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Spectral Characteristics of the Glow of Coal Flames during the Exposure to Laser Pulses

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The results of measuring the characteristics of the glow of coal flames at the initial moments of ignition under the action of laser pulses of a neodymium laser (1064 nm, 120 μs) are presented. In microparticles of coal grades DG, G, Zh, and K, when the corresponding critical energy densities are exceeded during a laser pulse, the surface ignites and flame propagates at a speed of $V \approx 50$ m/s. The emission spectra of flames are contributed by the emission of emitted hot coal particles, excited H_2^* and H_2O^* molecules, as well as the flame emission arising from the oxidation of carbon (CO^*) and carbon monoxide (CO_2^*) by atmospheric oxygen.

Keywords: coal, laser, ignition, flame, volatile substances, degree of coalification, coke residue.

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Introduction

At present, the main direction of the coal use is energy. Increasing environmental problems require a transition to new coal combustion technologies with reduced emissions of pollutants into the environment.

The development of new technologies for ignition and combustion intensification of coal is impossible without a detailed study of the initial mechanisms of its ignition. When studying the mechanisms of coals ignition the laser radiation [1–5] can be used as an ignition source, and to register fast processes it is possible to use electron-optical methods [1–3,5] and the method of spectroscopy with high time resolution [6–8]. We present some results of such studies.

In [2] the kinetic characteristics of the process of laser ignition of coal particles and carbon material by neodymium laser pulses (1064 nm, 120 μs) are obtained. For coal with a high content of volatile substances, the duration of the luminescence recorded by a photomultiplier reached 40−70 ms, for anthracite — 15−30 ms, for carbon material — 50−80 ms. For all samples the first short peak was observed on the kinetic dependence of the luminescence, the duration of which for coal with high content of volatile substances and carbon material reached ~ 1 ms, for anthracite — 3 ms. For anthracite and carbon material a second luminescence component was observed, which significantly exceeded the duration of the first peak. For coal with a high content of volatile substances, in addition to the first and second components a third, even longer luminescence component was registered.

The authors of paper [9] proposed a mechanism for flame propagation in pulverized coal aerosol under the action of laser radiation, which was as follows. Initially, when under the action of laser radiation a small number of coal particles are ignited, from them volatile substances are released during thermal decomposition. As the combustion front spreads, the nearby coal particles heat up, undergo thermal decomposition, volatile substances are released and then ignited.

In [10] a model of coal ignition by a laser pulse was developed. The main provisions of the model are as follows. As a result of the laser pulse action the surface of the coal particle is heated and undergoes thermal decomposition, volatile substances are released, which, mixing with an oxidizing agent, absorb part of the laser radiation, which leads to the gas phase heating and the chemical reactions development in it. The coal particles can be ignited by two mechanisms: 1 — the gas phase is initially ignited, 2 — the solid surface of the coal particles is initially ignited, then the gas phase is ignited.

In [1] the presence of two mechanisms of laser ignition of coal samples was experimentally confirmed, as was assumed in [10]. In paper [5] it is noted that the coal ignition mechanism is determined by its position in the genetic series.

In our previous papers studies were carried out on the laser ignition of brown and a number of hard coals from the Kuznetsk coal basin under the action of neodymium laser radiation with a pulse width of 120 μs [11–14], and the spectral-kinetic regularities of ignition of brown coal were studied [15].

In [11–15] three stages of laser ignition of coals are distinguished with corresponding thresholds, which have characteristic burning times at each stage. It is concluded that at the first stage, when the threshold energy density $H_{cr}^{(1)}$ is reached, thermal luminescence is observed on the surface of the coal sample heated to a temperature of

$T = 3,100$ K. At the second stage, when the threshold energy density $H_{cr}^{(2)}$ is reached in a millisecond time interval, the coals are ignited, and the flame luminescence is observed, attributed in [15] to the radiation of volatile substances and thermal luminescence at $T \sim 2,300$ K. At the third stage of ignition, when the threshold energy density $H_{cr}^{(3)}$ is reached in the time interval ~ 100 ms, thermal luminescence of the flame with a temperature of $T \sim 1,800$ K is observed, referred in [15] to the combustion of the non-volatile residue of coal particles.

This paper is a continuation of previous studies and develops understanding of the processes occurring at the initial stages of ignition of coal particles.

Samples and experimental procedure

We used coals from the Kuznetsk coal basin (RF) of various stages of metamorphism: long-flame gas (LFG), gas (G), fatty (F) and coke (C), taken from the same batch of samples used in paper [13]. After grinding in a ball mill and screening through sieves the coal fractions with a wide distribution by sizes $d \leq 63\,\mu$m were selected. The results of the coals technical analysis are given in [13]. We used samples with bulk density $\rho = 0.5$ g/cm^3, weight 10 mg, which were placed in a copper cartridge 5 mm in diameter and 2 mm deep. The functional diagram of the experimental setup and the description of the measuring procedure for the emission spectra of solids and flames are given in paper [15].

