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Laser Monitor for Studying the Combustion of Thin Layers of Metal Nanopowders

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Abstract—In this paper, we propose a laser monitor with a horizontally located observation area for studying laser initiation and combustion of thin layers of metal nanopowders. Three configurations of the optical scheme with different inputs of igniting laser radiation and different magnifications are considered. Visualization of combustion of a 0.4 mm layer of aluminum nanopowder demonstrated the possibility of studying the surface of a nanopowder thin layer during combustion using a laser monitor. The bright glowing of the sample and the bright radiation of the igniting laser do not interfere with the imaging of the surface. The proposed system allows us to study surface changes caused by the propagation of combustion waves. It is demonstrated that in the region of laser initiation, combustion proceeds in one-stage, and combustion products are formed during laser action. Outside the initiation area, combustion proceeds in two stages. The results reveal the prospects for designing a laser monitor for studying the combustion of thinner layers of metal nanopowders.

1. INTRODUCTION

One of the modern areas of research and technology is nanotechnology related to the combustion of materials [1–3]. Combustion of metal nanopowder foils is no less interesting area of research than the study of combustion of relatively massive samples [4]. In contrast to the combustion of massive samples, in which combustion is filtration, and the oxidizer spreads through the sample, when burning thin layers, the oxidant is accessed from the surface. The nature of combustion propagation over the layer surface is of interest, including initiation, smoldering, and inflammation. In the case of aluminum nanopowder, smoldering, in fact, is the propagation of the first combustion wave, and inflammation is the appearance and propagation of the second combustion wave.

Knowledge of the regularities of the combustion of powder material layers is important both for solving technological problems associated with materials production by the method of high-temperature combustion and for assessing their fire hazard and determining their application areas in construction design of buildings and structures [5]. A thin layer of nanopowder can serve as a model for studying the heat balance at the combustion surface, in particular, the processes of ignition of large and small particles [6]. In this regard, it is important to develop appropriate techniques for studying the combustion of multilayer materials and thin layers. The use of such systems can be useful for studying the combustion of multilayer nanofilms, whose combustion differs from systems of a similar composition obtained by mixing nanopowders [3].

Combustion of metal nanopowders, in particular, mixtures of aluminum-based nanopowders, proceeds at high temperatures and is accompanied by a bright glow, which makes difficulties to the observation of the sample surface. Passive direct observation methods using cameras and light filters

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allow high-speed video recording of the processes [7–9], including with microscopic magnification [10]. High-speed imaging is an essential component of much of today's combustion research. At the same time, observation of the surface during heating and combustion with microscopic magnification is of interest for understanding the physical and chemical processes and regularities of combustion [11, 12]. The use of microscopic systems to study combustion opens up additional opportunities in comparison with the study of macroscopic parameters of flame propagation [10]. High-speed visualization systems based on brightness amplifiers and high-speed video cameras — laser monitors — are used in diagnostics of fast processes [13]. Such systems are built on the principle of a laser projection microscope based on copper or copper bromide gain medium [14–16]. In [17], we demonstrated the possibility of using a low-intensity laser monitor to study the combustion of aluminum nanopowder. In [18], the possibility of observing laser ignition of aluminum nanopowder using such a system was demonstrated. Works [7–9] were devoted to the use of laser monitors based on copper bromide vapor active medium for studying various aluminum-containing mixtures of nano- and micron metal powders. In the previous works [7–9, 17–19], we observed the side surface of a massive sample (up to 3 g) located vertically.

The combustion of thin layers of metal nanopowders has not previously been visualized using laser monitors. At the same time, visualization of surface changes under a bright luminous layer simultaneously with the observation of the propagation of a bright luminous layer over the surface can provide interesting and new data on the combustion process of thin layers of metal nanopowders. In this regard, the aim of this work was to develop a laser monitor for studying the surface of metal nanopowder thin layers placed horizontally and to test it when studying the combustion of a thin layer of aluminum nanopowder during combustion.

