

PACS numbers: 82.33.Vx; 42.55.Lt
DOI: 10.1070/QE2012v042n01ABEH014722

Initiation of ignition of a combustible gas mixture in a closed volume by the radiation of a high-power pulsed CO_2 laser

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Abstract. **The results of experiments on initiating the ignition of a CH_4–O_2–SF_6 triple gas mixture in a closed volume by the radiation of a high-power CO_2 laser are presented. It is shown that spatially nonuniform (in the direction of the laser beam) gas heating by the laser radiation leads to formation of a fast combustion wave, propagating along the chamber axis and giving rise to 'instantaneous' ignition. At the threshold value 16.5 J of the laser radiation energy the fast combustion wave is transformed into a detonation wave, which causes an explosion and destruction of the reaction chamber**

Keywords: *initiation of ignition, combustible gas mixtures, laser heating, detonation wave, deflagration wave*

1. Introduction

Great interest in the problem of initiating the combustible gas mixture ignition, observed recently (see, e.g., the published review [1]), is stimulated both by the necessity to satisfy the requirements of current technology and by the development of fundamental studies of formation, maintaining and propagation of flames.

Two groups of studies may be selected among those devoted to mechanisms of plasma influence on the ignition of combustible mixtures. In the first one (see [2]) the attention is focused on the character of inflammation in the volume simultaneously subjected to short-time action of gas-discharge plasma. In the second group [3–9] the dense plasma is localised in a relatively small volume and affects the process of ignition in the surrounding combustive gas mixture not involved directly in the gas discharge process. In the latter studies the role of local ignition initiators was played by high-power high-current slipping surface discharges [3, 4], microwave discharges [5, 6], laser sparks at the metal and dielectric surface [7], as well as laser sparks in free space [8, 9]. The results of the experiments have shown that when the power of the used electric discharges considerably exceeds the power of common spark igniters (sparking plugs) the character of combustion waves (deflagration) and the waves described by the traditional 'thermal' model strongly differs [10]. Moreover, it was shown that the velocity of the combustion waves appears to be significantly higher than $V = V_n\rho_0/\rho_h$ (where V_n is the normal velocity of the combustion wave, ρ_0 and ρ_h are the gas densities before and after the combustion wave front) and considerably grow with increasing power and energy, deposited into the discharge volume. Unfortunately, in Refs [3–9] the basic physical and chemical processes that determine the properties of high-velocity combustion waves were not specified.

The present work is a continuation of studies performed in [3–9] with CO_2 laser radiation involved as an initiator of combustible mixture ignition. The scheme of the experiment is such that the process of initiating the ignition is determined by the local heating of the gas mixture, thus eliminating a number of factors, accompanying the electric discharge (e.g., chemical reactions with charged particles and UV radiation, exciting the surrounding medium). In addition, an important feature of laser initiation of the combustion wave in free space is the absence of electrodes and other elements necessary to organise electric discharges. This, in turn, essentially affects the characteristics of gas-dynamic processes that accompany propagation of combustion waves when using such methods of initiating the ignition.

The effect of laser radiation on combustible mixtures was investigated in some papers (see, e.g., [11–15]). However, in these studies the attention was mostly paid to the process of ignition and maintaining the stable combustion of the mixtures under study, initiated by the absorption of laser radiation. The issues of formation of combustion waves and the velocity of their propagation were left open.

The goal of the present paper is to study the formation and propagation of fast combustion waves in flammable mixtures, excited by the radiation of a high-power CO_2 laser, as well as to clarify the effect of the given initial profile of the gas temperature, produced by the laser action, on the characteristics of combustion waves.

2. Experimental

The scheme of the experiment is shown in Fig. 1. The cylindrical silica camera (*1*) (inner diameter 44 mm, length 25 cm) was evacuated up to the pressure $p \leqslant 0.1$ Torr and filled with combustible gas mixture $CH_4:O_2:SF_6 = 45:90:15$ with total pressure $P = 150$ Torr. The faces of the camera were closed with flanges. One of them was blind metallic, and the other had a BaF_2 window, through which the laser radiation was let into the camera. Different apertures and calibrated filters were placed in front of the window to attenuate the laser radiation.

