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The Ignition Energy Characteristics and Glow Kinetics of the Flames of Dispersed Coal Particles of Different Ranks under the Action of Laser Pulses

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Abstract

Laser ignition (1064 nm, 120 μs) of the dispersed particles of different coal marks was studied. For all coal marks, three consecutive ignition stages with characteristic threshold radiation energy densities (H_{cr}) for each stage were discovered. With an increase in the rank, the values of the first ignition limit $H_{cr}^{(1)}$ remain almost constant, the second ignition limit $H_{cr}^{(2)}$ decreases, while the third ignition limit $H_{cr}^{(3)}$ increases. Results of the investigation of kinetic dependencies of flame glow at different ignition stages arising under the action of laser pulses on coal particles are presented. The glow duration at the first ignition stage slightly exceeds the duration of the laser pulse and reaches 150 μs for all marks of coal. The glow duration at the second ignition stage at $H = H_{cr}^{(2)}$ is in the millisecond time interval. With an increase in the energy density of laser radiation, the glow duration at the second ignition stage decreases to the submillisecond range as a result of an increase in the rates of thermochemical reactions. At the third ignition stage at $H = H_{cr}^{(3)}$, the glow duration is 10–100 ms for different marks of coal. It was established that for the studied coals, the glow intensity increases from the moment of exposure to laser pulse. In the submillisecond range, a decrease in glow intensity was observed. The glow amplitude of coals increases linearly with an increase in the energy density of laser radiation.

Keywords: coal, laser ignition, combustion, glow kinetics, ignition thresholds, ignition stages

INTRODUCTION

The development of physical methods for the ignition of fuel composed of coal particles opens outlooks for mazut-free ignition of coal in industrial furnaces, which will provide substantial economic and ecological effects. In particular, developments based on plasmatrons are already available for these purposes [1–3]. The studies into laser ignition of dispersed coal particles are promising for the development of new systems for the ignition of coal-dust fuel. One of the possible schematics of a laser ignition system is as follows. At the stage of firelighting, the coal particle fuel is supplied to the lighting muffle burners in which a highly efficient solid-state laser is used as a firing device; the main burners of the furnace are switched off during this procedure. Laser ignition systems are supposed to possess a number of advantages in comparison with plasmatron-based developments: lower energy consumption, technical simplicity of the systems, the absence of consumables to be replaced frequently (such as nozzles and cathodes for a plasmatron). To make the systems of laser ignition of coal dust fuel in industrial furnaces, fundamental research of the processes taking part at the initial stages of coal ignition is necessary. These studies were launched in [4–9] and are continued in the present work. In addition, this direction is significant for the improvement of the technologies for intensification of the combustion of solid organic fuel, as well as for the development of methods for the prevention of coal dust explosions in mines. The use of pulsed laser radiation for initiation of thermochemical coal ignition processes is distinguished by the simplicity of energy input to coal particles and by the possibility to provide control over the processes at different stages of ignition with the help of the electron-optical method [4, 5] and optical spectroscopy with high temporal resolution [6].

Detailed investigation of threshold energy and spectral-kinetic characteristics of the glow of the flames of dispersed coal particles along the metamorphism sequence has not been carried out yet. Previously we determined the characteristics of laser ignition of coal of B, DG, G, Zh, and K marks, and measured the kinetic dependences of flame glow for the corresponding threshold energies [5, 6]. The present work is a continuation of those studies with the expansion of a set of coal samples and the generalization of the regularities discovered in the metamorphism sequence from brown coal to anthracite.

EXPERIMENTAL

Materials

The objects of the investigation were coal samples from the Kuznetsk coal basin; coals marks B (Kaychakskiy open-pit mine), D (Kamyshanskiy open-pit mine, Severo-Taldinskoye deposit, course 73), DG (V. D. Yalevskiy mine, course 52), G (Kirov mine, Polenovskiy course), Zh (Tikhov mine, course 23), K (LC Uchastok Kokosoviy, course II internal), OS (open-pit mine Tomusinskiy), SS (Bachatskiy open-pit mine), T (JSC Kuznetskinveststroy, course 19a), A (open-pit mine Bungurskiy). Coal samples were obtained from the coal bank of the Institute of Coal Chemistry and Chemical Materials Science of the Federal Research Centre of Coal and Coal Chemistry, SB RAS.