2. EXPERIMENTAL DETAILS

During the experiments, aluminum nanopowder in the form of a thin layer with 0.35–0.4 mm thickness was placed on a ceramic substrate. The layer was compacted by rolling a 100 g metal roller. We used the aluminum nanopowder obtained by the electric explosion of a metallic wire under argon atmosphere [20]. The nanopowder was characterized by the following activity parameters: temperature of oxidation start, 450°C; degree of oxidation, 63.8%; maximum oxidation rate, 0.13 wt%/°C; specific heat effect, 4995 J/g. The particle size distribution was measured using a Shimadzu Nano Particle Size Distribution Analyzer SALD-7101 in isopropanol (99.99%). The distribution was near lognormal with the maximum of 80 nm. The content of metallic impurities in the powder did not exceed 0.2 wt%.

A laser monitor for observing thin layers of metal nanopowders differs from that for observing massive samples. To study horizontally placed layers of aluminum nanopowder, it is necessary to illuminate the sample from above. Because of the design features of metal vapor active elements, their placement in a vertical position is practically impossible; therefore, the laser monitor scheme [18] was modified. Fig. 1 presents schematically options for the arrangement of the elements of the laser monitor scheme for horizontal observation of combustion. The main difference from the systems that we used in previous works is a part of the experimental setup where the object is placed, and the image is formed. All variants are monostatic schemes, in which a beam-deflecting mirror is installed to direct the beam of the brightness amplifier in the vertical direction. In the variant in Fig. 1(a), we used a lens 5 with 50 mm focus. A beam-deflecting mirror was located between the lens and the object. The emission of the laser monitor illuminated the area of the object ~ 4.5 mm in diameter. LAPSUN USB-HDMI Microscopic Camera with Canon Macro Lens EF 180 mm and a bandpass filter was installed to record the general view of the nanopowder layer combustion. The sharpness of images in all schemes was adjusted using a vertical linear translator.

The options of the optical path of a laser monitor for horizontal observation with greater magnification are presented in Figs. 1(b) and (c). Helios-44M objective lens ($F = 58$ mm) was located above the object of study, while the radiation of the brightness amplifier was rotated by a beam-deflecting mirror. The emission of the laser monitor illuminated the area of the object ~ 1.5 mm in diameter. The difference between the schemes in Fig. 1(b) and Fig. 1(c) is the input of igniting laser radiation. In case (b), lasing is coupled similarly to case (a) by placing a beam-deflecting mirror in close proximity to the object of observation. In case (c), the igniting laser radiation is focused on the object surface by the same objective lens, which forms an image of the laser monitor. Since the sharpest image is obtained

