The effect of aluminum particles dispersity on characteristics of ammonium perchlorate—aluminum composition laser ignition

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Abstract. Ammonium perchlorate–aluminum compositions taken in stoichiometric ratio were ignited in air and under the cover with 1.06-µm 0.8-ms-long laser pulses. The ignition energy thresholds were measured for samples at various dispersity of Al. The causes of the difference in examined compositions sensitivity to the influence of laser radiation is considered from the perspective of the thermal theory.

1. Introduction

It is known that pyrotechnic mixtures based on ammonium perchlorate (AP) and aluminum can be used to produce pyrocartridges and other ignition means. Laser system can be successfully applied for that kind of devices in the function of igniter [1, 2]. For that reason, it is necessary to understand what characteristics of the mixture affects the ignition process.

It was previously ascertained that ignition characteristics of such mixtures, on top of everything else, can depends on the size of Al particles [3]. The sensitivity to laser pulse (pulse duration 4 ms) comparison between AP mixtures with ultrafine (UF) Al (size of one particle $d = 0.25$ µm) and coarse Al ($d = 80$ µm) was conducted. The energy density threshold (ET) was used as the criterion of ignition. The ET of mixture AP/coarse-Al appears to be significantly higher than ET of AP/UF-Al.

It has been assumed that mixture sensitivity decreases with increasing the size of Al particles. To verify that assumption it is necessary to carry out the experimental research with greater numbers of mixtures of different Al particle size.

2. Materials and methods

In this work we studied the ignition of a stoichiometric AP/Al compositions by laser radiation pulse (wave length λ=1.06 µm) with pulse duration $\tau_p = 0.8$ ms. Analytical-grade ammonium perchlorate was milled in an agate mortar and sieved through a Capron sieve with a mesh size of ~70 x 70 µm. Al powders with average particle size of 0.14; 3; 4; 9 and 80 µm was mixed with AP.

Ultrafine aluminum powder with a mean diameter of 0.14 µm was prepared by electric explosion. It contained less than 93 wt. % of active aluminum. The particles had a spherical shape and a log-normal size distribution [4].

The mixtures samples of ~ 9 mg weight had been placed into a polymethyl methacrylate (PMMA) capsule having 3 mm inner diameter and 3 mm depth. The samples were pressed in the PMMA

capsule up to the pressure of 800 kgf/cm². The end surfaces of the both open and covered samples were exposed to laser radiation. The covered samples were tested in the absence of gas-dynamic unloading as shown in figure 1.
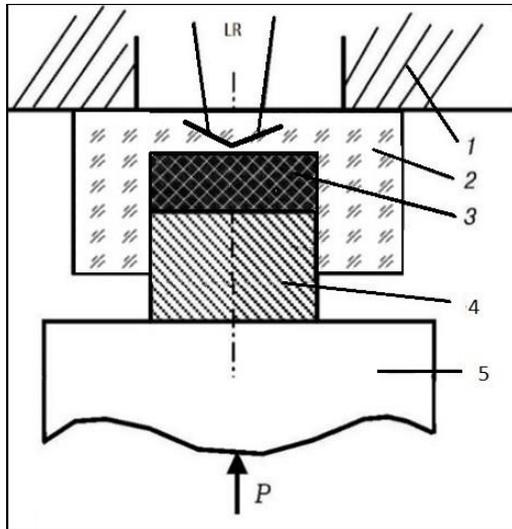

Figure 1. Cross-sectional view of covered sample 1 – Upper press stop; 2 – PMMA capsule; 3 – mixture sample; 4 – lower press stop; 5 – press forcer.

The neodymium laser (wavelength λ=1.06 μm) was used for defining the ET of each sample (table 1). It generates the quasicontinuous laser pulse with the modulation depth not higher than 10 % and duration of 0.8 ms. Laser beam diameter is 2 mm. Non-uniformity of the irradiation was not over than 10 %.