For the preparation of experimental samples, coarse coal cobs of each mark were ground with a Pulverisette 6 ball mill (Fritsch, Germany). After grinding, coal particles were sieved through a vibratory sieve with mesh size 63 μm.

The parameters of the technical analysis of coal were determined using standard procedures. Water content (W^a) in the analytical coal sample was calculated as the mass loss by coal portion during heating in a drying box at 378–383 K to the constant mass (GOST R 52917–2008); ash content (A^d) was calculated as the mass of the residue measured after burning the coal portion in the muffle furnace at a temperature of 1078–1100 K (GOST 11022–95), the resulting A^d values are given per the dry state of coal portion. The yield of volatiles (V^{daf}) was calculated as the mass loss by coal portion during heating without air access up to 1173 K for 7 min after subtraction of the mass loss due to the presence of water in the sample (GOST 6382–2001), the resulting values of V^{daf} are given per the dry ash-free state of coal portion.

Results of the technical analysis of analytical coal samples are presented in Table 1. The elemental composition of coal samples was determined with the help of automatic C, H, N, S, O

TABLE 1

Results of technical analysis of the analytical samples of coal

Sample No.	Sample No., coal mark, mining site	W^a, %	A^d, %	V^{daf}, %
1	27, B, Kaychakskiy open-pit mine	11.8	10.1	53.1
2	72, D, Kamyshanskiy open-pit mine, the Severo-Taldinskoye deposit, course 73	7.6	6.2	44.5
3	64, DG, V. D. Yalevskiy mine, course 52	5.7	4.7	42.6
4	40, G, Kirov mine, Polenovskiy course	1.2	3.3	40.4
5	15, Zh, Tikhov mine, course 23	0.8	7.8	33.3
6	10, K, LC Uchastok Koksoviy, course II internal	0.6	4.9	21.3
7	34, OS, Tomusinskiy open-pit mine	0.1	6.7	19.8
8	45, SS, Bachatskiy open-pit mine	1.3	4.7	19.0
9	81, T, JSC Kuznetskinveststroy, course 19a	0.5	6.2	14.4
10	33, A, Bungurskiy open-pit mine	0.4	3.6	7.7

Note. W^a – analytical humidity, A^d – ash content, V^{daf} – yield of volatile substances.

TABLE 2

Results of the elemental analysis of coal

Sample No.	Sample No., coal mark	Content, %				
		C	H	N	S	O (from difference)
1	27, B	61.4	5.1	1.0	0.5	31.9
2	72, D	74.4	5.3	2.3	0.5	17.5
3	64, DG	74.3	5.3	2.3	0.3	17.7
4	40, G	81.3	5.8	3.1	0.2	9.6
5	15, Zh	80.2	5.2	3.0	0.4	11.2
6	10, K	87.7	4.6	2.2	0.4	5.1
7	34, OS	84.8	4.2	2.0	0.3	8.7
8	45, SS	83.8	4.0	2.1	0.1	10.0
9	81, T	89.7	4.1	2.0	0.4	3.8
10	33, A	89.6	3.3	1.8	0.4	4.9

element analyzer Flash 2000 (Thermo Fisher Scientific, Great Britain). Analysis results calculated for the organic mass of coal (OMC) are shown in Table 2.

Experimental samples with the bulk density $\rho = 0.5$ g/cm^3 in the form of a portion of 10 mg were placed in a copper capsule 5 mm in diameter and 2 mm deep.

Investigation procedure

The functional scheme of the experimental set-up is presented in Fig. 1.

The source of laser radiation was a pulsed YAG:Nd^{3+}-laser (L) operating in the free generation mode (wavelength $\lambda = 1064$ nm). Pulse duration was 120 μs; the diameter of the incident laser spot on the sample was 2.5 mm. The instability of the energy of the laser pulse did not exceed 2 %. The distribution of radiation intensity over the beam section was quasi-rectangular.