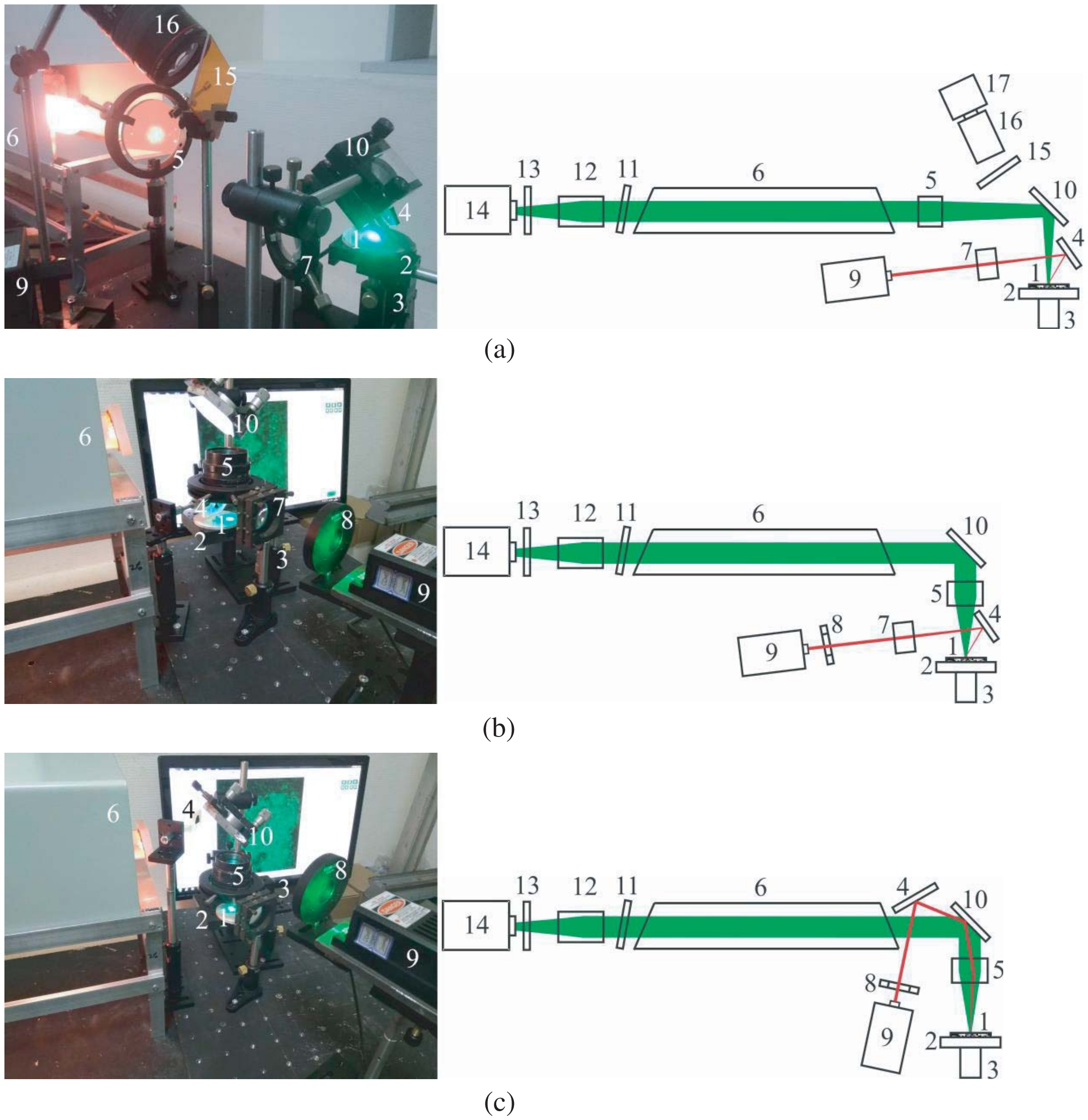


Figure 1. Schemes of the experimental setup. (a)–(c) Various implementations of laser monitors. 1 — nanopowder on a substrate; 2 — horizontal linear translator; 3 — vertical linear translator; 4, 10 — beam-deflecting mirrors; 5, 12, 16 — objective lenses; 6 — brightness amplifier; 7 — lens; 8 — mechanical shutter; 9 — igniting laser; 11 — neutral light filter; 13 — narrow-band filter 510 ± 5 nm; 14 — high-speed camera; 15 — band-pass filter; 17 — USB-camera.

when the object is located near the objective lens focus, the power of the igniting laser beam focused by the lens is sufficient to initiate combustion. In this regard, one of the tasks of this work was to test the possibility of using such a combined scheme. Fig. 2 presents the images of test objects obtained using various imaging schemes. A steel ruler with 0.5 mm gradations and a microscopic calibration ruler with 10 μm gradations were used as test objects. The spatial resolution of the scheme in Fig. 1(a) was 12.5 μm, and the resolution of the schemes in Figs. 1(b) and (c) was 5 μm.

In this work, we used a compact laboratory-made copper bromide brightness amplifier previously applied in [19]. The gas-discharge tube of the brightness amplifier had an aperture of 3 cm and the length of the active area of 60 cm. The temperature parameters of the tube operation were provided by highly stable (±1°C) independent heating of the active area and containers with copper bromide. The active medium was pumped by a thyratron pulsed power supply with a pulse repetition frequency of 20 kHz.

