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OPTICS AND SPECTROSCOPY

IONIZATION PROCESSES IN A CARBON AEROSOL UPON EXPOSURE TO LONG LASER PULSES. II

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An experimental setup and a technique for measuring the temperature and concentration of carbon and barium oxide particles in an ionized dispersed medium are described. It is shown that in a wide-aperture Nd *laser beam at a wavelength of 1.06 μm with a pulse duration of ~1 ms, the ionization threshold of a carbon dispersed medium is lower than the ionization threshold of a focused beam. The ionization threshold is 5−10 times higher for the medium with barium oxide particles (with a low ionization potential) than for the medium with carbon particles for both focused and wide-aperture beams. It has been experimentally confirmed that exothermic reactions prevail over other processes during ionization of the examined media.*

Keywords: laser radiation, optical breakdown, plasma, pulse, ionization, intensity, carbon, chemical reactions.

In part I of this work [1], mechanisms of interaction of low-energy long duration laser pulses with carbon particles dispersed in air were considered, and the basic equations describing the processes of evaporation, thermal emission, and exothermic reactions proceeding at temperatures above 1000°C were presented. The estimations showed that atoms and molecules with high ionization potential φ_ε (for example, $\varphi_\varepsilon = 11$ eV for carbon) contribute to a decrease in the temperature and electron concentration in the formed plasma, whereas atoms and molecules with low ionization potential (for example, $\varphi_\varepsilon = 5$ eV for aluminum oxide and $\varphi_\varepsilon \approx 5$ eV for barium oxide), on the contrary, increase the gas temperature and the electron concentration. The temperature changes can reach several hundred and even thousand degrees, and the electron concentration can change by an order of magnitude and even more.

Part II of this work is aimed at experimental verification of the theoretical estimates and results of calculations performed in part I of this work and is devoted to the prevailing contribution of the exothermic reactions to the ionization level in the laser beam channel with dispersed carbon and barium oxide particles having sizes of 10−100 μm and concentrations of 10^4–10^6 cm^{-3} at a wavelength of 1.06 μm, radiation intensity of ~10^5−10^6 W/cm^2, and pulse duration of ~1 ms.

Write the energy balance equation describing the process of laser radiation interaction with dispersed media in the form:

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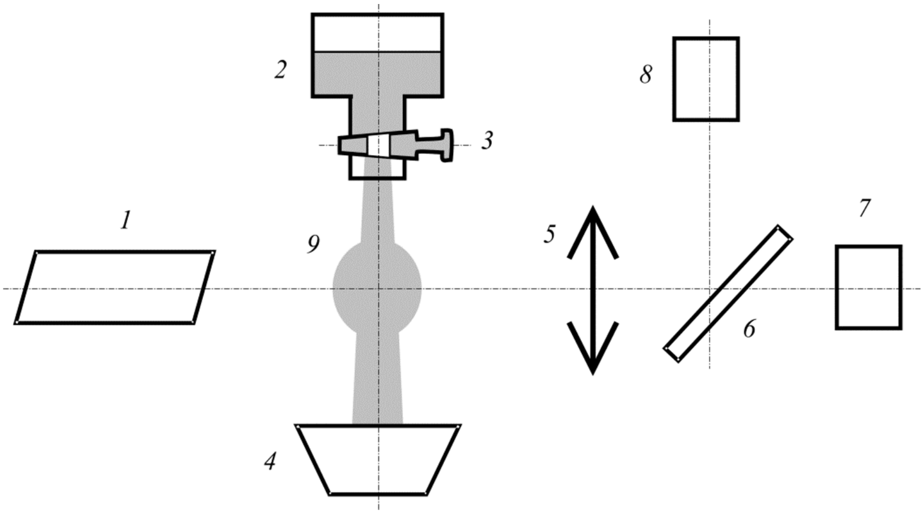

Fig. 1. Experimental scheme comprising GOS-1001 Nd laser *1*, container *2* with dispersed particles, tap *3*, cell *4*, focusing lens *5*, optical beam splitter *6*, MLE-200 energy meter *7*, MS-200 spectrograph *8*, and ionization zone *9*.