The experimental installation for the laser ignition is schematically presented on figure 2. Further details about installation described in works [5, 6].
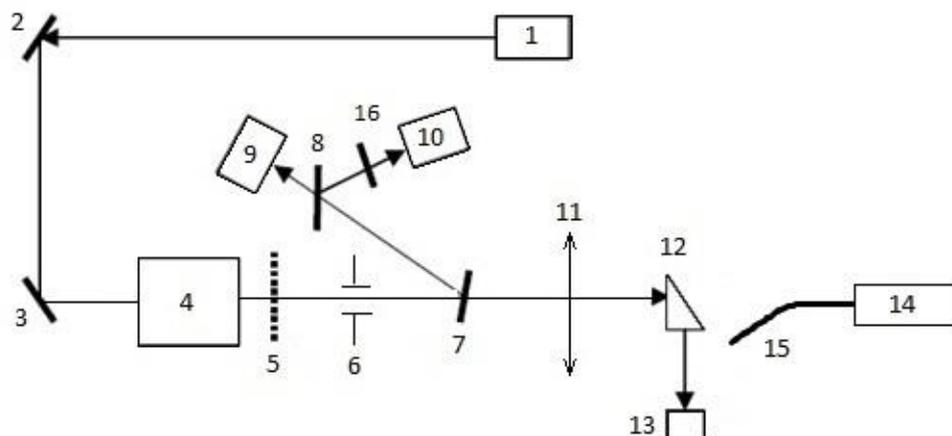

Figure 2. The scheme of the laser ignition experiment of pyrotechnic mixtures: 1 – alignment laser; 2, 3 – rotary mirrors; 4 – pulse laser block; 5, 16 – neutral filters; 6 – diaphragm; 7,8 – beam splitters; 9 – calorimeter; 10, 14 – photodetectors; 11 – lens; 12 – rotary prism; 13 – sample; 15 – fiber optic cable.

The ignition thresholds were determined from the initiation function curve (figure 3). Firstly, the dependence $P(E)$ within the range of 0–1 was obtained, where P is the ratio between the number of successful ignitions and all attempt; E is the energy density of laser radiation. Each point of this function contains 25 experiments with statistic range distribution less than 10 % by the calorimeter

results. The ignition threshold H_{50} (or H_{TR}) is a value for which ignition took place with possibility $P = 0.5$.
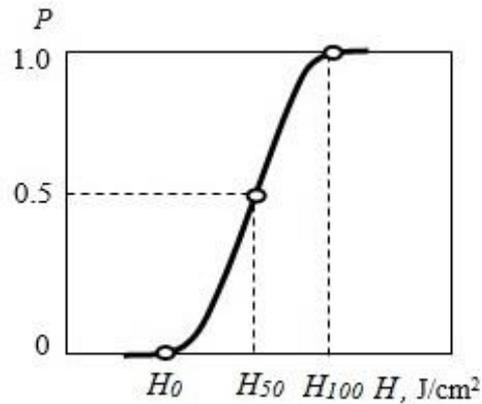

Figure 3. The function of the ignition threshold.

3. Experimental results
AP/UF-Al samples with opened surface burned out with bright and more than 10 cm tall flame jet. The ET is 1.3 J/ cm^2 and the ignition delay amount to 15 ms. That results corresponding the obtained one from the work [3].

From all examined mixture types only AP/UF-Al was able to ignite in case of open surface.

The other mixtures with coarse Al showed no substantial activity even with increase of the energy exposure up to 40 J/cm^2. However, the plasma jet appears on $H \geq 3$ J/ cm^2.

In case of closed surface all mixtures burned out along with blast sound and disruption of PMMA capsule. The ignition delay did not exceed laser pulse duration and decreased with energy level rising. ET are listed in the table 1.

4. Discussion
So the AP/UF Al with open surface is more active than other mixtures that based on coarse Al. In case of covered surface there is an opposite situation. Furthermore, covered samples sensitivity increases with decreasing Al particle size.

To interpret such results, it is necessary to analyze the heating of Al particle and the matrix during the duration of laser pulse. For that purpose, it is enough to be confine by the rough estimate of heating temperature. The two different approaches should be considered.

4.1. Isolated particle approach
It is possible to assume some microzones with isolated from each other Al particles. The particle heating process taking place without heating influence of surrounding Al particles.

In that context the hot spot appears in AP matrix near the Al particle, even in case of high concentration of Al (mixtures AP/UF-Al and AP/Al-10 at table 2).