(a) (b) (c)

Figure 2. Test-objects images. (a), (b) Laser monitor in Fig. 1(a); (c) laser monitor in Fig. 1(b).

The amplified spontaneous emission (ASE) of the gain medium with a total average power of 20 mW at wavelengths of 510.6 and 578.2 nm illuminated the object under observation. The illumination power was measured at the site where the object was placed. The illuminating power density was 1.3 mW/mm^2 for the scheme in Fig. 1(a) and 11.3 mW/mm^2 for the schemes in Figs. 1(b) and (c). The experimentally determined ignition-threshold value for the aluminum nanopowder was about 250 mW/mm^2. Thus, the influence of the brightness-amplifier emission on the combustion process in our work can be neglected.

A high-speed camera Phantom Miro C110 captured the laser monitor images. The camera was operated in an external sync mode to provide synchronization with the ASE pulses. The frame rate of the camera was equal to 500 fps at a resolution of 1024 $\times$ 1024 pixels and an exposure of 10 µs. The actual exposure in the system was equal to the ASE duration of the brightness amplifier $\sim$ 30 ns. To ensure the recording mode, in which the image is formed by one pulse of the brightness amplifier ASE, synchronization was carried out using a two-channel pulse generator Aktakom AWG-4122. The generator formed synchronized pulses with frequencies of 500 Hz and 20 kHz, which were fed, respectively, to the sync input of the high-speed camera (500 Hz) and the input of the external trigger of the pulsed power supply. Synchronization ensured the exposure of each frame recorded by a high-speed camera by one pulse of the brightness amplifier emission. The advantage of this mode of imaging was discussed earlier in [19].

Ignition in the scheme in Fig. 1(a) was provided by a semiconductor laser with a power of 2 W in a continuous mode with a wavelength of 660 nm. The laser had an external trigger for controlling the radiation power, which made it possible to set the duration of exposure. In these experiments, the exposure time was 0.4 s. Ignition in the schemes in Figs. 1(b), (c) was provided by a solid-state laser with a power of 0.2 W in a continuous mode with a wavelength of 532 nm. A mechanical shutter Thorlabs SHB1 with external triggering was used to control the ignition. The exposure time was $\sim$ 1 s. The high-speed camera was triggered simultaneously with the igniting laser switching-on.

3. RESULTS

The combustion of a thin layer of aluminum nanopowder differs significantly from the combustion of a massive sample [7–9]. Fig. 3 presents the direct imaging frames of nanoAl thing layer combustion recorded by the USB camera in Fig. 1(a). The red spot in the center of the images is the igniting laser beam in standby mode ($\sim$ 1 mW), except for the moment of time 0.33 s, which corresponds to the igniting laser power of 2 W providing combustion initiation. After initiation, the propagation of combustion seems disordered. During propagation, the bright combustion spots branch into several parts, which then also branch. Bright spots spread over the surface corresponding to a high-temperature combustion wave. Typically the combustion of aluminum nanopowder in air proceeds in two stages with a relatively low (600–800°C) and high ($>$ 2000°C) temperatures [1, 2], which are clearly identified when observing the combustion of massive samples.

Figure 4 presents a typical view of the initial aluminum nanopowder layer and combustion products. After combustion, the sample has an inhomogeneous structure. Some of the nanopowder did not react