To ignite the coals a pulsed YAG:Nd^{3+} laser was used, operating in the free-running mode at the main frequency ($\lambda = 1,064$ nm) with a pulse with of $\tau_i = 120\,\mu$s and pulse energy $E \leq 1.5$ J. To register the luminescence of the flames a polychromator and a photochronograph based on an electronic image converter (EIC) operating in the linear sweep mode (streak camera) were used. The area of the flame luminescence, limited by the size of the space-time slot 0.1×0.2 mm, was projected onto the input of the polychromator using a lens system and converted into a spectrum in the wavelength range $\Delta\lambda = 350-750$ nm. The image of the spectrum was projected onto the photocathode of EIC, which scanned the spectrum in time. The light matrix on the output screen of EIC was read by CCD matrix and transferred to a computer for further processing. According to the vertical rows of the light matrix, it is possible to build a luminescence spectrum at a specific time moment. According to the horizontal lines — the luminescence intensity versus time at a fixed wavelength. The spectral and temporal resolutions were determined by the size of the spectral-time gap and amounted to 10 nm and 2 ns, respectively, at the shortest sweep of the photochronograph. On the sweep used in this paper the time resolution was 2 μs, which is much less than the laser pulse duration.

The reference lamp method [16] was used to correct the emission spectra for the spectral sensitivity of the measuring path.

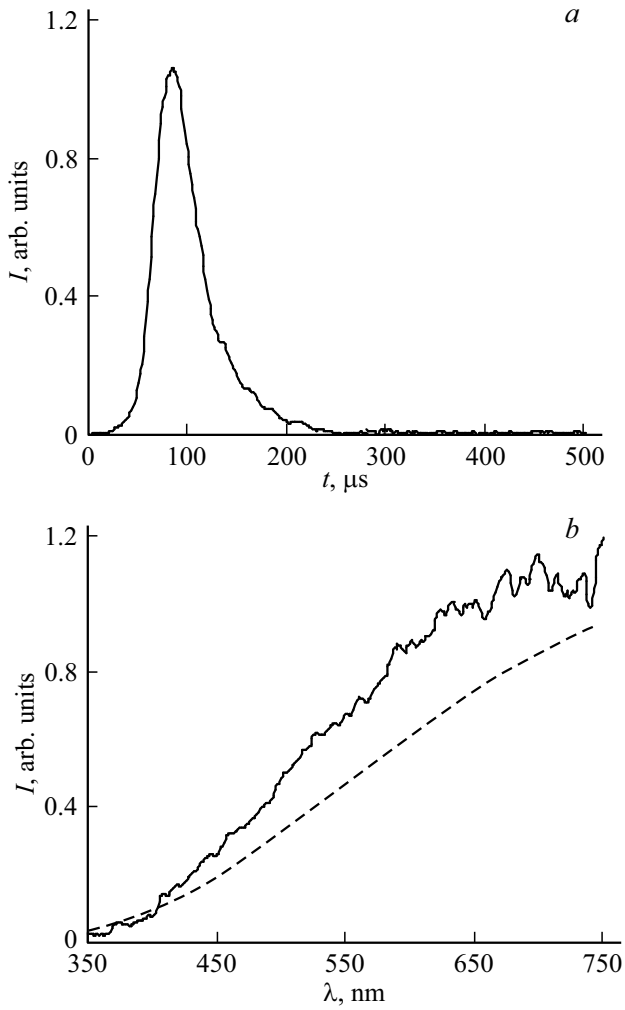

Figure 1. Kinetic dependence of the luminescence intensity and the spectrum of the flame luminescence above the sample at a distance of 1 mm from the surface of the coal sample at $H_{cr}^{(2)} = 1.6$ J/cm^2. (*a*) luminescence intensity vs. time; (*b*) spectrum at the maximum of the kinetic dependence of the luminescence, dashed line — approximation of the luminescence of carbon particles by the Planck formula with a temperature of 3,100 K.

The sample surface was located perpendicular to the laser radiation and parallel to the optical axis of the measuring path. The sample could move vertically so that different sections of the flame could fall into the space-time slot.

Experimental results

Flame spectra were studied using threshold radiation energy densities at which flame appears associated with volatile substances ($H_{cr}^{(2)}$) and ignition of the non-volatile residue of coal particles ($H_{cr}^{(3)}$), measured for the used coals in paper [13] and given in Table 1.