$$P\frac{d\upsilon_{\text{p.f}}}{dt} + E_{\text{em}} + E_{\text{therm}} + E_{\text{endoth}} = \sigma_{\text{a}} I + E_{\text{heat}} + E_{\text{exoth}}\,, \qquad (1)$$

where $\upsilon_{\text{p.f}}$ is the plasma formation volume; P is the pressure; E_{em}, E_{therm}, and E_{endoth} are energies lost by the plasma formation on thermal emission, thermal conductivity, and chemical reactions proceeding with energy absorption, $\sigma_{\text{a}}I$ is the total energy absorbed by the plasma formation; and E_{heat} and E_{exoth} are the energies liberated during particle heating and exothermic reactions. Let us consider the case

$$E_{\text{exoth}} > \sigma_{\text{a}} I, \qquad (2)$$

that is, when the exothermic reactions prevail, and the situation is possible in which the plasma formation energy exceeds that of the laser pulse spent for initiation of the chemical reactions.

To verify the statement on the prevailing role of the exothermic reactions in the dispersed carbon medium upon exposure to long laser pulses, we performed some experiments. Figure 1 shows the block diagram of the experimental setup. Radiation of pulsed laser source *1* with a wavelength of 1.06 μm, beam diameter of 45 mm, pulse duration of ≈1 ms, and energy of 700–1200 J passed through ionization zone *9* and then through lens *5* and beam-splitting plate *6* and was incident on MLE-200 laser energy meter *7* and MS-200 spectrograph *8*. To weaken the incident radiation energy, the attenuator operating in the range of 0.4–2.5 μm and having the extinction coefficients up to 10^3 was used together with the MS-200 spectrograph. Dispersed particles from tank *2* fall down in the vertical direction and entered into receiving cell *4*. In the zone of laser radiation interaction with particles, a plasma formation arose with sizes exceeding the laser beam diameter. The laser pulse energy was controlled with MLE-200 laser pulse energy meter *7*.

The gas temperature in the plasma was determined based on relative intensities of the first N_2^+ $(B_2\Sigma_u + {}^{-}X_2\Sigma_g^+)$ negative molecular nitrogen system and the second N_2 $(C_3\Pi_u^+ - B_3\Pi_g^+)$ positive system. The particle concentration was determined by the mass loss of particles arriving from container *2* to the laser beam interaction zone with the dispersed medium. If M_{p} is the total mass of particles precipitated from container *2*, $\tilde{m}_{\text{p}}$ is the average mass of one particle, and $\tilde{\nu}_{\text{p}}$ is the average volume of particles in the flow, then the average particle concentration is determined as

$$\tilde{N}_{\mathrm{p}} = \frac{M_{\mathrm{p}}}{\tilde{m}_{\mathrm{p}}\tilde{\nu}_{\mathrm{p}}}. \tag{3}$$

Considering that

$$\tilde{m}_{\mathrm{p}} = \frac{4}{3}\pi\,\tilde{R}_{\mathrm{p}}^{3}\,\rho_{\mathrm{p}} \text{ and } \tilde{\upsilon}_{\mathrm{p}} = \tilde{\upsilon}_{\mathrm{fl}}\tilde{s}_{\mathrm{fl}}t\,, \tag{4}$$

we obtain

$$\tilde{N}_{\mathrm{p}} = \frac{3M_{\mathrm{p}}}{4\pi\,R_{\mathrm{p}}^{3}\rho_{\mathrm{p}}\tilde{\upsilon}_{\mathrm{fl}}\tilde{s}_{\mathrm{fl}}t}, \tag{5}$$

where $\tilde{R}_{\mathrm{p}}$ is the average particle radius, ρ_{p} is the particle density, $\tilde{s}_{\mathrm{fl}}$ is the average cross section of the particle flow. Since $\tilde{s}_{\mathrm{fl}} = \pi\tilde{R}_{\mathrm{fl}}^{2}$, where $\tilde{R}_{\mathrm{fl}}$ is the average radius of the particle flow, then

$$\tilde{N}_{\mathrm{p}} = \frac{3M_{\mathrm{p}}}{4\pi^{2}\,R_{\mathrm{p}}^{3}\rho_{\mathrm{p}}\tilde{\upsilon}_{\mathrm{fl}}R_{\mathrm{fl}}^{2}t}. \tag{6}$$

For $M_{\mathrm{p}} = 200$ g, $\tilde{R}_{\mathrm{p}} = 50$ μm, $\rho_{\mathrm{p}} = 2.2$ g/cm^3, $\tilde{\upsilon}_{\mathrm{fl}} = 4$ m/s, $\tilde{R}_{\mathrm{fl}} = 1.78$ cm, and $t = 4$ s, we obtain $\tilde{N}_{\mathrm{p}} = 6.8\cdot10^{5}$ cm^{-3}, which is within the limits of the measured particle concentration.