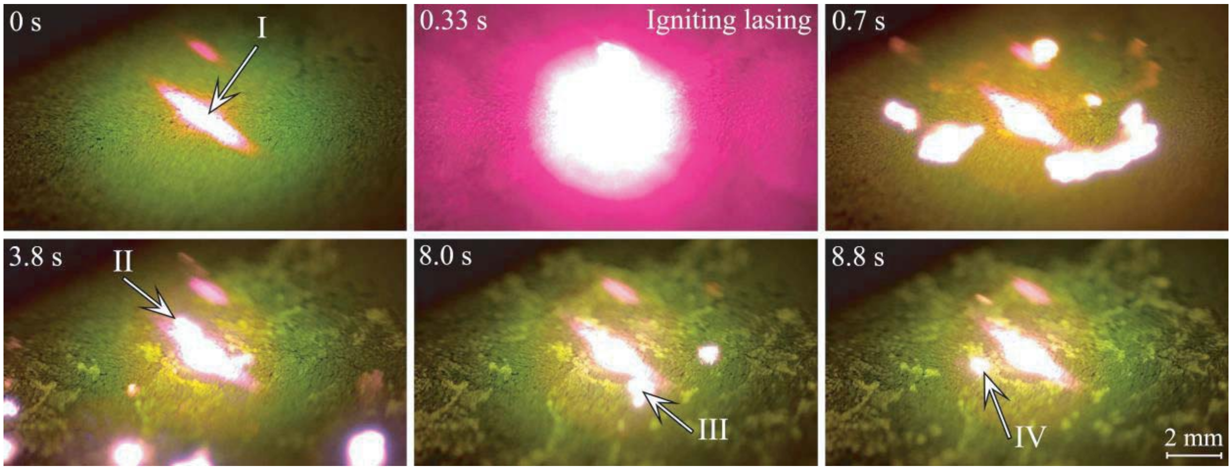
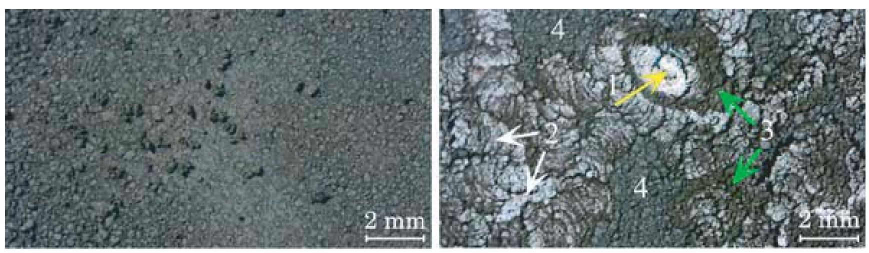


Figure 3. Combustion of a thin layer of aluminum nanopowder. I — area of laser action, II–IV — areas corresponding to those marked in Fig. 5.



(a) (b)

Figure 4. Typical view of the surface of the aluminum nanopowder layer. (a) Before combustion; (b) after burning out. 1 — area of laser action; 1, 2 — combustion products; 3 — unreacted aluminum; 4 — remains of the original aluminum nanopowder.

at all; this part has the original color. White combustion products AlN, Al_2O_3, Al_5O_6N alternate with unburned dark products, which are darker than the original. Dark products are agglomerates of unreacted aluminum, formed during the first wave of combustion because of heating. Probably, there was not enough time and energy to heat the nanopowder for ignition and transition to the high-temperature stage of combustion. Thus, in the case of thin layers of aluminum nanopowder, the first combustion wave is not always accompanied by a second high-temperature wave.

The bright glowing of the burning nanopowder, on the one hand, gives information about the nature of the process, on the other hand, does not allow observing surface changes in the combustion area. In particular, it is impossible to identify the propagation of the first wave of combustion; it seems that the combustion proceeds in one stage. The proposed laser monitors (Fig. 1) make it possible to observe the surface through bright glowing. In this case, the high-speed camera does not record the high-temperature glowing of the burning sample and the lighting of the igniting laser. The gain medium on copper bromide vapors performs narrow-band amplification of radiation and operates as an active band-pass filter. The choice of the appropriate scheme depends on the required magnification and the size of the observation area.

Imaging with high magnification does not allow observing the complex behavior of combustion propagation over the surface due to the small size of the observation area. The scheme in Fig. 1(a) with the 50 mm lens provides a large enough observation area of $3 \times 3\,\mathrm{mm}^2$, making it possible observing both the area of laser ignition and the propagation of combustion waves from the ignition area. Fig. 5 presents the frames of high-speed imaging of a thin layer combustion of aluminum nanopowder, obtained using this scheme. Frames in Fig. 3 and Fig. 5 represent the same process; time points and details (I–IV) correspond to each other. A laser monitor with a relatively large observation area allows us to observe the intermittent nature of the combustion. Intermittent combustion results in nonuniform combustion products. Comparison of the images allows us to conclude that the bright fragments in Fig. 3 correspond to the propagation of the second stage of combustion of aluminum nanopowder, during which white combustion products 2 (AlN, Al_2O_3, Al_5O_6N) are formed. The propagation of the first combustion wave, which is not noticeable when being directly observed (Fig. 3), is clearly identified using the laser monitor (relatively dark region 3 in Fig. 5).