As the experiment showed, the spectral and kinetic patterns of luminescence for the studied coals have the same

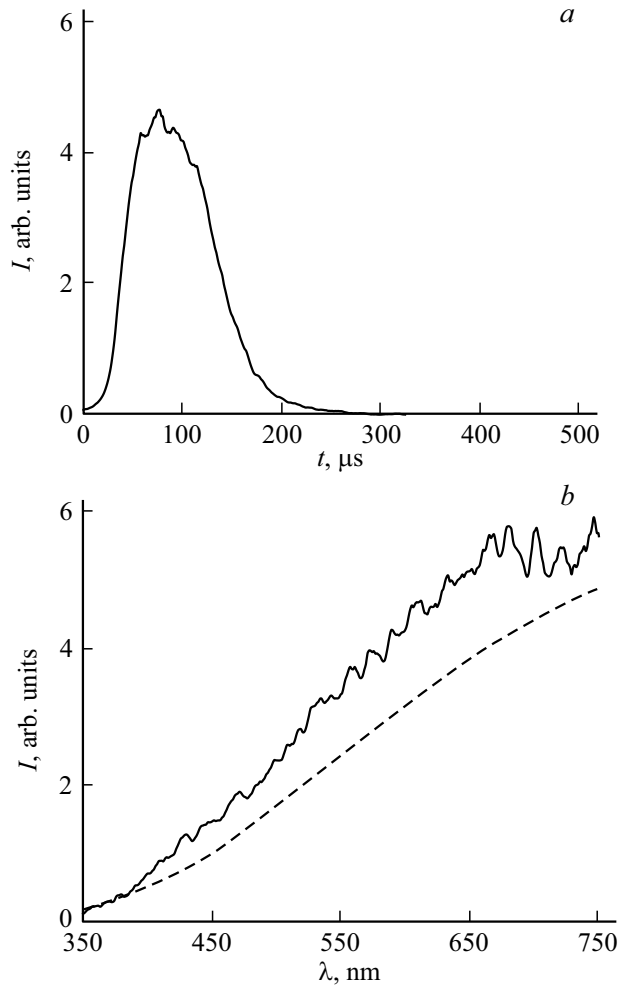

Figure 2. Kinetic dependence of the luminescence intensity and the spectrum of the flame luminescence above the sample at a distance of 1 mm from the surface of the coal sample at $H_{cr}^{(3)} = 2.4\,\text{J/cm}^2$. ($a$) luminescence intensity vs. time; (b) spectrum at the maximum of the kinetic dependence of the luminescence. Dashed line — approximation of the luminescence of carbon particles by the Planck formula with a temperature of 3,100 K.

Table 1. Ignition thresholds for coals at various stages of ignition [13]

Coal grade	$H_{cr}^{(2)}$, J/cm^2	$H_{cr}^{(3)}$, J/cm^2
LFG	1.6	2.4
G	1.85	3.3
F	1.1	5.5
C	0.9	6.5

qualitative nature when exposed to the corresponding H_{cr} indicated in Table. Due to the limited scope of the paper, below we present illustrative material based on the LFG grade coal.

Fig. 1 shows the kinetic dependence of the luminescence at a wavelength of $\lambda = 560$ nm (Fig. 1, a) and the spectrum of the luminescence at the intensity maximum of the kinetic dependence of the luminescence at a distance of 1 mm from the sample (Fig. 1, b) under exposure with energy density $H_{cr}^{(2)}$. The luminescence front corresponds to the duration of the laser pulse, the beginning of the luminescence is delayed by $\sim 20\,\mu$s relative to the laser pulse. luminescence decay ends in $\sim 200\,\mu$s.

Fig. 2 shows the same dependences under the exposure of the laser pulse with an energy density of $H_{cr}^{(3)}$. The kinetics and the luminescence spectrum have a similar character with the luminescence regularities obtained under the effect with an energy density of $H_{cr}^{(2)}$, but the luminescence intensity increases by ~ 5 times.

Fig. 3 shows the kinetic dependence of the flame luminescence at a wavelength of $\lambda = 560$ nm and the luminescence spectra at the intensity maxima of the kinetic dependence of the luminescence at a distance of 2 mm from the sample surface under exposure with an energy density $H_{textcr}^{(2)}$. The beginning of the luminescence is delayed by $\sim 40\,\mu$s relative to the beginning of the laser pulse. The kinetic dependence of the luminescence has a non-elementary nature, the duration of the luminescence is ~ 2 ms (Fig. 3, a). At first thought, the difference between the luminescence durations in Figs. 1, a and 3, a is puzzling, but the presentation of these figures in the same scale allows us to conclude that the long components of the luminescence with maxima *2* and *3* in Fig. 3, a are not visible in Fig. 1, a due to the limited dynamic range of brightness gradations of the EIC. At a distance of 1 mm the luminescence intensity at the maximum in Fig. 1, a exceeds the luminescence intensity at the maxima in Fig. 2, a by 50–100 times, so the luminescence intensity shown in Fig. 3, a has a noise level in Fig. 1, a.