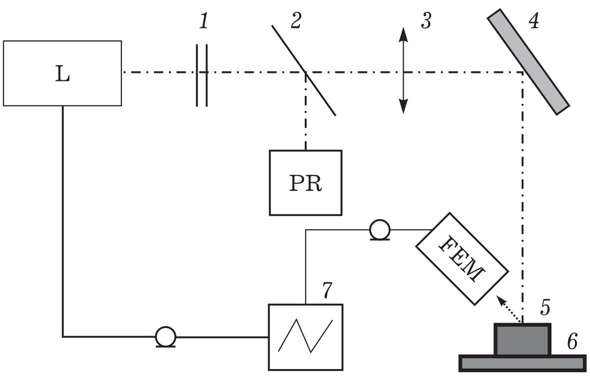

Fig. 1. Functional scheme of the experimental set-up: *1* – neutral light lifters; *2* – beam splitter; *3* – lens; *4* – rotary mirror; *5* – sample; *6* – massive basement; *7* – oscillograph; L – pulsed YAG:Nd^{3+}-laser (1064 nm, 120 μs); PR – pyroelectric receiver; FEM – photoelectron multiplier.

The energy of laser radiation was adjusted with the help of a set of optical glass filters (*1*) with known extinction coefficients. To control the energy, a portion of radiation (8 %) was directed by a beam splitter (*2*) to the pyroelectric receiver PE50BF-C (Ophir Photonics, Israel) (PR). With the help of the focusing lens (*3*) with a focal distance of 25 cm, and a rotary mirror (*4*), laser radiation was directed to coal sample (*5*) placed on a massive basement (*6*). Sample glow arising under the action of pulses was detected with the help of a photoelectron multiplier H-10721-01 (Hamamatsu, Japan, temporal resolution 0.5 ns) (FEM), transformed into the electric signal and recorded with a WJ332A oscillograph (LeCroy, USA) (*7*).

To determine threshold characteristics of coal ignition, ten samples were irradiated sequentially with a single laser pulse of definite energy, and the kinetic dependences of coal glow were recorded with the help of the photomultiplier.

The probability of ignition (P) was determined as

$$P = n/10 \quad (1)$$

where n is the number of recorded flashes.

Then radiation energy was increased, and eth experiment was repeated. As a result, we obtained dependence of the probability of ignition on the density of laser radiation energy for the samples of all the indicated coal marks.

Experimental results were approximated by the probability integral:

$$p(H) = \frac{1}{\sigma\sqrt{2\pi}} \int_{-\infty}^{H} \exp\left(-\frac{(t - H_{cr})^2}{2\sigma^2}\right) dt \quad (2)$$

where H is the density of laser radiation energy; H_{cr} is energy density corresponding to a 50 % probability of ignition (ignition threshold); σ is the square mean deviation.

RESULTS AND DISCUSSION

Three stages of ignition were detected for the samples of all coal marks under study. These stages are characterized by definite threshold values of H_{cr} and glow duration (t, ms).

Dependences of the probability of ignition for coal of D mark on the density of laser radiation energy (the curves of relative frequency) are shown as examples in the inserts (Fig. 2). One can see that three ignition thresholds H_{cr} corresponding to different processes in the samples may be distinguished.

Figure 2 also shows the kinetic dependences of glow intensity (I, V) recorded with the photomultiplier for the flames of D coal with the probability $P = 0.5$ for the three above-indicated processes.

The values of ignition threshold H_{cr} for all coal marks under study are presented in Table 3.

The dependences of ignition threshold values H_{cr} on the coalification degree of coal samples under study are shown in Fig. 3.

Errors of measured H_{cr} values (see Table 3) and confidence ranges (see Fig. 3) correspond to the values of square mean deviation σ in the case if the curves of relative frequency are approximated by equation (2).

One can see that $H_{cr}^{(1)}$ values remain almost constant with an increase in coalification time but they exhibit substantial statistical scattering (see Fig. 3, *a*), while $H_{cr}^{(2)}$ values decrease (see Fig. 3, *b*), and $H_{cr}^{(3)}$ values increase (see Fig. 3, *c*).

The effect of the density of laser energy on the kinetic dependences of flame glow for coal samples was studied in a series of experiments for three detected ignition stages.

The kinetic dependences of glow at the first stage of ignition have a similar nature for the studied coal marks. The duration of glow only insignificantly exceeds the time of laser pulse and reaches 150 μs within the energy density range from $H_{cr}^{(1)}$ to $H_{cr}^{(2)}$ (see Fig. 2, *a*).