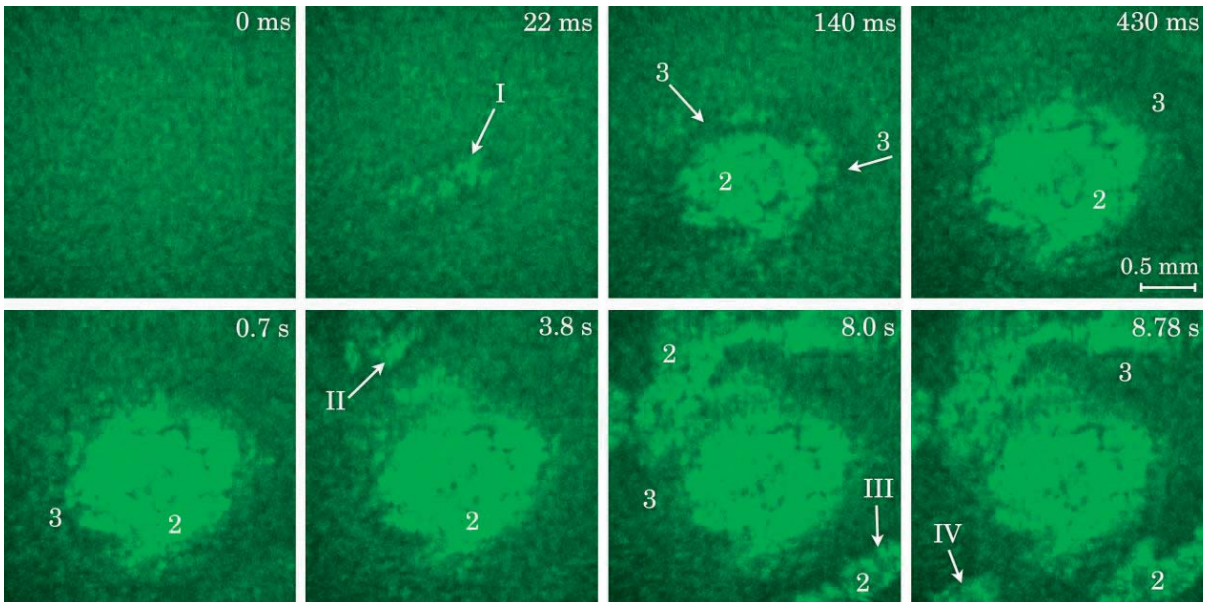


Figure 5. Frames of the high-speed imaging of nanoAl thin layer combustion obtained at the area of laser ignition using the scheme in Fig. 1(a). I — area of laser action; II–IV — areas corresponding to those marked in Fig. 3; 2 — combustion products/second combustion wave, 3 — first combustion wave.

The laser monitors in Figs. 1(b) and (c) allow us to study the initiation and propagation of combustion with higher magnification. Fig. 6 presents the frames of high-speed imaging of the combustion of an aluminum nanopowder thin layer, when the initiation was carried out at the edge of the observation area. It follows from the presented frames that the combustion process proceeds in two stages. In the ignition area, combustion proceeds in one stage, then the high-temperature combustion stops for a while (time between 62.5 and 304 ms), while the low-temperature wave continues to propagate along the sample, which is clearly seen in the dynamics in the video. The presence of a liquid phase at this time indicates the propagation of combustion inside the sample. After an induction period (304 ms), the second stage of combustion comes to the surface again.

