PETN was studied by the comparison of threshold, spacial, spectral and kinetic characteristics of the emission of the selected materials occurring under power laser impact.

The excitation source was Q-switching YAG:$Nd^{3+}$ laser at fundamental frequency ($\lambda$ = 1.06 μm) with pulse width at half maximum of 16 ns. The detailed scheme of the laser testbed for the investigation of explosive decomposition of energy-yielding materials is presented elsewhere [7]. A part of LR was deflected by glass plates to the calibrated IKT-1N calorimeter (Russia) and FEK photoreceiver (Russia) to measure energy, duration and shape of laser pulses. The density of the pulse energy (*H)* on the surface of sample was measured by QE25LP-S-MB pyroelectric energy meter (Gentec-EO, Canada) and varied using calibrated glass filters in the range of (0.1–6) J/$cm^2$. The laser beam was focused by a lens (with focal length of 0.3 m) into a 2-mm spot and incident on the plane of the open surface of sample at the angle of 75°. The experiments were performed in atmospheric air. The luminance kinetics of the excited target was recorded by H5773-04 photosensor (Hamamatsu, Japan) and DPO 3034 oscilloscope (Tektronix, USA). To record the spacial distribution of luminance along the irradiated surface of the targets, the testbed was supplemented by a microscope. The luminance induced by pulsed irradiation of the samples was captured by Sony Alpha DSLR-A500 (Sony, Japan) via MPD-10 microscope (Russia). The channels of the photomultiplier tube and photoelectric colorimeter were synchronized by the pulse of CdSe crystal two-photon luminescence excited by the first harmonic of Nd-laser. The time resolution of the measurement system was ~10 ns, while the spacial resolution was ~10 μm.

## 3. Results and Discussion

It was experimentally shown that the irradiation of pure PETN samples by the laser pulse with *H* = (0.5–3) J/$cm^2$ is accompanied by the formation of bright spots on the background of less intense diffused luminance (Figure 1a).

(a) (b) (c)

**Figure 1.**Microphotograph of surface luminance of PETN (*a*), sugar (*b*) and MgO (*c*) under the first laser irradiation pulse. *a*) *H* = 3 J/$cm^2$; *b*) *H* = 2.5 J/$cm^2$; *c*) *H* = 3.5 J/$cm^2$.

A typical oscillogram of local region (HS) luminance (upper oscillogram) in Figure 2 was obtained simultaneously with photo capturing. The lower oscillogram in Figure 2 is the pulse of Nd-laser generation. The data on spacial distribution of HSs along the irradiated PETN surface, luminance kinetic and LR oscillogram were obtained after primary LR pulse. The repeated irradiation of the surface of sample by a sequence of pulses decreased the number of HSs and their luminance intensity, i.e. HSs were annealed. The increase in *H* from 0.5 to 3 J/$cm^2$ for irradiation of the untreated surface of the specimen increases the number of HSs, the intensity and duration of their luminance after the first excitation pulse. The HS luminance duration at pulse half maximum increases from 20 to 36 ns.

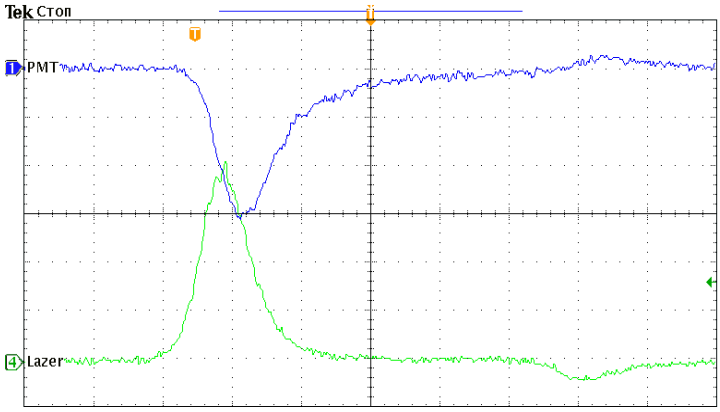
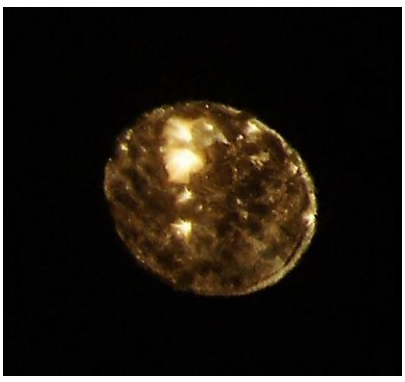


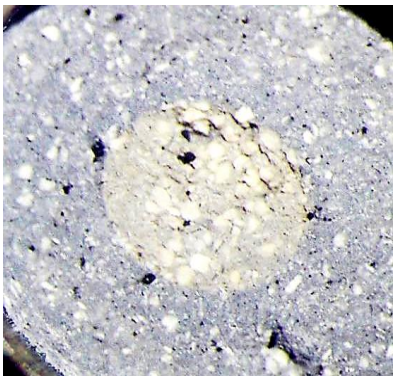

**Figure 2.** Oscillograms of hot spot luminance in PETN after the first irradiation pulse (upper) and excitation laser pulse (lower), H = 3 J/cm$^2$.