Qualitatively similar nature is also traced for the kinetic dependences of glow at the second stage of ignition. Within the energy density range from ~$H_{cr}^{(2)}$ to (2–3)$H_{cr}^{(2)}$, a decrease in glow duration is exhibited by these dependences. The kinetic dependences of glow at the second stage of ignition for D and T coal marks are shown in Fig. 4 as an example.

At sufficiently high densities of laser radiation energy, glow duration decreases to the submillisecond time interval. For instance, the kinetic dependences for coal of T mark measured at different energy densities are shown in Fig. 5. The maximum of glow intensity is achieved within the time t = 110 μs for any energy density involved. Qualitatively similar dependences are observed for all the studied coal marks.

Dependences of the amplitude of glow intensity at the moment t = 110 μs on the density of radiation energy for the samples of five coal marks are presented in Fig. 6.

Measurement of the kinetic dependences of glow in coal flame corresponding to energy densities $H \geq H_{cr}^{(3)}$ showed that glow duration at the third stage of ignition is practically independent of the density of laser radiation energy. It was stressed that the maximal energy density was 15 J/cm^2. However, pulse shape changes even with fixed energy density.

In our previous works [4–6], we linked the sequence of ignition stages and the dependences of threshold ignition $H_{cr}^{(i)}$ values on coalification degree with the occurrence of thermochemical processes described below.

At the first stage of ignition, the surface of coal particles is heated, and the ignition of microasperities occurs. The second stage of ignition is connected with the release and ignition of volatile substances in the gas phase, while the third stage is due to the ignition of non-volatile residues of coal particles. These conclusions are confirmed by the data reported in [10] and the results of our experiments [4–6].

It was demonstrated in [10] that ignition of a particle of black coal under the action of laser radiation is initiated on microasperities. A dependence of the time of coal particle ignition and the minimal laser radiation intensity necessary for coal particle ignition on the number of nonuniformities on the surface of coal particle [10]. So, the ignition of microasperities is connected with particle size and the geometry of the particle surface, which are characterized by substantial statistical scattering and finally give $H_{cr}^{(1)}$ values measured in the experiments. To remind, we used coal samples with particle size less than 63 μm in this work. It may be the process of microasperity ignition that makes the decisive contributionnto $H_{cr}^{(1)}$ and leads to a weak dependence (or its absence) on the degree of coalification of the studied coal samples. In addition, as electron-optical studies showed, the used coal samples are characterized by broad size distributions with a substantial amount of particles of ~1 μm in size. Because of this, in addition to microasperities on

