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Absorption of pulsed laser radiation by composites based on hexogen and aluminium nanoparticles

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Abstract. **Absorption of the second-harmonic radiation from a pulsed YAG : Nd^{3+} laser by composites based on hexogen and aluminium nanoparticles (of a diameter 100 nm) is theoretically and experimentally investigated. The absorption efficiency factor (the ratio of the light absorption cross section to the geometrical cross section) is calculated by using the Mie theory for Al inclusions in a hexogen medium for various particle diameters and wavelengths corresponding to laser fundamental and second-harmonic radiation. Dependences of the absorption coefficient and amplitude of an acoustic signal on the concentration of inclusions under laser irradiation of composite samples are measured by using a piezo-acoustic transducer. It is shown that the radiation is directly absorbed by nanoparticles. The optimal concentration is found, which provides the maximal signal of the piezo-acoustic transducer.**

Keywords: *hexogen, aluminium nanoparticles, composite materials, laser radiation, radiation absorption, optoacoustic.*

1. Introduction

Explosion and detonation phenomena have been studied for more than hundred years [1 – 5]. One direction in these investigations is explosive decomposition of matter under the action of laser radiation, which started in the 1960s [6]. For researches, of particular interest was the development of the materials sensitive to laser radiation, which can be used as detonators of conventional blasting agents (BAs) [7, 8]. One of the ways to enhance sensitivity of a normal BA to laser radiation is fabrication of composites with light absorbing additives included into the BA. Such works started in the 1980s [9, 10], but those did not continue later.

In the last decade, we have actively investigated pentaerythritol tetranitrate (PETN) composites with inclusions of metals or carbon materials. More than 20 papers have been published (see, for example, [11 – 18]). It was shown that at the optimal concentration of nanodiperse-size metal particle inclusions, for example Al, at the fundamental radiation of a pulsed neodymium laser, the index k_{eff} of extinction on particle inclusions reaches ~200 cm^{-1}, and the radiation energy is absorbed in a layer of thickness $h = 1/k_{\text{eff}} \approx 50$ μm [12]. In this case, in a time inerval of pulse action the temperature of metal inclusions enhances and an exothermic chemical reaction starts with a release of additional heat in a layer adjacent to the inclusion particle, that is, a ‘hot point’ is formed [18]. At a sufficient number of ‘hot points’, a shock wave is formed, which propagates to a non-irradiated part of the sample and finally leads to explosion [18]. However, as in our works, so and in works of other authors, PETN is used as a matrix of a composite material. For establishing similar and different features of blast laser initiation in various composite materials it is reasonable to study materials with other matrices, for example, hexogen (RDX), which is transparent for laser radiation and is less sensitive to all kinds of action than PETN. At a first stage, it is necessary to perform theoretical calculations and experiments concerning a pre-blast regime. In the present work, we present investigation results for possible absorption of laser radiation by RDX – Al composites. The latter not only are interesting themselves, but also allow one to further optimise experiments with blast laser initiation in composite materials.

2. Investigated objects and experimental methods

The initial material was an RDX powder with a narrow granulometric distribution. At the distribution maximum a particle size is 1 – 2 μm. The other component is Al powder of Alex type with a particle diameter at the distribution maximum of 100 nm. All particles have an oxide envelope of thickness ~5 nm, which is transparent for the laser emission. The method for preparing samples with various Al content is generally similar to that for PETN – Al samples, which is thoroughly described, for example, in [11]. The samples were pressed to a hole at the centre of a copper plate (sample holder) by a hydraulic press. While preparing samples, the pressure increased to 1.8 GPa for 60 min. Finally, samples with a density of 1.80 ± 0.02 g cm^{-3} were obtained.

Laser initiation was realised by using second harmonic radiation (λ = 532 nm) of a YAG : Nd^{3+} laser, operated in a Q-switched regime. The FWHM pulse duration was 14 ns. Experiments were performed in the pre-blast regime; therefore, the pulse energy density was 30 mJ cm^{-2}, in which case sample destruction under the pulse action was minimal. However, such an action is enough for reliable signal detection by a TsTS-19 ceramic piezotransducer similar to one employed in [11, 17]. A detected signal from the piezotranstucer passed to a LeCroy 332 WJ oscilloscope. The experimental method is thoroughly described in [18].

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3. Results and discussion

Earlier, basing on the Mie theory we have developed a method for calculating the factor of electromagnetic radiation absorption efficiency Q_{abs} (the ratio of the light absorption cross section to the geometrical cross section) for metal nanoparticles in a PETN matrix at various particle radii and radiation wavelengths. The method is thoroughly described in [19, 20].