Fig. 4 shows the kinetic dependence of the luminescence and the luminescence spectra at a distance of 2 mm from the surface of the samples at the radiation energy density of $H_{cr}^{(3)}$. The front of the flame luminescence corresponds to the duration of the radiation pulse. The luminescence ends in $\sim 400\,\mu$s. In some cases in the time interval ≥ 1 ms the luminescence of low intensity is registered (Fig. 4, a).

When samples are exposed to radiation with an energy density greater than $H_{cr}^{(2)}$, but less than $H_{cr}^{(2)}$, on the solid spectra three emission lines ($\lambda = 555, 590, 625$ nm) in the time interval of $300-600\,\mu$s are superimposed after exposure to laser pulse. This result is shown in Fig. 5.

The luminescence spectra are interpreted in the next Section.

Discussion of results

The beginning of the flames luminescence in Figs. 1, a and 2, a is synchronized with the beginning of the laser pulse action on the samples. The delay in the beginning of the luminescence in Fig. 1, a and 2, a corresponds to the propagation time of the luminous products at a distance

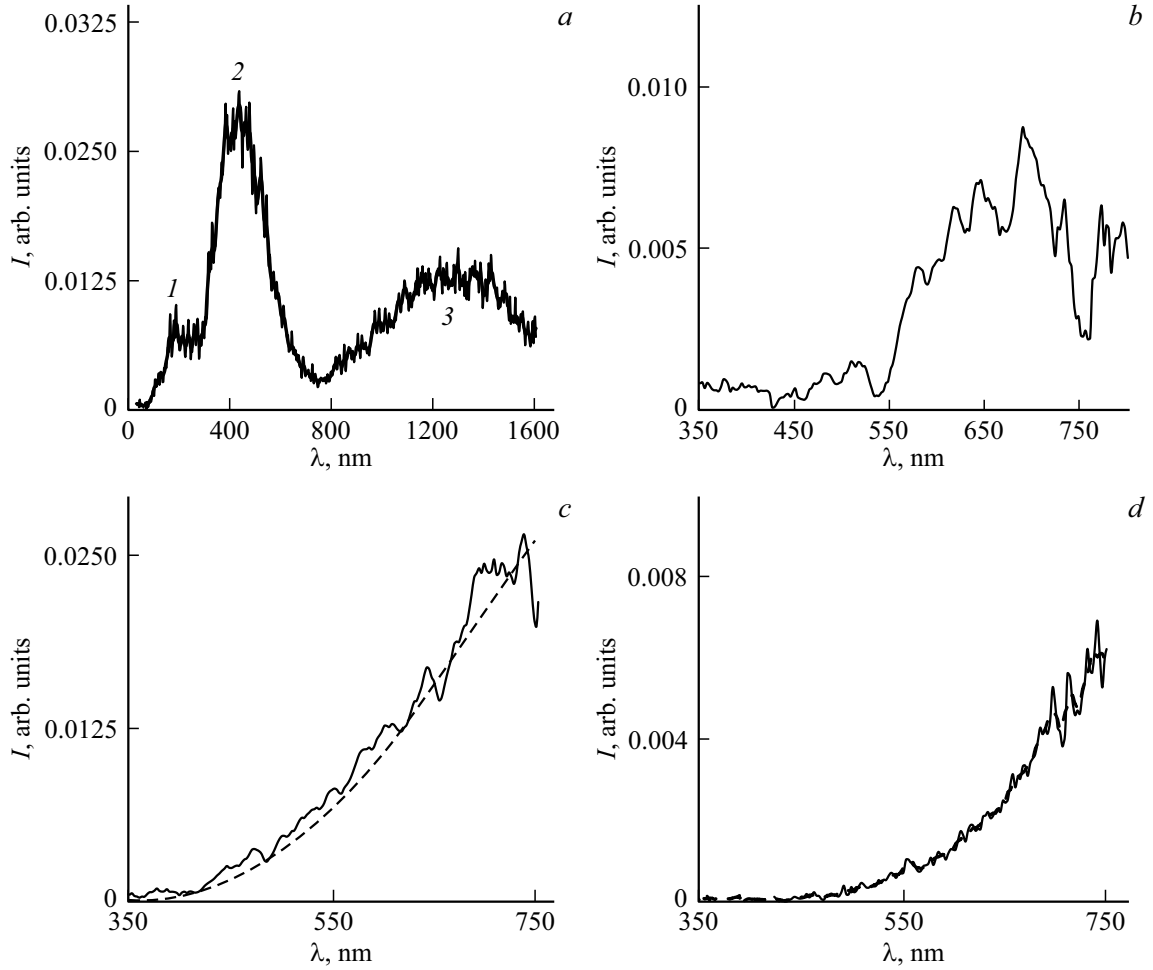

Figure 3. Kinetic dependence of the luminescence intensity and the spectra of the flame luminescence above the sample at a distance of 2 mm from the surface of the coal sample at $H_{cr}^{(2)} = 1.6\,\mathrm{J/cm^2}$ at different times. (*a*) Luminescence intensity vs. time; (*b*) emission spectrum at time *1* in Fig. 3, *a*; (*c*) emission spectrum at time *2* in Fig. 3, *a*; (*d*) emission spectrum at time *3* in Fig. 3, *a*. Dashed line — approximation of the luminescence of carbon particles by the Planck formula with a temperature of 3,100 K.

of 1 and 2 mm from the surface of the coal particles and is, respectively, ~ 20 and $40\,\mu$s. From here we can estimate the flame propagation speed at this stage: $V \approx 50$ m/s. An important conclusion also follows from these results, that the luminescence shown in Figs. 1, *b* and 2, *b* is associated with flame occurred on the surface of coal particles directly during the laser pulse action, followed by propagation. Thus, at energy densities $H > H_{cr}^{(2)}$ the particle surface ignites directly during the laser action.