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Received 16 Sepember 2011; revision received 2 November 2011
Kvantovaya Elektronika **42** (1) 65–70 (2012)
Translated by V.L. Derbov

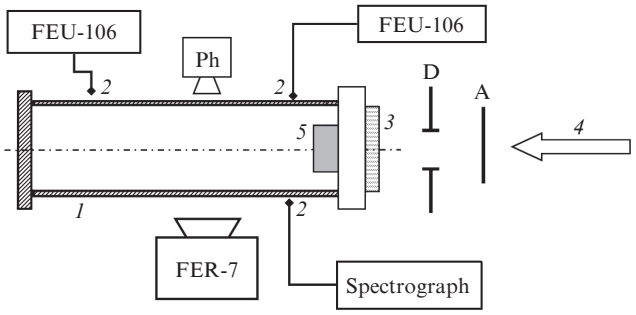

Figure 1. Scheme of the experimental setup: (*1*) reactor chamber (silica tube); (*2*) optical fibres; (*3*) BaF_2 window; (*4*) CO_2 laser beam; (*5*) schematic image of the gas region having the initial temperature higher than 1000 K; (Ph) photodiode; (D) circular aperture; (A) calibrated attenuators of the laser radiation.

The pulsed electric-discharge CO_2 laser [16] operating at the P(20) line ($\lambda = 10.6$ μm) was used in the experiments. The maximal laser radiation energy in the pulse was 60 J, the radiation pulse shape being typical of CO_2 lasers with transverse discharge pumping and having the duration $\tau_{0.1} \approx 3$ μs at the 0.1 level [17]. The laser radiation energy distribution over the beam cross section within the zone selected by an aperture was close to uniform.

The addition of a small amount of SF_6 (15 Torr), efficiently absorbing the CO_2 laser radiation, to the combustible gas mixture allowed the gas mixture heating. The gas temperature and its distribution along the axis of the cell were varied by changing the laser radiation energy density, incident on the gas. As shown by the analysis [17], the time, during which the energy stored in the vibrational degrees of freedom of SF_6 molecules is transformed into heat, is less than 1 μs under the conditions of the described experiment. Therefore, before the end of the laser pulse all the energy absorbed by the gas is completely transformed into heat. The temperature distribution along the cell axis immediately after the laser pulse was measured as follows. The dependences of the transmission of CO_2 laser radiation through gas cells with the absorbing length 19 and 29 mm upon the radiation energy density at the cell entrance were measured (in a way similar to [17]). The mixture $SF_6:CH_4:N_2 = 30:45:90$ Torr was let into the cells. The validity of replacing the combustible $SF_6–CH_4–O_2$ mixture with the $SF_6–CH_4–N_2$ mixture was checked in a separate experiment. It was found that at the partial pressure of N_2 or O_2 up to 200 Torr the transmission of the mixtures $SF_6–N_2$ and $SF_6–O_2$ does not differ from that of SF_6 (at the same partial pressure as in the mixture). The measurements of transmission of radiation through the cells with the abovementioned absorption lengths allowed us to obtain the dependence of CO_2 laser radiation energy absorbed by the 10-mm-thick layer of the gas mixture on the energy density of the radiation incident on this layer. To get the temperature distribution along the chamber axis, the whole volume of the gas was divided into 10-mm-thick layers, the energy absorbed by each layer was calculated, and the gas temperature in the layer was calculated by analogy with [16]. The data on the temperature dependence of the heat capacity were taken from Ref. [18], the processes of heat exchange between the gas layers and the walls of the gas chamber were not taken into account, because the time of laser pulse action was relatively small (~3 μs).

To analyse the ignition dynamics of the combustible mixtures and propagation of combustion waves, we used the FD-25k photodiode, the FEU-106 photoelectric multipliers, and the FER-7 high-speed photorecorder that allows taking photos of the glow from the reactor in the continuous scanning regime. The slit of the photorecorder was positioned along the axis of the cylindrical chamber (z axis). To analyse the gas glow and the temporal evolution of the glow intensity in different cross sections along the silica tube, we used the AvaSpec-2048 (AVANTES) spectrograph and FEU-106 photomultipliers, respectively. The gas radiation from the silica tube was transported to the entrance slit of the spectrometer and photomultiplier by optical fibres.