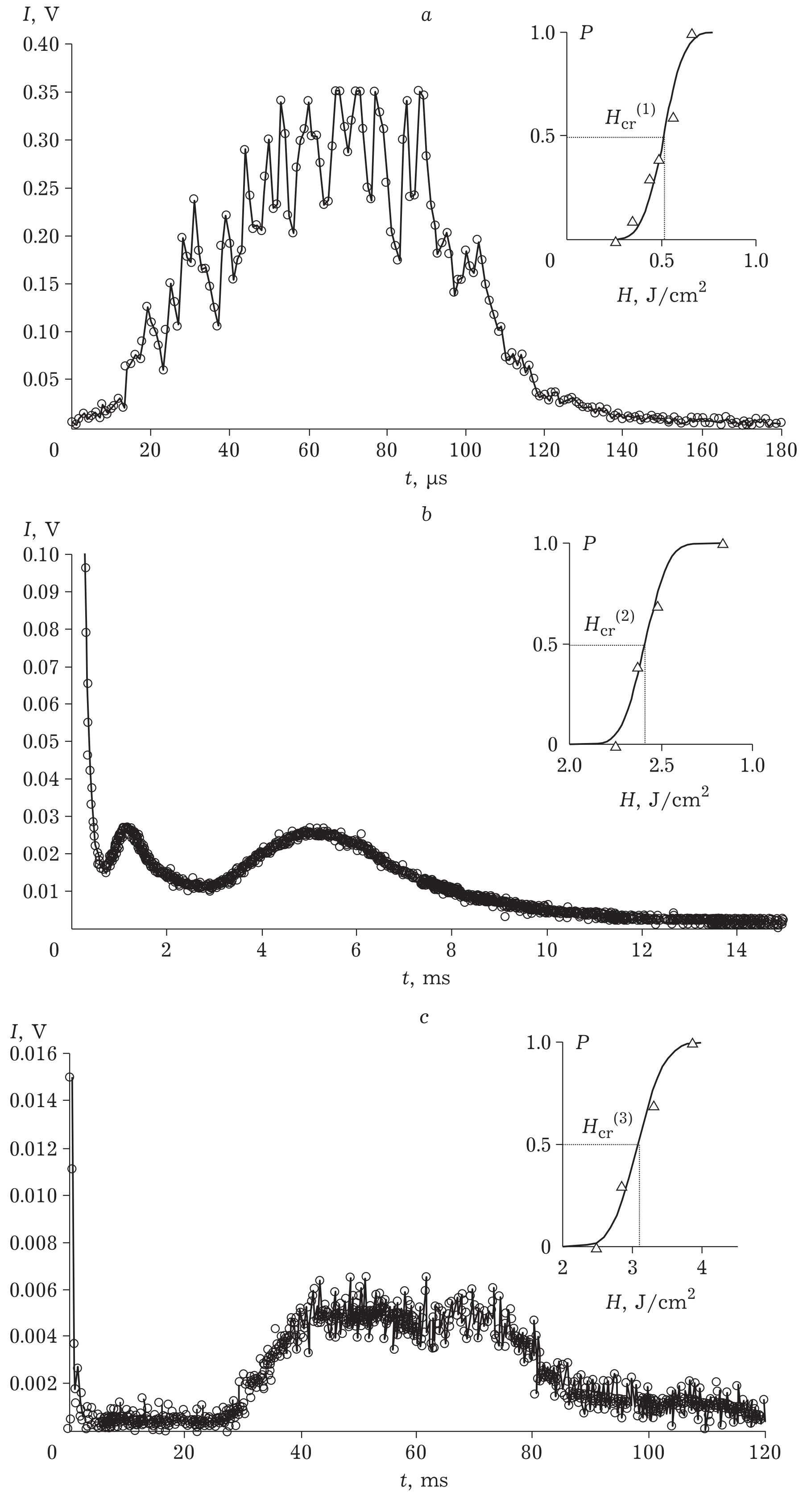

Fig. 2. Kinetic dependences of glow intensity (I, V) in the flames of coal of D mark with the probability $P = 0.5$ for three stages of ignition: $H_{cr}^{(1)} = 0.51$ J/cm^2 (a), $H_{cr}^{(2)} = 2.41$ J/cm^2 (b), $H_{cr}^{(3)} = 3.08$ J/cm^2 (c). Inserts: dependences of the probability (P) of ignition of the coal of D mark on the density of laser radiation density (H).

the particles of relatively large sizes, the first stage of ignition is also initiated by the ignition of coal particles with a size of ~1 μm.

To explain the dependences of the second $H_{cr}^{(2)}$ and the third $H_{cr}^{(3)}$ thresholds of coal ignition on coalification degree, at the present stage of investigation similarly to [4–6], a model described below is proposed.

Initiation of chemical reactions is connected with the absorption of laser radiation energy. With an increase in coalification degree, the absorption coefficient (k) of coal increases [11]. The temperature of laser ignition (T_{ig}) is almost constant in the metamorphism sequence [12]. Specific hear capacity (c) of coal samples under study varies insignificantly, for example, the true thermal capacity of rough coal of mark D at a temperature of 300 K is c ~ 1.11 kJ/(kg · K), and the corresponding value for mark A is c ~ 0.83 kJ/(kg · K) [13]. The true density of coal samples under study also varies within a narrow range: the true density of the coal of D mark is ρ_{true} ~ 1.40 g/cm³, while for the coal of A mark it is ρ_{true} ~ 1.32 g/cm³ [14]. So, the volume density (Q_v) of absorbed energy to achieve T_{ig} changes only slightly.

$$c\rho\Delta T = Q_v \quad (3)$$

On the other hand,

$$Q_v = Hk \quad (4)$$

It follows from these considerations that $H_{cr}^{(2)}$ decreases with an increase in coalification degree and absorption coefficient k.