At a distance of 1 mm from the sample the luminescence spectra have a non-elementary nature (Fig. 1, *b* and 2, *b*), in particular, they are not fully thermal. The shape of the spectra suggests that excited molecules of volatile substances and escaping hot coal particles, the luminescence of which has thermal nature, contribute to the luminescence. The presence of the latter can be detected by a black coating on a transparent glass plate, if it is placed at a distance of 1–2 mm from the sample. If we assume that the temperature of the emitted coal particles is $T = 3,100$ K (the dashed line in Fig. 1, *b* and 2, *b*), then the difference spectra *1* and *2* in Fig. 1, *b* and 2, *b* gives the spectrum of volatile substances, which is shown in Fig. 6. The assumption for the temperature of the coal particles $T = 3,100$ K is somewhat arbitrary. However, it relies on the measured surface temperature at $H \leq H_{cr}$ for brown coal [15]. Note that such a representation is largely of a qualitative nature, since the scale factors for the luminescence of coal particles in Figs 1, *b* and 2, *b* are taken arbitrarily. Nevertheless, from our point of view, from the difference spectra in Fig. 6, we can make the conclusion about the qualitative composition of the flammable volatile components. The shape of the difference spectra (Fig. 6) suggests that, as in the spectra of brown coal flame [15], the flame of CO (300–550 nm) [17,18] and of the excited molecules H_2^* and H_2O^* (550–750 nm) contribute to luminescence [17,18].

At a distance of 2 mm the first maximum in the kinetic dependence of the luminescence (Fig. 3, *a*) is associated with the products emitted from the surface at the moment of exposure to the laser pulse. In this case, the luminescence is observed in the spectral range 550–750 nm (Fig. 3, *b*) only.