3. Experimental results

In the course of experiments it was found that for each value of the aperture diameter, limiting the transverse size of the laser beam before the entrance window of the reaction chamber, two values of the threshold laser radiation energy density exist (W_{1thr} and W_{2thr}), at which either 'nonexplosive' ignition of the combustive mixture, or the ignition accompanied by the explosion of the chamber (detonation regime) occurs. The fact that in the second regime of ignition the detonation takes place is justified below (see Section 4). Figure 2 presents the photographs of the glow from the reactor chamber, shot using the FER-7 photoelectric recorder in the case of 'nonexplosive' ignition of the gas mixture (Fig. 2a) and in the case, when the ignition is accompanied by detonation and the chamber explosion (Fig. 2b). The horizontal axis of the photograph is the temporal axis and the vertical axis is spatial. The characteristic temporal and spatial scales are presented in the pictures. The moment of time $t = 0$ corresponds to the appearance of the glow in the 'field of view' of the photoelectric recorder; τ_1 is the temporal interval during which the 'pri-

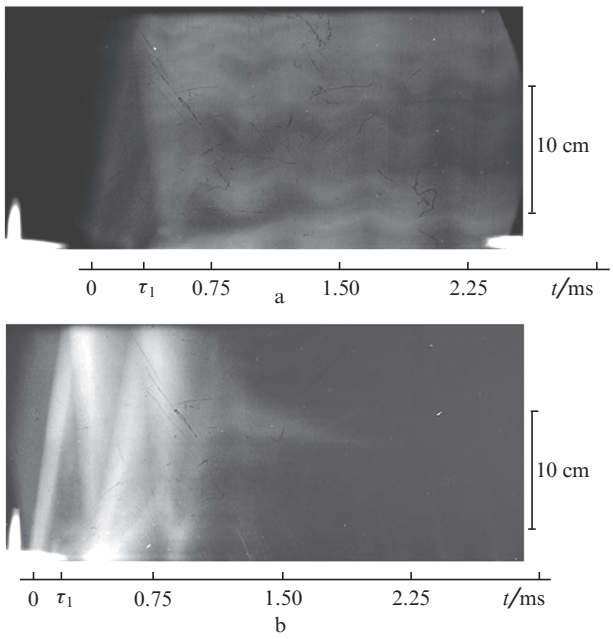

Figure 2. Photographs of the glow from the burning methane–oxygen mixture, shot using the photoelectric recorder. The diameter of the aperture before the entrance window is 29 mm, the laser radiation energy density at the reactor entrance is ~1.6 (a) and 2.5 J cm^{-2} (b).

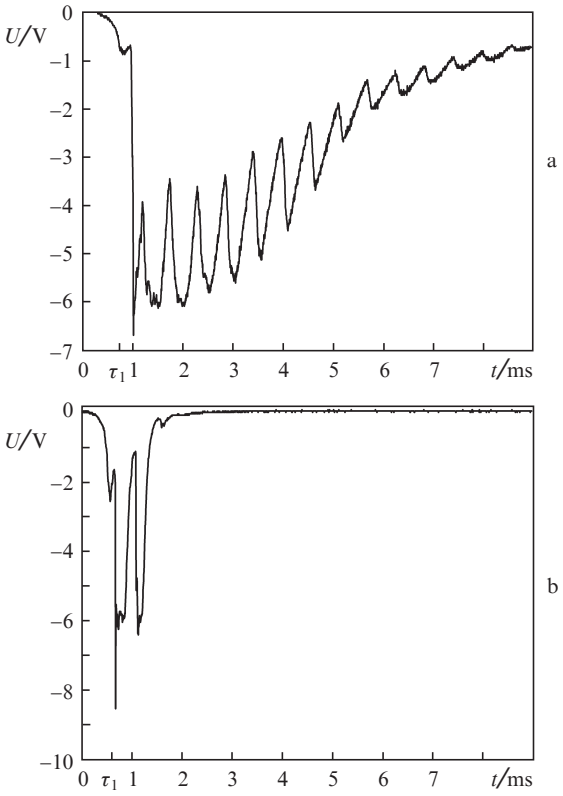

Figure 3. Oscillograms of the photomultiplier signal at $W \approx 1.6$ (a) and 2.5 J cm^{-2} (b)