A coarse estimate of the hot spot temperature ΔT for each particle size during laser pulse τ_u can be found from equation:

$$\Delta T = \frac{F(\rho_0) H_{TR} \cdot 3}{4 \cdot c\rho \cdot R_0} \cdot k(R_0, \lambda_0) \cdot \frac{R_0^3}{(R_0 + \sqrt{\alpha\tau})^3} \tag{1}$$

$F(\rho_0)$ is the coupling coefficient between light intensity on and under the surface; H_{TR} is the energy density threshold; ρ_0 is the diffusion reflectivity coefficient; ρ is the AP density; c is specific heat; α

is thermal diffusivity of AP matrix; $k(R_0, \lambda_0) = \dfrac{\sigma(R_0, \lambda_0)}{\pi R_0^2}$ - is relative cross-section of absorption at wavelength λ_0; $\sigma(R_0, \lambda_0)$ is the absorption cross-section. According to [3] coefficient ρ_0 equals to 0.35, while value of ρ_0 the coupling coefficient is $F(\rho_0) \approx 3$. For Al particles of considered size range $2R_0$ relative cross-section of absorption is $k(R_0, \lambda_0) \approx 0,1$. According to [7] α take a value of 10^{-3} cm^2/s. The results of temperature calculations ΔT_{oz} for different mixtures are listed in the table 1.

Maximum temperature of the hot spot (3500 ºC) corresponds to maximum particle size $2R_0$ = 80 µm (AP/Al-1 mixture). Whereby hot spot size is $2R = 2R_0 + 2\sqrt{\alpha\tau}$, take a value about 100 µm. The heating temperature and the enthalpy W ($W = F \cdot H_{TR} \cdot k \cdot \pi \cdot R_0^2$) decreases along with the particle size $2R_0$ decreasing.

So if $2R_0$ = 80 µm then enthalpy is about 3 mJ and for $2R_0$ = 0,14 µm (2R ~ 10 µm) is 3×10^{-10} J, in other words the difference is about 6 orders of magnitude.

The main paradox of that approach is about hot spot with low enthalpy actually are more active than hot spot with much higher enthalpy. That defies the conceptual issues of thermal theory and the common sense.

Hence, the experimental results can't be interpreted from the position of microzone thermal ignition theory. It follows that ignition from the single hot spot, that appears around isolated singe Al particle in such mixtures, is not possible.

Table 1. Energy thresholds and calculation results.

Types of mixture	Typical size Al particle $2R_0$, µm	Energy density threshold for ignition with covered surface H_{TR}, J/cm^2	Energy density threshold for ignition with uncovered surface H_{TR}, J/cm^2	Heating temperature of hot spot around the particle ΔT_{oz}, ºC	Hot spot enthalpy W, J
AP/Al -1	80	153	—	3500	~ 3 × 10^{-3}
AP/Al -4	8-10	16.9	—	228	~ 4 × 10^{-6}
AP/Al -8	3-4	11.8	—	86	~ 5 × 10^{-7}
AP/Al -10	2-3	8.3	—	35	~ 8 × 10^{-8}
AP/UF-Al	0.14	3.3	1.3	0.25	~ 3 × 10^{-10}

4.2. Near surface layer approach
The other limiting case has no dependence from sample structure (AP crystal size or Al particle size). Laser radiation is heating the subsurface layer with the depth $Z_l = \dfrac{1}{\mu} + \sqrt{\alpha\tau}$, μ is the absorbance index of mixture. Absorbance index can be found from equation:

$$\mu = \pi R_0^2 \cdot k(R_0, \lambda_0) \cdot C \qquad (2)$$

C is Al particle concentration ($C = \dfrac{\eta \cdot \rho_c}{M_{pat}(1-\eta)}$; η is the weight percentage of Al particles; ρ_c is the mixture density; M_{pat} is the mass of one Al particle). The value of μ-index for different mixtures are listed in the table 2. The temperature of heated layer can be found from:

$$\Delta T_l = \frac{F \cdot H_{TR}}{Z_l c \rho} \tag{3}$$

$F = 1 - \rho_F$ is absorption coefficient due to Fresnel reflection. The values of heated layer temperature are listed at table 2.

Table 2. The results of layer heating calculations.

Types of mixture	Typical size Al particle $2R_0$, μm	Al particle concentration, particle /cm^3	Layer absorbency index μ, cm^{-1}	Thickness of the heated layer ΔZ, cm	Heated layer temperature ΔT_l, $^{\circ}$C
AP/Al - 1	80	$\sim 2 \times 10^6$	9	~ 0.1	765
AP/Al - 4	8-10	$\sim 1 \times 10^9$	74	$\sim 1 \times 10^{-2}$	845
AP/Al - 8	3-4	$\sim 5 \times 10^9$	185	$\sim 6 \times 10^{-3}$	983
AP/Al - 10	2-3	$\sim 1.5 \times 10^{10}$	370	$\sim 4 \times 10^{-3}$	1037
AP/UF-Al	0.14	$\sim 3 \times 10^{13}$	3700	$\sim 1 \times 10^{-3}$	1650

It can be seen that the surface layer temperature increases with decreasing Al particle size alone with decreasing the thickness of heated layer ΔZ.