In the present work, we calculate dependences of the absorption efficiency factor on particle sizes at the wavelengths λ corresponding to a fundamental and second-harmonic radiation of a neodymium laser by the method described in [19, 20]. According to [21], the refractive indices for aluminium are 0.884–6.470i and 1.226–10.041i at $\lambda = 532$ and 1064 nm, respectively. The refractive index for RDX was found by extrapolating the dependence obtained experimentally in [22] in the spectral range of 2.5–18 μm. At the same wavelengths, the refractive index of RDX was 1.594 and 1.593, respectively. The calculation results are presented in Fig. 1.

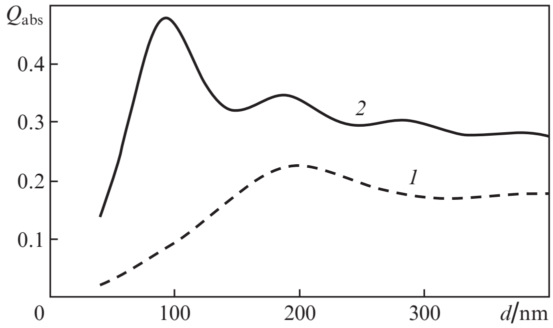

Figure 1. Calculated efficiency factors of electromagnetic radiation absorption by aluminium nanoparticles as functions of particle diameter d at λ = (*1*) 1064 and (*2*) 532 nm.

We used an aluminium powder of Alex type with a particle size of 100 nm at the distribution maximum. From calculations follows that the value of Q_{abs} for the laser second harmonic radiation is four times greater than that for the fundamental radiation (Fig. 1). Thus, all further experiments with the RDX–Al composite were carried out by using second harmonic radiation of the laser.

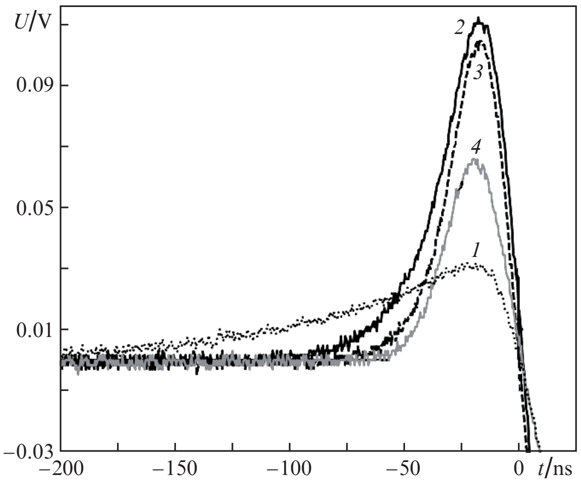

Figure 2. Oscillograms of an acoustic response to laser irradiation of hexogen with the aluminium concentration of (*1*) 0.05, (*2*) 0.2, (*3*) 0.3, and (*4*) 0.5 mass %.

Acoustic responses to the laser action on samples with the aluminium contents of 0, 0.025, 0.05, 0.1, 0.2, 0.3, 0.5, and 0.7 mass % were studied. In samples without aluminium inclusions, an acoustic response is not detected within the device sensitivity range. Hence, the RDX material weakly absorbs the laser radiation. Samples with inclusions exhibit well pronounced signals. For example, typical oscillograms of such signals are shown in Fig. 2. As in [18], the time start point corresponds to the instant of the pulse compressed stage transfer to the stretching stage. The time interval from 0 to −20 ns corresponds to a transducer transient response while passing from the compressed pulse phase to the stretched one. The pulse part in the range from −20 to −200 ns presents a distribution of heat sources over the sample depth [23] and is well described by the exponential dependence with the time constant

$$\tau_{\mathrm{a}} = (k_{\mathrm{eff}} c_0)^{-1}, \qquad (1)$$

where c_0 is the speed of sound in the medium.

The speed of sound in a medium was found experimentally from the time interval between the primary and reflected signals and was 3000 ± 200 m s^{-1}.

Experimental oscillograms of samples with various inclusion concentrations n were used for measuring τ_{a} and calculating k_{eff} by formula (1). Calculation results are shown in Fig. 3. One can see that the dependence of k_{eff} on the inclusion concentration is linear. Earlier, we obtained a similar dependence for PETN with Al inclusions in [12]. Basing on formulae for scattering media from [24–26], we have shown [12] that in the case of a linear dependence of k_{eff} on the inclusion concentration the absorption is mainly related to the inclusions whereas a contribution of radiation scattering in the matrix into k_{eff} is negligible.