The frames of imaging of the combustion of a thin layer of aluminum nanopowder in the area of laser action using the scheme in Fig. 1(c) are presented in Fig. 7. The presented frames confirm the observation results presented in Fig. 6 that combustion in the initiation area proceeds in one stage. The propagation of the combustion wave from the point of laser action to the sides is clearly visible. The combustion front is identified as a dark stripe in front of the highly reflective combustion products, visually light. From the images it is possible to determine the width of the combustion front (pointed by segments 2–5 in Fig. 7), which is 47–100 μm under experimental conditions. A significant variation in the width of the combustion front indicates the inhomogeneity of the combustion process.

The advantage of the scheme in Fig. 1(c) is the simplicity of alignment of the initiation and observation areas. In the scheme of Fig. 1(b), it is relatively difficult to provide initiation in the center of the observation area. We believe that this design will be useful when laser initiation is investigated using short focus objective lenses.

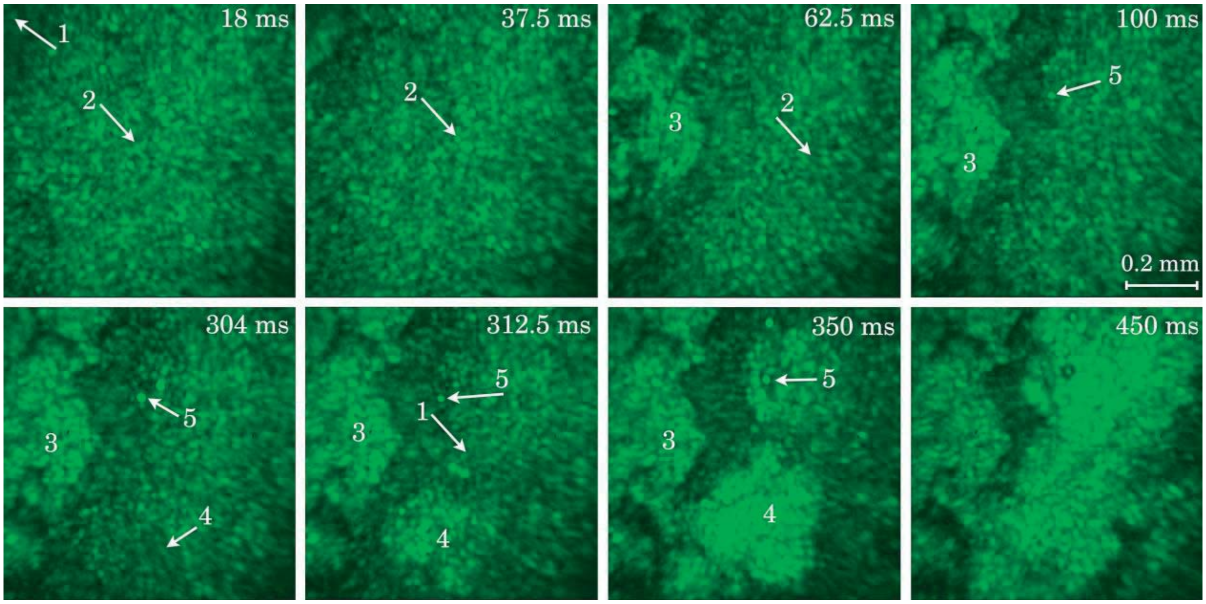


Figure 6. Frames of the high-speed imaging of nanoAl thin layer combustion obtained using the scheme in Fig. 1(b). 1 — area of laser action; 2 — front of the first combustion wave; 3, 4 — combustion products/second stage of combustion; 5 — liquid phase.