Nevertheless, the reactivity of the flat form hot spot with heating temperature 1650 $^{\circ}$C and layer thickness about 10^{-3} cm (AP/UF-Al mixture) is significantly higher than the reactivity of the another hot spot with heating temperature 765 $^{\circ}$C and layer thickness about 10^{-1} cm (AP/Al-1 mixture). This situation corresponds to classic ideas of thermal hot spot ignition and in particular, it corresponds the critical Frank-Kamenetskii parameter [8].

Thus, the increasing mixtures sensitivity along with increasing Al particle size points at thermal macrozone nature of AP/Al mixtures ignition. According to that theory the ignition begins from the hot spot limited by laser beam size, light attenuation depth and the thermal front range into the sample.

The ignition tests of the opened surface samples with coarse Al were not successful. That situation is similar to explosives with gas reaction phase [10]. But experiment result of AP/UF Al mixture ignition invalidate such idea because of lower threshold with open surface rather then covered. That fact points at possibility of condensed phase reaction.

From that position it is easy to explain the sensitivity difference of opened and closed surface of AP/UF-Al samples. In case of opened surface, the additional heating taking place due to Al particles oxidizing by atmospheric oxygen and nitrogen. The oxidation process appears due to high Al particles temperature (about 1650 $^{\circ}$C). Perhaps, for mixtures with AP and coarse Al the heated layer temperature is not enough for effective Al particle heating.

In that case the surface closed by transparent dielectric material improves the reaction condition through increasing the reaction velocity along with the pressure into the reaction zone. It can be confirmed by the ignition delay reduction in case of closed surface ignition in comparison with opened surface (figure 4).
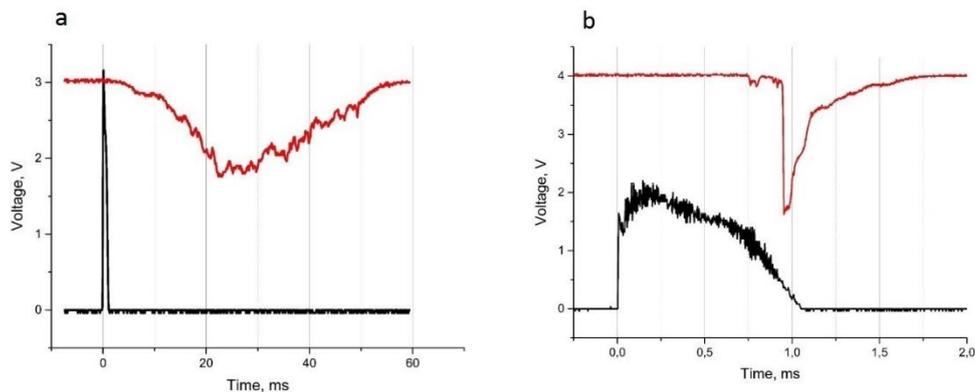
Figure 4. Typical AP/Al mixture ignition oscillogram for a) opened surface; b) closed surface. Upper curve corresponds to the glow of combustion products and lower curve is the laser pulse signal.

5. Conclusions

The energy thresholds for pulse laser ignition of stoichiometric AP/Al mixtures were measured. The mixtures distinguished in the Al particles size. The samples surface was irradiated with and without transparent dielectric.

For laser pulse duration $\tau_p = 0.8$ ms the mixture sensitivity increases with decreasing the specific size of Al particles in the range from 80 µm to 0.1 µm.

The AP/Al mixture ignition begin from the hot spot limited by laser beam size, light attenuation depth and the thermal front range into the sample in a time of laser pulse duration.

The absorbance index, temperature and enthalpy increases along with Al particle size which leads to increasing mixture sensitivity. That parameters were determined to estimate the hot spot reactivity.

It has been suggested that the presence of oxygen and nitrogen facilitate the ignition process only for AP/UF-Al mixture in case of opened surface. The influence of atmospheric oxygen and nitrogen on AP/Coarse Al mixture ignition is not substantial.

In this work the mixtures wherein the AP Crystal size excels the size of Al particles were examined. The mixtures with equal components dispersity should be considered at the next work.

Acknowledgments

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