Figure 2 shows the characteristic plasma formation generated by carbon particles with sizes of 10–100 μm and concentration of ≈10^5 cm^{-3} upon exposure to the wide-aperture radiation beam with energy of ≈1000 J. The plasma formation has a diameter of ≈70 mm and luminescence time of ≈1–2 s. The measurement of the plasma formation energy $E_{\mathrm{p.f}}$ showed that it was 3–5 times higher that the laser pulse energy. This suggests that in the course of radiation interaction with the medium, an additional energy is liberated due to the initiation of the chain of exothermic reactions proceeding with participation of ions, excited atoms, and carbon, oxygen, and nitrogen molecules.

Experiments were performed for the following conditions: particle diameter $\varnothing_{\mathrm{a}} = 10$–$100$ μm, concentration $N_{\mathrm{p}} = 10^4$–10^6 cm^{-3}, and laser pulse energy in the range of 700–1200 J. From the data obtained it follows that during laser radiation interaction with the carbon particles, the intensive ionization started at laser pulse energies of 1100–1200 J and carbon particle concentration $N_{\mathrm{p}} = 10^6$ cm^{-3}. In this case, the average laser radiation intensity was 69−75 kW/cm^2, which is much less than the intensity of initiation of plasma formations reported in works [3, 4]. This result is characteristic for wide-aperture beams when plasma formations can merge in a large volume and hence, a variety of exothermic reactions is excited that contribute to an increase in the gas temperature.

The experiments performed with the focused laser beam in the zone of carbon dispersed particles for radiation intensities of 1–1.2 MW/cm^2 showed that the average luminescence time of the plasma formation decreased to 0.1 s, and the temperature decreased to 3500–4000 °C. Similar results were also obtained for the BaO particles with the ionization potential (5 eV) lower than that of carbon. In this case, the plasma formation temperature also did not exceed 4000 °C [5, 6]. The processes of exothermic interaction of barium oxide were much less pronounced than of carbon. Figure 3 shows the curves illustrating changes in the average gas temperature of the plasma formation depending on the laser pulse energy for the indicated concentrations of carbon particles.

The basic chemical reactions with participation of BaO are:

$$\mathrm{BaO} + \mathrm{H_2O} \rightarrow \mathrm{Ba(OH)_2} + Q, \tag{a1}$$

$$\mathrm{BaO} + \mathrm{CO_2} \rightarrow \mathrm{BaCO_3}, \tag{a2}$$

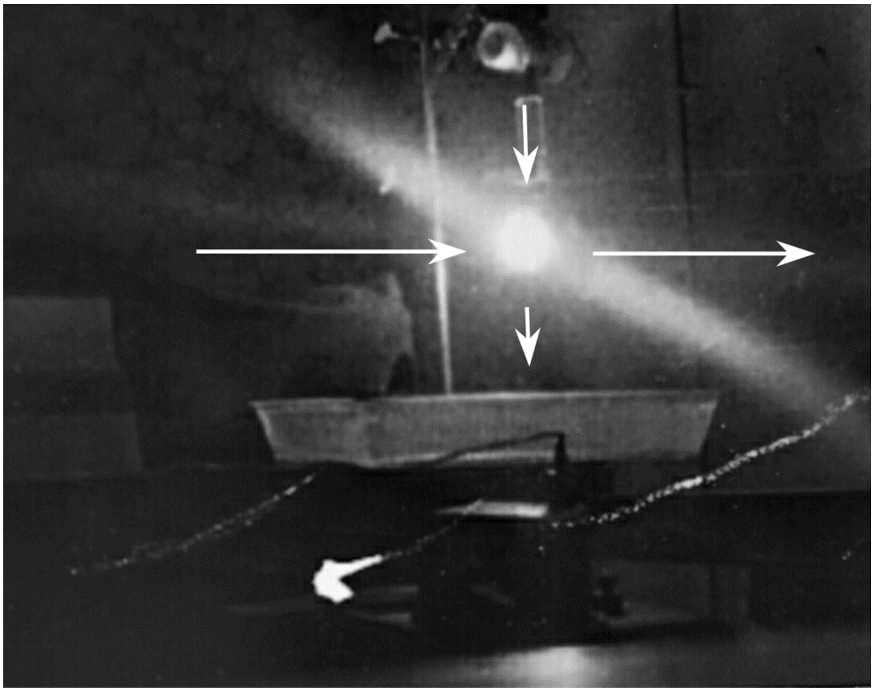

Fig. 2. General view of the plasma formation in dispersed carbon.