At the third stage of ignition, with an increase in coalification degree, an increase in $H_{cr}^{(3)}$ is observed. So, dependence may be traced which is qualitatively opposite to the second stage of ignition. At the third stage of ignition, coal particles are heated to T_{ig} as a result of the action of two factors: 1) heating due to the absorption of laser pulse energy; 2) coal particles are heated as a result of the combustion of the gas phase. With

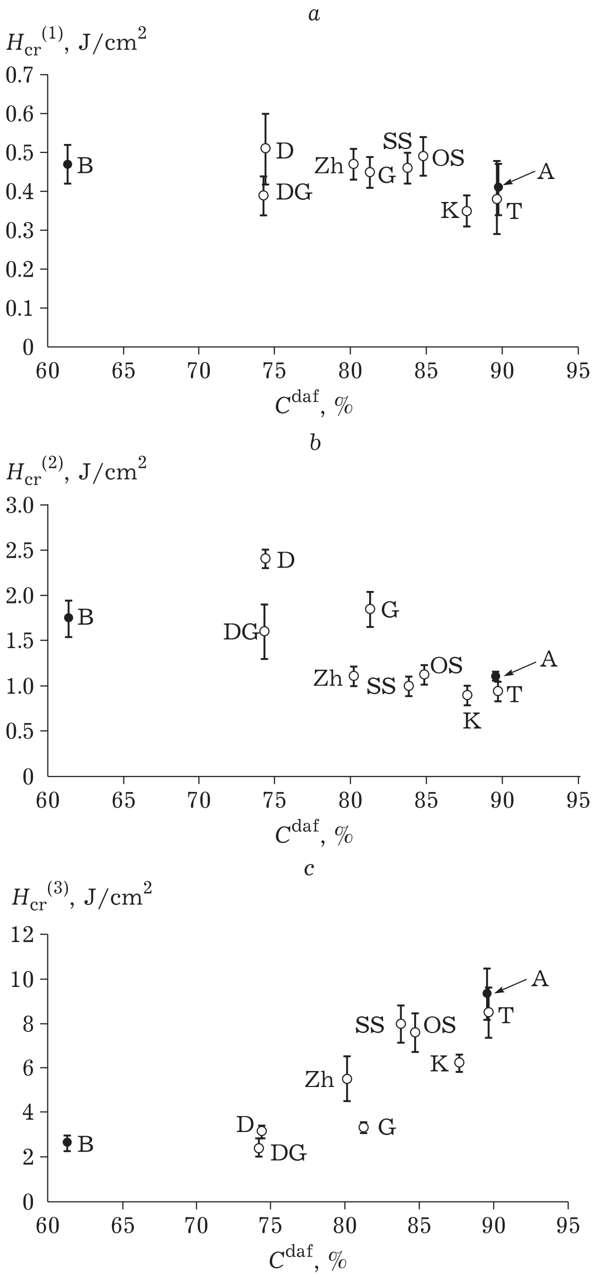

Fig. 3. Dependences of ignition thresholds $H_{cr}^{(1)}$ (*a*), $H_{cr}^{(2)}$ (*b*), $H_{cr}^{(3)}$ (*c*) on coalification degree (C^{daf}, %) of the studied coal samples.

TABLE 3

Values of ignition thresholds H_{cr} for the studies coal samples

Sample No.	Sample No., coal mark	$H_{cr}^{(1)}$, J/cm²	$H_{cr}^{(2)}$, J/cm²	$H_{cr}^{(3)}$, J/cm²
1	27, B	0.47±0.05	1.7±0.2	2.6±0.3
2	72, D	0.51±0.09	2.4±0.1	3.1±0.3
3	64, DG	0.39±0.05	1.6±0.3	2.4±0.4
4	40, G	0.45±0.04	1.8±0.2	3.3±0.2
5	15, Zh	0.47±0.04	1.1±0.1	5.5±1.0
6	10, K	0.35±0.04	0.9±0.1	6.2±0.4
7	34, OS	0.49±0.05	1.1±0.1	7.5±0.8
8	45, SS	0.46±0.04	1.0±0.1	7.9±0.8
9	81, T	0.38±0.09	0.9±0.1	8.5±1.1
10	33, A	0.41±0.07	1.10±0.05	9.3±1.1