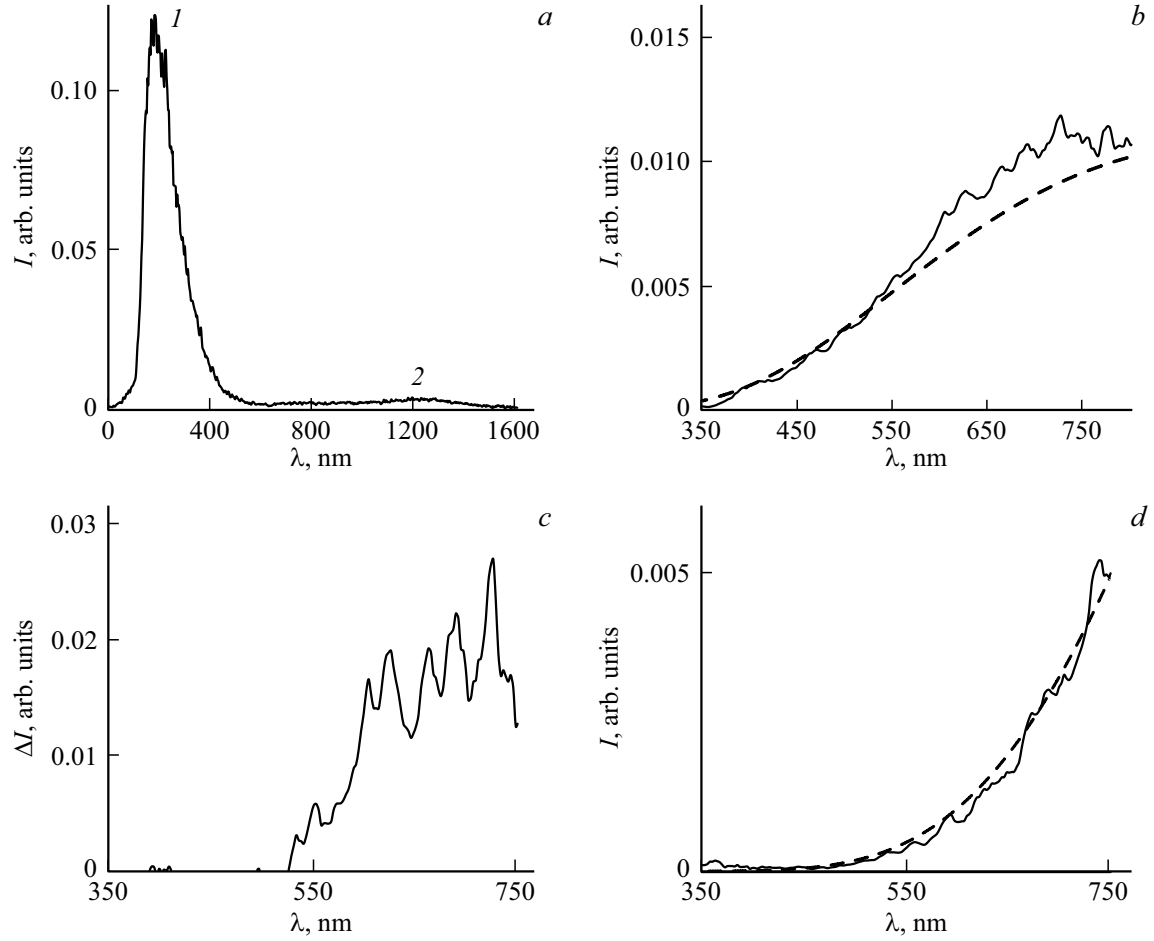

Figure 4. Kinetic dependence of the luminescence intensity and the spectra of the flame luminescence above the sample at a distance of 2 mm from the surface of the coal sample at $H_{cr}^{(3)} = 2.4\,\mathrm{J/cm^2}$ at different times. (a) Luminescence intensity vs. time; (b) emission spectrum at time 1in Fig. 4, a; (c) difference spectrum of solid an dashed curves in Fig. 4, b; (d) emission spectrum at time 1.2 ms. Dashed line — approximation of the luminescence of carbon particles by the Planck formula with a temperature of 3,100 K.

We associated this luminescence with the luminescence of excited molecules H_2O^* and H_2^*, as in [15]. The luminescence of the CO flame in the short-wavelength part of the spectrum is absent, in contrast to the luminescence in Fig. 1, b, 2, b, 6. The CO flame is associated with the reaction progress

$$2CO + O_2 \rightarrow 2CO_2^*. \qquad (1)$$

Apparently, immediately after the CO molecules formation they are oxidized on the surface of coal particles by air oxygen according to reaction (1) already at the first millimeter from the surface of coal particles. Therefore, the luminescence of the CO flame at a distance of 2 mm from the surface of the coal particles is no longer observed. Spectra at subsequent times at the maxima of the kinetic dependences *2* and *3* Fig. 3 (Fig. 3, c, 3, d) are described by the Planck formula at temperatures $T = 2,400\,\mathrm{K}$ and $T = 1,800\,\mathrm{K}$, respectively, i.e., are thermal.

It follows that as a result of exothermic reactions in coal particles, combustion is initiated with complex kinetics in the millisecond time interval, leading to the escape of hot coal particles.

The emission spectra of the flame at a distance of 2 mm when exposed to the critical energy density $H_{cr}^{(3)}$ (Fig. 4) during exposure to laser radiation differ from the emission spectra at a distance of 1 mm at $H_{cr}^{(3)}$ (Fig. 2) by the absence of CO flame luminescence in the short-wavelength part of the spectrum (Fig. 4, b and 4, c). The contribution to the luminescence spectrum comes from incandescent soot particles at $T = 3,100\,\mathrm{K}$ and the luminescence of excited molecules H_2^* and H_2O^* (Fig. 4, b, 4, c). According to the time characteristics (Fig. 4, a) this flame appears during the laser pulse action, i.e. as a result of surface ignition.

In a millisecond time interval (Fig. 4, d) the escape of incandescent coal particles is observed at $T = 1,800\,\mathrm{K}$.