There was no damage to the PETN sample surface after the irradiation. At $H > 4$ J/cm$^2$, on the regions of the brightest HSs there are microcraters with the size of ~100 μm. Interestingly, according to early studies [1], the crater formation threshold ($H^*$) in compacted PETN samples after irradiation by the first harmonic of Nd-laser depends on the PETN dispersity and varies in the range of ($4 \geq H^* \geq 9$) J/cm$^2$ with the dispersity changing from 3200 up to 12,000 cm$^2$/g. The authors of [1] have associated $H^*$ with the threshold of PETN optical breakdown recorded by the luminance of the laser torch and intense sound (click). However, that work lacks data on the processes taking place in PETN at $H < H^*$.

To understand the physical nature of HS luminance occurring in PETN under pulsed irradiation, additional experiments were performed using the inert samples (compacted tablets of sugar and MgO). During the studies, we have established that in the inert materials and at the energies of ($0.5 \geq H \geq 3.5$) J/cm$^2$, the regularities of formation and annealing of HSs are similar to those in PETN. All the above was confirmed experimentally and the results are presented in Figs. 1b and 1c. The results state unified character of formation and annealing of HS under pulsed irradiation of compacted samples. One can assume that HSs originate in solid bodies due to localization of LR energy on absorbing irregularities. According to [8], one of such irregularities in the compacted samples of pure PETN are carbon microparticles. To check this assumption, we have studied aged (stored for 20 years) PETN samples with 0.1 wt% of carbon. The photo images of the specimens demonstrated dark microparticles (assumably, the agglomerates of carbon particles) with the dimensions from 10 to 150 μm. We have established that at the moment of pulsed irradiation, HSs are formed in the locations of microparticles which dimensions exceed those of the particles (Figure 3a). After LR action, one can observe a light patch on the irradiated surface of sample along with the microparticles. Assumably, it is caused by the burning of soot particles having the dimensions of less than 10 μm (Figure 3b).



(a)



(b)

**Figure 3.**Microphotographs of hot spot luminance formed in PETN subsurface region with addition of 1 wt% of carbon (*a*) and surface of sample after single irradiation (*b*).

The combination of received experimental data (nonuniform surface luminance structure with spots, delayed HS luminance maximum in relation to the laser pulse, their increased luminance duration at increased laser radiation energy density, annealing effect) and analysis of literature on the effect of absorption irregularities on the optical breakdown threshold of various materials [9–10] allows concluding that the reason of HS formation in pure PETN samples and studied inert materials is low-threshold optical breakdown evolving in local (defective) regions of solid bodies in the electric field of the light wave. The host spots are subsurface laser plasma that forms after the evolution of the optical breakdown in the vapor of absorbing irregularities. The discovery of the nature of the absorbing irregularities responsible for the initiation of the optical breakdown in the studied samples requires additional investigation. We assume that one of the origins of absorbing irregularities are impurities introduced into the specimens after the compaction in the metal mold. It is confirmed by the authors of [11] who have established the absence of HSs for the energy densities of $0.16 < H < 2.5$ J/cm$^2$ in sugar samples prepared in a glass cell. Separate microzones of white luminance, in this case, were detected only for $H \leq 2.5$ J/cm$^2$.

## 4. Conclusion

The studies have demonstrated that the excitation of the compacted samples of studied materials by the first harmonic of Nd-laser causes formation of bright hot spots (local micro- and macrozones of white luminance) forming after the development of low-threshold optical breakdown on absorbing irregularities. The role of absorbing irregularities is played by microparticles introduced into the subsurface region of the samples during their compaction in a metal mold. Multi-pulse excitation of the samples leads to the evaporation of absorbing impurities (annealing of defects) and increased threshold of optical breakdown.

Thus, we can conclude that the HS formation mechanism in the samples of pure PETN under LR pulse is electrical, similarly to the irradiation of explosives and inert materials by high-current electron beam [12–13]. Under laser action, the electrical breakdown develops in the subsurface region of the sample on absorbing irregularities in the electric field of the light wave, while under electron irradiation it happens in the zone of electron beam breakage, in the electric field of volumetric negative charge injected into the sample. Finally, the received results in addition to the scientific value bear practical interest, because the significance of the nature and parameters of the absorbing irregularities present in energy-yielding materials and possibility of their annealing under sequential irradiation enable the adjustment of the explosive material sensitivity to laser radiation.

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