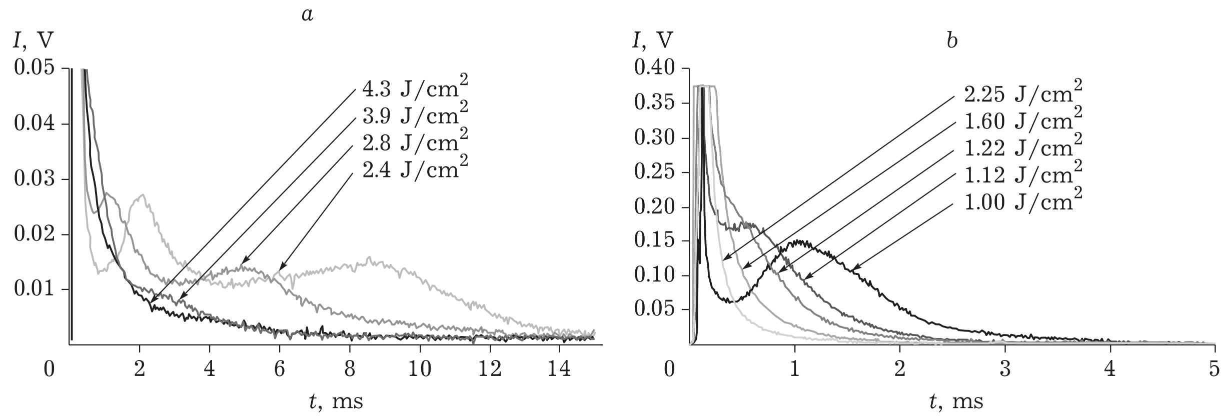

Fig. 4. Kinetic dependences of glow of coal samples of D mark (*a*) and T mark (*b*) at the second stage of ignition under the action of laser pulses with energy density $H > H_{cr}^{(2)}$.

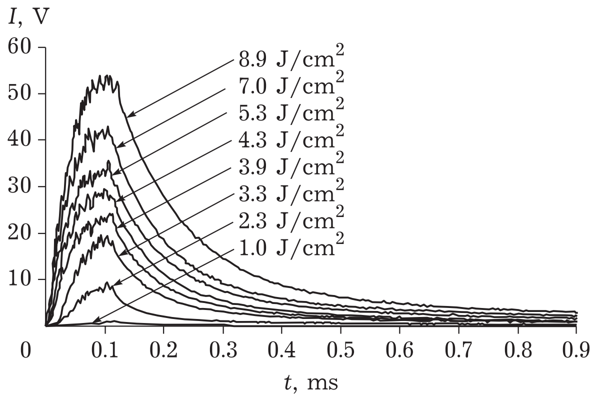

Fig. 5. Kinetic dependences of glow in the submillisecond time range for coal of T mark, measured at different densities of laser radiation energy.

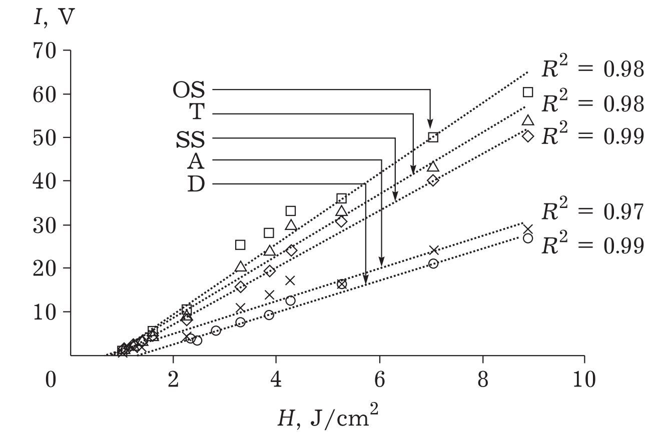

Fig. 6. Dependences of the amplitude of glow intensity on the density of radiation energy for coal marks OS, T, SS, A, D (glow duration $t = 110$ μs).

an increase in coalification degree, the mass of burnt volatile substances decreases, which correspondingly leads to relatively lower heating of coal particles. To achieve T_{gi}, it is necessary to increase the energy of laser pulse, which leads to the observed dependence of $H_{cr}^{(3)}$ on coalification degree.