When the radiation energy density is greater than $H_{cr}^{(2)}$, but less than $H_{cr}^{(3)}$, lines 555, 590, 625 nm are superimposed on the continuous spectrum. According to reference data [17,18], the line 625 nm can be associated with the luminescence of excited CO^* molecules. It can be assumed

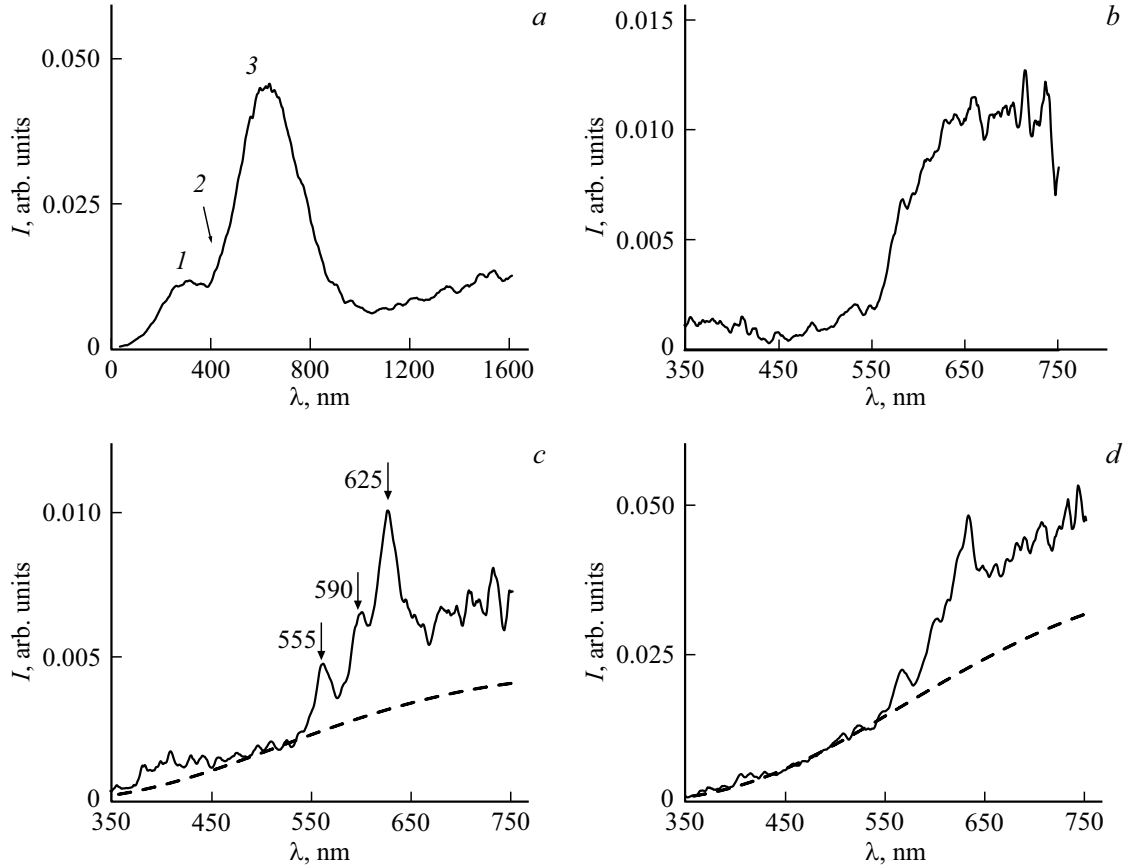

Figure 5. Spectral and kinetic characteristics of the luminescence of a LFG grade coal flame at a distance of 2 mm from the surface when the sample is exposed to radiation with energy density of $H_{cr}^2 = 2\,\mathrm{J/cm^2}$. (*a*) Luminescence kinetics, (*b*) luminescence spectrum at time *1*, (*c*) luminescence spectrum at time *2* , (*d*) emission spectrum at time *3*. Dashed line — approximation of the luminescence of carbon particles by the Planck formula with a temperature of 3,100 K.

that carbon particles are oxidized by air oxygen according to the reaction

$$2C + O_2 \rightarrow 2CO^*. \qquad (2)$$

Luminescence lines with maxima 555 and 590 nm, according to reference data [17,18], can be associated with the luminescence of excited molecules C_2^*.

Upon the radiation energy density increasing (Fig. 4), the luminescence of CO^* and C_2^* is not observed. This may be due to the intensity increasing of the continuous spectrum luminescence. If there is no proportional increasing of the yield of CO and C_2 molecules, then their luminescence is difficult to distinguish against the background of the continuous spectrum.

Qualitatively similar results were obtained using coal grades G, F, C.

