Study of synthesis dynamics of ceramic materials nanopowders at different processing modes by ultrafast laser pulses

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Abstract. The paper presents a developed experimental setup for the synthesis of spherical nanoparticles using different processing modes by exposure of femtosecond laser pulses in an argon atmosphere. Using the combined processing approach leads to both an increase in ablation process efficiency and an increase in the dispersion of particle size of the resulting fraction as a result of coagulation of ablation products due to prolonged stay in the discharge plasma. When processing is carried out in an electrostatic field, there is a close to linear growth in ablation process efficiency in the investigated range of laser pulse energy. In this case, compared to the treatment with only a laser beam without an electrostatic field the dispersion of particle size of the resulting fraction is reduced. The difference in efficiency between these modes is about 70% at pulse energy of 145 μJ.

1. Introduction

The problem of efficient synthesis of metallic, composite, ceramic powder materials is of exceptional relevance in connection with the development of both powder metallurgy and the emergence of new approaches to the production of high-quality ceramic materials, including optical ones. The main problem in creating high-quality ceramics is the creation of materials for pressing, such materials are powders of the initial components (oxides, fluorides, etc.). The problem of obtaining spherical nanopowders is relevant in the direction of creating transparent laser ceramics [1-4]. Granules of these powder materials should have a minimum size to ensure high surface activity, have a low porosity of the resulting workpiece and isotropy of the properties of the resulting material. The main requirements for such powders are a high bulk density, this parameter is achieved through the use of spherical granules with a size of not more than 100 nm. The powder used should not contain hard agglomerates. The fractional composition of this powder must provide a bulk density of at least 0.7 of the density of a similar crystalline material. The synthesis of spherical nanopowders is a rather difficult task due to the high activity of the resulting granules and the tendency to form agglomerates. Nevertheless, when using laser ablation processing, it is possible to synthesize this material [5-7]. One of the effective methods for the synthesis of highly dispersed powder metal, composite, and ceramic materials with a narrow particle size distribution, perfect spherical shape is the method of materials dispersion using laser radiation [8].

The paper presents a developed experimental setup for the synthesis of spherical nanoparticles using different processing modes by exposure of femtosecond laser pulses in an argon atmosphere,
conclusions are drawn about the effectiveness of each of the processing modes used.

2. Experimental setup and processing modes
The developed experimental setup for the synthesis of spherical nanoparticles is an isolated metal chamber with optical window for inputting laser radiation, window for observing the processes occurring in the affected area. The chamber contains electrical conductor entry units into the internal volume, channels for the introduction of various media in which the surface of materials is processed [9]. The processing took place under the following conditions: scanning with a laser beam in two axes was carried out by a galvo scanner system equipped with a flat-field lens with a focal length of 200 mm source of laser radiation was femtosecond Yb:KGW – laser system (wavelength 1029 nm, pulse width 280 fs, pulse repetition rate of 10 kHz, pulse energy 55-150 μJ). Moving the sample along with the reaction chamber along the vertical axis and setting the plane of the processed sample to the focal plane of the focusing system was carried out using a motorized linear translator. The inner volume of the working chamber was filled with highly pure argon through a leak, which is located on the chamber body.

The treatment of aluminum oxide was carried out under normal conditions in an argon atmosphere: only by laser radiation, by the combined action of laser radiation with an electric arc discharge and by laser radiation in an electrostatic field. The processing mode is common for all 3 experiments: pulse width 280 fs, pulse repetition rate of 10 kHz, pulse energy 55-145 μJ, the processing area is 8x1 mm, the scanning density is 30 lines/mm, the scanning speed is 10 mm/s.

A schematic representation of the developed experimental setup is shown in figure 1.

Figure 1. Schematic representation of experimental setup for the synthesis of spherical nanoparticles: (a) - laser radiation combined with an electrostatic field; (b) - laser radiation combined with an arc discharge.
The intensity of the electrostatic field is 5-15 kV/cm. The need to change the intensity is due to the adhesion of ablation products to the electrodes during prolonged exposure and the formation of an arc discharge in the electrode gap. An electrostatic field was used to remove the ablation products from the laser beam propagation area and collect them (figure 1 a). Ablation products, acquiring a charge, are deposited on the positive and negative electrode. Ablated particles have a speed of the order of tens to hundreds of meters per second, which makes it possible to exclude repeated interaction of the laser beam with nanoparticles. This method is convenient for collecting laser ablation products. During processing, the resulting granules do not stick together and do not deform, in this case there is no need to use filtration systems.