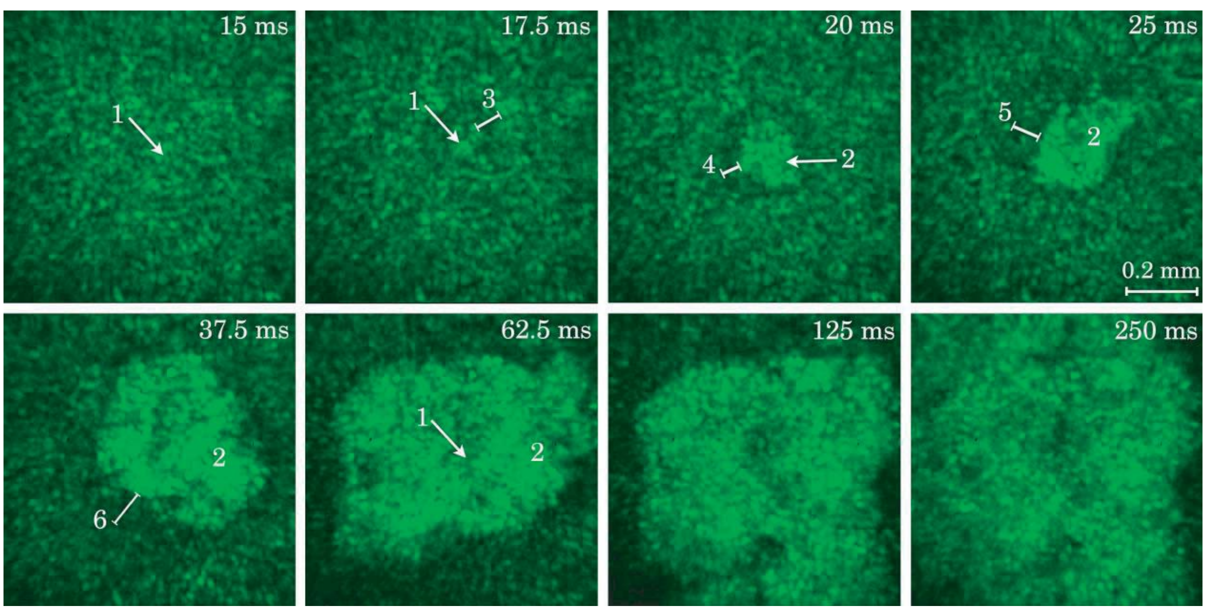


Figure 7. Frames of the high-speed imaging of nanoAl thin layer combustion obtained at the area of laser ignition using the scheme in Fig. 1(c). 1 — area of laser action; 2 — combustion products; 3–6 — combustion wave front.

4. CONCLUSION

The paper proposes the laser monitor for studying the combustion of thin layers of metal nanopowders. In contrast to laser monitors previously used to study the combustion of massive metal nanopowder samples, the emission of the brightness amplifier and the initiating lasing are directed to the object from above. The use of an appropriate lens allows a small imaging area with high magnification or a large imaging area with relatively low magnification to be obtained. A brightness amplifier based on a copper bromide vapor medium with a low power of amplified spontaneous emission was used in this work, which minimizes the effect of the visualization tool on the object of study. The use of additional passive video recording makes it possible to compare the surface changes observed by the laser monitor with the visible bright glowing of the surface in this area.

Three options of a laser monitor for horizontal surface imaging were proposed, which were used to study the combustion of thin layers of aluminum nanopowder. The system made it possible to observe the inhomogeneity of combustion propagation over the surface of the sample, the formation of combustion products and areas with partial combustion of the nanopowder. In these areas, only the first combustion wave passes through, and there is not enough energy to go to the second stage. In the area of laser initiation, combustion proceeds in one-stage, and combustion products are formed during the laser action. Outside the initiation area, as in the combustion of massive samples, combustion proceeds in two stages.

A combined scheme of the laser monitor was used for the first time for aluminum nanopowder combustion imaging, in which an igniting laser beam was fed into the objective lens (focus 58 mm) that forms an image. The system alignment allowed focusing the radiation of the initiating laser in the observation area of the laser monitor. This implementation makes it possible to use objective lenses with a shorter focal length in the future. The results reveal the prospects for designing a laser monitor for studying the combustion and laser heating of thinner layers of metal nanopowders such as metal nanopowder foils and multilayer materials. The promising applications of such devices might include in situ imaging of laser-driven integration of metal nanopowders into polymers [21] and other applications.

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