There is some difference in the luminescence temperatures at *2* and *3* maxima at a distance of 2 mm from the sample surface for coals of grades G and F, for coals of grade C at a distance of 2 mm from sample two luminescence maxima are observed, but not three, as for other grades of coal. Besides, line spectrum is not observed for coal grade C.

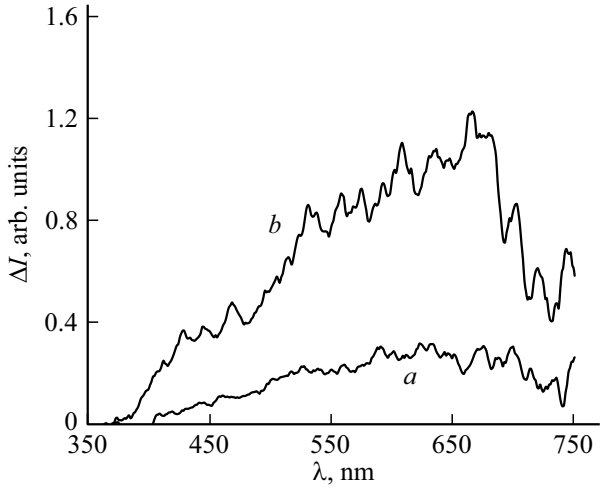

Figure 6. Difference spectra between curves *1* and *2*: *a* — in Fig. 1, *b*, *b* — in Fig. 2, *b*.

The main result is that when the critical energy density $H_{cr}^{(2)}$, characteristic for each grade of coal, is exceeded, the surface of the coal particles ignites already during the

Table 2. Intensity of the first peak of the flame luminescence of different grades of coals at the corresponding threshold energy densities at a distance of 1 mm from the surface of the coal sample ($\lambda = 560$ nm)

Coal grade	I, rel. units	
	$H_{cr}^{(2)}$	$H_{cr}^{(3)}$
LFG	1.0	4.2
G	2.4	7.5
F	2.5	8.0
C	4.0	9.0

laser pulse. The luminescence spectra of flame occurred on the surface of coal particles contribute to the luminescence of emitted coal particles and volatile substances, among which excited molecules H_2^* and H_2O^* can be identified, as well as carbon monoxide CO, which ignites by reacting with atmospheric oxygen to form CO_2^* [17].

A quantitative difference between the grades of coals is observed in the luminescence intensities immediately after exposure to the laser pulse, which is demonstrated by the measurement results in Table 2. With the degree of carbonification increasing, the increasing of the luminescence intensity is observed. Noteworthy is that $H_{cr}^{(2)}$ decreases in this series of coals (Table 1). At this stage, it can be assumed that when the corresponding ignition thresholds are reached, the rates of chemical reactions leading to the ignition of the surface of coal particles increase with the degree of carbonification increasing, which leads to the increased luminescence intensity of the flames. Also note that for a particular grade of coal, the luminescence intensity increases with the increasing of the radiation energy density H, as evidenced by the data in Fig. 1, 2 and Table 2. This indicates the increased chemical reactions rate, as a result of which the luminescence intensity increases with H increasing.

As a result of the energy absorption of laser pulses, the non-volatile residue is ignited when $H_{cr}^{(3)}$ is exceeded in the time interval 10−100 ms, as shown in paper [13]. However, the study of the mechanism of ignition of the non-volatile residue was beyond the scope of this paper.

Analyzing the results of spectral-kinetic measurements of the luminescence of flames, we can conclude that already during the laser pulse the processes of ignition of the surface of coal particles and gaseous pyrolysis products occurs in parallel.

Conclusions

1. Under the action of YAG:Nd^{3+}-laser pulses ($\tau = 120\,\mu$s) on coals of grades LFG, G, F, C when the corresponding critical ignition energy densities ($H_{cr}^{(2)}$) are exceeded during the laser pulse, the surface of the coal particles is ignited, and the flame propagates at a speed of $V \approx 50$ m/s.

2. The emission spectra are of non-elementary nature. The contribution to the luminescence is made by the incandescent coal particles present in the flame, volatile substances: excited molecules H_2^*, H_2O^*, as well as excited molecules CO^* and CO_2^*, which are formed during the carbon molecules oxidation by atmospheric oxygen.

3. The luminescence intensity of the flames increases with carbonification increasing at the corresponding ignition energy densities $H_{cr}^{(2)}$ and $H_{cr}^{(3)}$.

4. For a particular grade of coal the luminescence intensity increases with laser radiation energy density increasing.

5. Under the experimental conditions of the present paper, the processes of ignition of the coal particles surface and of gaseous pyrolysis products, that arise during the exposure to laser pulse, occur in parallel.

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Conflict of interest

The authors declare that they have no conflict of interest.

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