A dependence of an acoustic signal intensity in an RDX-Al composite on the Al concentration is presented in Fig. 4.

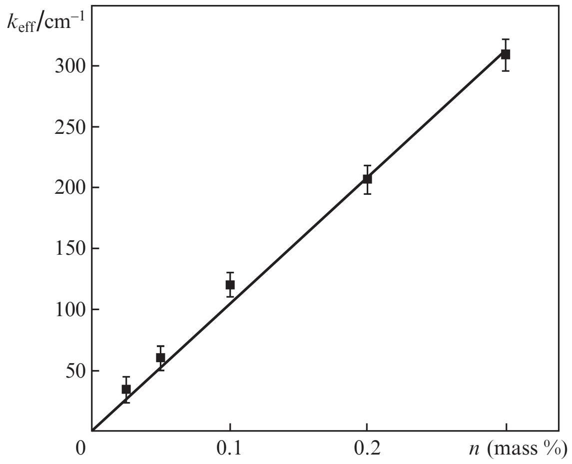

Figure 3. Extinction index k_{eff} vs. inclusion concentration n for hexogen–aluminium composites.

Similar dependences we obtained for acoustic signal intensities in PETN–Al [12] and PETN–Fe [27] composites. It was found that the concentrations n with the maximal optoacoustic signal amplitudes correspond to the minimal thresholds of explosive decomposition under laser initiation [12, 27]. This effect was explained in [13]. The essence is that at the inclusion concentrations from zero to $n_{\text{opt}} = 0.2$ mass %, the laser irradiation regime is adiabatic, and the layer of thickness $h = k_{\text{eff}}^{-1}$ in which sample heating occurs in a result of pulse energy absorption has no time to expand. At n above 0.2 mass %, k_{eff} accordingly increases (with a correspondent reduction of layer thickness h). The wavelength of an acoustic wave $\lambda < \tau_{\text{p}} c_0$ (τ_{p} is the laser pulse duration) becomes comparable with the absorbing layer thickness h. Due to the acoustic unload, already during the pulse irradiation the radiation absorbing layer expands more extensively at higher n. Thus, at $n > n_{\text{opt}}$, a transfer from adiabatic to quasi-stationary expansion occurs [23]. As a result of irradiation, a pressure in the irradiated layer falls; hence, the acoustic signal amplitude also reduces (Fig. 4). As shown in [12, 27], in explosion-regime experiments with the inclusion concentration exceeding n_{opt}, the threshold of blast decomposition increases. Thus, data presented in Fig. 4 allow one to find the optimal concentration n_{opt} corresponding to the minimal threshold of blast decomposition, which will reduce labour expenditures in finding this threshold for RDX–Al composite.

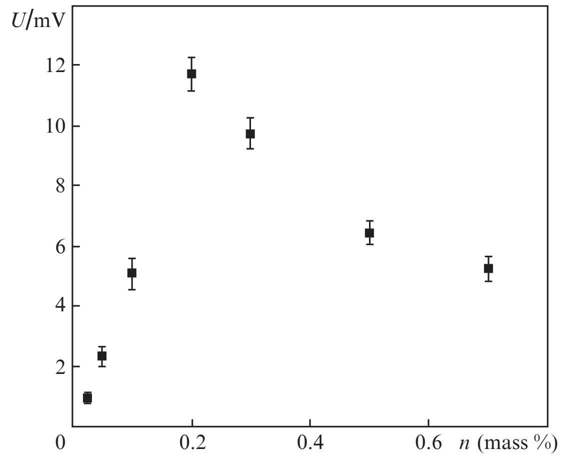

Figure 4. Acoustic signal amplitude under the action of second-harmonic laser radiation on the RDX–Al composite vs. concentration of Al inclusions.

4. Conclusions

1. Radiation absorption efficiency factors Q_{abs} at $\lambda = 532$ and 1064 nm are calculated for Al inclusions in a RDX medium for various particle diameters, from which follows that it is more appropriate to carry out further experiments with composite blast initiation by using the second harmonic of a neodymium laser.

2. It is shown that in an RDX–Al composite, second-harmonic radiation of a neodymium laser is mainly absorbed by Al particles.

3. Study of a piezetransduser signal amplitude as a function of the inclusion concentration yields the optimal concentration $n_{\text{opt}} = 0.2$ mass % at which the minimal threshold is predicted for blast decomposition of RDX–Al under the action of second-harmonic radiation of a neodymium laser.

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