The electric arc discharge was created by a high-voltage pulsed source (25 kV/cm), with a generation frequency from 0 to 200 Hz. The discharge was carried out through the air gap between the tungsten electrode and the stage through the sample (figure 1 b). The localization of the electric arc was carried out using a laser-induced plasma channel. The focusing area was located at an equal distance from the electrodes. Scanning rate and pulse repetition rate are the same. In the interaction of a sub-picosecond laser study with a medium, the formation of a laser-induced plasma channel is observed, the conductivity of which is tens of orders of magnitude higher than that of a gas under normal conditions. The formation of an electric arc and its propagation occurs strictly along the area of spreading of the laser beam, which allows for combined processing.

As a result of the experiments performed using different processing modes, spherical nanomaterials were obtained, which will be discussed in more detail below.

3. Results and discussion

SEM images of aluminum oxide based oxide ceramics powder materials obtained in argon atmosphere under different processing modes and histograms of nanoparticle distribution is shown in figure 2.

Figure 2. SEM images of the obtained aluminum oxide nanoparticles and histogram of the nanoparticle size distribution: (a) - laser ablation mode; (b) - laser treatment in an electrostatic field; (c) laser - treatment combined with an arc discharge.
The difference in the dispersion of granules, depending on the processing mode, can be explained by the duration of the thermal effect on the ablation products. During laser ablation treatment, particles ejected into the vessel space can be repeatedly exposed to laser radiation and a laser-induced plasma torch, which leads to their fusion among themselves (figure 2 a). The use of an electrostatic field allows the particles to be removed from the propagation region of the laser radiation until the next laser pulse arrives, which also helps to reduce the laser erosion torch (figure 2 b).

When processing combined action of laser radiation with an electric arc discharge, particles ejected during ablation serve as conductors for the arc drain, at the same time increasing the intensity of luminescence of both the laser-induced plasma torch and the electric arc discharge (figure 3 c). Based on the results obtained, it can be concluded that the use of the electrostatic field allows to remove the ablation products from the laser radiation propagation region without compromising the surface quality and dispersion of the resulting micro and nanopowders. When a high-intensity electrostatic field is used, the scattering of ablated particles takes on a predetermined character. Ablation products, acquiring a charge, are deposited on high-voltage electrodes. To confirm the assumption of an increase in the efficiency of laser ablation, we estimate particle ejection rate from the propagation region of laser radiation. It is necessary to determine a number of conditions: the distance that the particle must travel to get out of the probable propagation region of laser radiation, we take 0.5 mm; the time for which the particle must leave the specified region of 100 μs (the time interval between laser pulses at laser repetition rate of 10 kHz).

Figure 3 shows graphs of the dependence of the weight of removed material (aluminum oxide) on pulse energy of laser radiation and the presence of an electrostatic field.

![Figure 3. Graphs of the dependence of the weight of removed material (aluminum oxide) on pulse energy of laser radiation and the presence of an electrostatic field.](image)

In the figure 3, red triangles indicate the processing mode using an electrostatic field, gray dots indicate the processing mode by laser exposure without using an electrostatic field. As can be seen, the use of an electrostatic field has a beneficial effect on the overall dynamics of the laser ablation process. The graph shows that an increase in the pulse energy starting from 55 μJ to 95 μJ in 5 μJ steps is accompanied by an increase in the weight of the removed material. With a further increase in pulse energy, a significant decrease in efficiency is observed when using the processing mode without an electrostatic field, and for the mode using an electrostatic field, the weight of removed material continues to grow (when the pulse energy reaches 145 μJ, the difference in efficiency between these modes is about 70%).
4. Conclusion
As a result of the experiments, it was found that the laser-induced plasma channel has a sufficient concentration of free electrons, as well as conductivity for dynamic localization of a pulse-periodic electric arc discharge during laser ablation processing. Using the combined processing approach (exposure to laser radiation together with electric arc discharge) leads to both an increase in ablation process efficiency and an increase in the dispersion of particle size of the resulting fraction as a result of coagulation of ablation products due to prolonged stay in the discharge plasma. This approach can be effectively used in erosional surface processing or in the laser ablation synthesis of nanomaterials which are subject to low uniformity requirements of the fractional composition.

When processing is carried out in an electrostatic field, there is a close to linear growth in ablation process efficiency in the investigated range of laser pulse energy. In this case, compared to the treatment with only a laser beam without an electrostatic field the dispersion of particle size of the resulting fraction is reduced. For the synthesis of nanomaterials of a given fractional composition, it is effective to use an electrostatic field combined with exposure to laser radiation. Laser processing of the material in an electrostatic field allows make it possible to increase the ablation process efficiency by removing ablated particles from the area of laser radiation propagation until the next pulse, at the same time collecting nanoparticles, and preventing repeated interaction of ablation products with the laser beam.

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