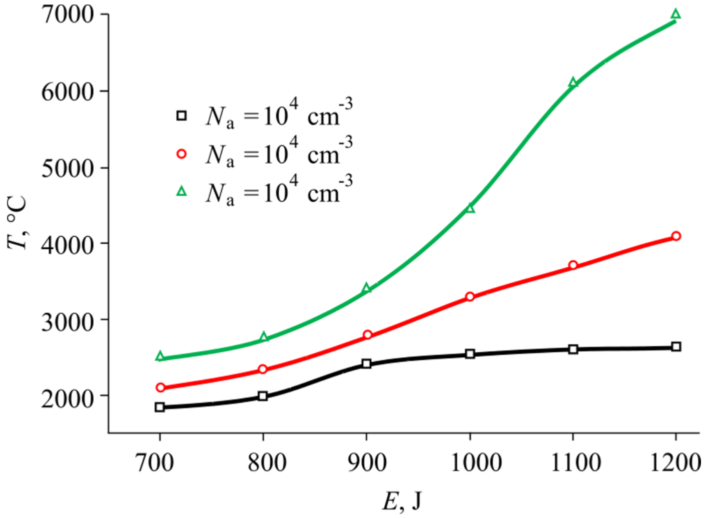

Fig. 3. Dependence of the plasma formation temperature on the laser pulse energy.

$$BaO + SO_3 \rightarrow BaSO_4, \tag{a3}$$

$$BaO + 2HCl \rightarrow BaCl_2 + H_2O, \tag{a4}$$

$$BaO + H_2SO_4 \rightarrow BaSO_4 + H_2O, \tag{a5}$$

$$2BaO + O_3 \rightarrow BaO_2, \tag{a6}$$

$$BaO + Mg \rightarrow Ba + MgO, \tag{a7}$$

$$BaO + H_3 \rightarrow BaH_2 + H_2O, \tag{a8}$$

$$BaO + NH_3 \rightarrow Ba + N_2 + H_2O, \tag{a9}$$

$$2BaO + CO \rightarrow Ba_2CO_3. \tag{a10}$$

Only reaction (a1) from all above-indicated reactions proceeds with considerable energy liberation [5]. Therefore, the processes of evaporated substance vapor ionization prevail in BaO upon exposure to laser radiation [1]. And exothermic reaction (a1) contributes to an increase in the ionization level.

The least breakdown threshold q of dispersed carbon was observed at $\varnothing_a = 80$ μm and particle concentration $N_p = 1.6 \cdot 10^3$ cm^{-3}; it was ≈1.9 MW/cm^2. For Al_2O_3, for example, at $\varnothing_a = 42$ μm and particle concentration $N_p = 2.1 \cdot 10^3$ cm^{-3}, $q = 0.37$ MW/cm^2 [3]. In work [7] it was established that the laser radiation intensity required to initiate a breakdown on the surface of a solid was $q \sim 10$ MW/cm^2. A considerable decrease in the dispersed carbon breakdown threshold with high particle concentration ($N_p = 10^6$ cm^{-3}) in the experiment with wide-aperture laser radiation beam indicates a wide variety of exothermic reactions in the collective breakdown mode of type a_1–a_{15} [1]. As a result, the temperature of the medium increased together with the free electron concentration.

CONCLUSIONS

1. During the chemical reactions proceeding in the high-power laser radiation channel, a significant increase in the temperature and electron concentration in the gas can be reached only when the chain of exothermic reactions prevails in the wide-aperture beams at particle concentrations of 10^5–10^6 cm^{-3}.

2. Upon exposure to the wide-aperture long duration beam of the Nd laser, the plasma formation in the carbon dispersed medium with particle concentration of 10^6 cm^{-3} and sizes of 10–100 μm, the plasma formation reached lifetimes of 1–2 s and temperatures of 6000–7000 K, and its liberated energy is 3–5 times higher than the laser pulse energy.

3. In the process of the focused laser beam interaction with the medium consisting of BaO particles with ionization potential less than that of carbon, the lifetime of the plasma formation does not exceed 0.1 s, and its average temperature is 4000 K, which suggests a lower chemical activity of this medium in comparison with carbon one.

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