An explanation given below may be proposed for the obtained dependences of threshold $H_{cr}^{(2)}$ and $H_{cr}^{(3)}$ values on coalification degree. It is known that a coal molecule is composed of aromatic carbon rings linked to each other with the help of aliphatic linear chains. The linear aliphatic chains are also bound to radicals and form a higher reactive peripheral part of the molecular structure of coal [15]. The core of the molecule possesses the highest bond strength and thermal stability; the side aliphatic groups with different polymerization degrees are characterized by relatively lower stability. Because of this, the destruction of aliphatic chains occurs at the second stage with energy density $H_{cr}^{(2)}$. The emission from excited H_2, H_2O molecules and carbon particles is observed in the spectra of the flame [6]. With an increase in coalification degree, the amount of aromatic carbon increases, while the amount of aliphatic carbon decreases. A small decrease in $H_{cr}^{(2)}$ may be connected with the individual structure of coal molecules or, as assumed above, with an increase in absorption coefficient *k*.

Then we may assume that the third stage proceeding at higher energy densities is connected with the destruction of thermally stable aromatic cores of molecules, followed by ignition at $H_{cr}^{(3)}$. From this point of view, an increase in $H_{cr}^{(3)}$ with an increase in coalification degree is quite clear, since the degree of aromaticity increases in the metamorphism sequence.

Then the observed kinetic dependences of glow at the second stage of ignition in the millisecond time interval may be explained as follows. With an increase in the density of radiation energy $H > H_{cr}^{(2)}$ coal heating increases, which results in an increase in the rates of thermochemical reactions in aliphatic chains. This leads to the decomposition of the latter, the release and ignition of volatile substances, as well as to the ignition of carbon particles.

Measurements of the glow of coal flames in the submillisecond time range (see Fig. 5) showed that for $H > H_{cr}^{(3)}$ ignition of coal particles is observed directly during irradiation pulse. The intensity of flame glow increases linearly with an increase in the density of laser radiation energy. The attenuation of radiation within the submillisecond range is not described by the exponential law. Attenuation time (a decrease in the intensity to the value corresponding to the half of the amplitude) increases within the range $\Delta t = 29-130$ μs with an increase in energy density within the range $\Delta H = 1.0-8.9$ J/cm^2.

The observed dependence of glow intensity on energy density (see Fig. 6) provides evidence that laser pulses within the used power range do not involve the processes connected with the optical breakdown, chain reactions, etc., that would lead to the nonlinear dependence of the flame glow intensity on energy density.

CONCLUSION

Three stages of ignition having threshold nature were distinguished in the laser ignition of fine (less than 63 μm in size) particles of coals of different marks. The first stage of ignition is connected with heating of the surface of coarse coal particles and ignition of microasperities along with the ignition of particles ~1 μm in size present in the sample. The second stage of ignition is characterized by the release and ignition of volatile substances, as well as the ignition of carbon particles that are most probably formed through the destruction of aliphatic chains. The third stage of ignition involves thermochemical reactions in the aromatic part of the coal macromolecule leading to the ignition of non-volatile coal residue

With an increase in coalification degree, the values of the first ignition threshold $H_{cr}^{(1)}$ remain almost unchanged, the values of the second ignition threshold $H_{cr}^{(2)}$ decrease, while the values of the third ignition threshold $H_{cr}^{(3)}$ increase.

The duration of glow at the first stage of ignition exceeds the duration of laser pulse only insignificantly and reaches 150 μs for all coal marks. Glow duration at the second stage of ignition at $H = H_{cr}^{(2)}$ is within the millisecond time interval. At the third stage of ignition with $H = H_{cr}^{(3)}$ glow duration is within 10–100 μs for the samples of the studied coal marks.

With an increase in the density of laser radiation energy, glow duration at the second stage of ignition decreases as a result of an increase in the rates of thermochemical reactions and reaches the submillisecond range.

For all coal marks under study, glow intensity increases since the moment of laser pulse action. A decrease in glow intensity is observed in the submillisecond range. The amplitude of coal glow increases linearly with an increase in the density of laser ration energy.

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