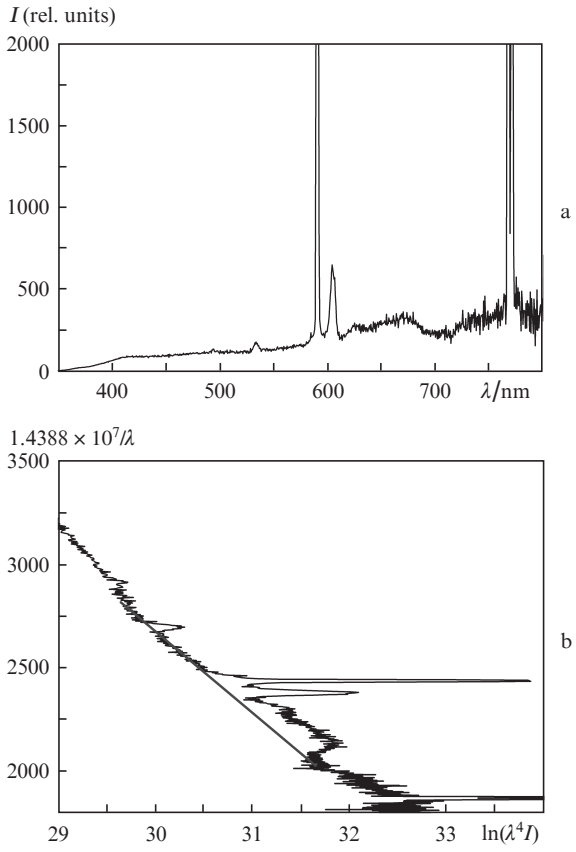

Figure 4. Spectral characteristics of the radiation from the combusting methane–oxygen mixture, integrated over time 1.2 ms from the moment of initiation (a) and the same spectrum in the coordinates $x = \ln(I\lambda^4)$ and $y = 1.4388\ 10^7/\lambda$ (*б*); $T_g = 4000$ K.

mary' wave is recorded, preceding the bulk ignition of the mixture in the reactor. The oscillograms of the signals recorded by the photoelectric multiplier in the same two cases are presented in Fig. 3.

Figure 4a shows a typical spectrogram of the radiation from the ignited mixture in the range of wavelengths $400 \leqslant \lambda \leqslant 800$ nm (integrated over 1.2 ms starting from the initiation moment) for the case of 'nonexplosive' ignition, which represents a combination of continuous and discrete line spectra. The brightest emission lines in the spectrum are the doublets of sodium and potassium. Sodium and potassium are impurities of silica, the material of the reactor walls. The spectral range shown in Fig. 4a does not cover the region where the emission band $A^2\Sigma^+$ of electron-excited OH molecules (the edge near $\lambda \approx 306$ nm) is located. This band significantly contributes to the spectrally integral intensity of radiation, recorded by FER-7 (Fig. 2) and FEU (Fig. 3) [9]. The sources of the quasi-continuous spectral component are not identified. However, the analysis shows that this radiation has nearly a Planck distribution and can be used to determine the gas temperature following the method described in [19]. The result of procession of the recorded spectrum following this method is presented in Fig. 4b with the coordinates $x = \ln(I\lambda^4)$ and $y = 1.4388 \times 10^7/\lambda$ (I is the number of photons, detected at the wavelength λ, measured in nanometres). The obtained dependence allows identification of the continuous part of the spectrum with the spectrum of Planck radiation and estimation of the gas temperature averaged over the combustion time, which yields $T_g \approx 4000$ K.

Figure 5 shows the temperature distribution of the gas mixture along the axis of the cylindrical chamber, established just after the end of the CO_2-laser pulse at the stage, preceding the ignition of the mixture (the distance is measured from the entrance window of the chamber). The calculation is performed for two values of the radiation energy density incident onto the gas, at which the stable ignition ($W = 1.6$ J cm^{-2}) or the detonation of the combustible mixture ($W = 2.5$ J cm^{-2}) occurred. The diameter of the circular aperture, placed before the entrance window, was equal to 29 mm. As seen from Fig. 5, the gas is heated by the laser radiation through a significant distance from the entrance window of the chamber.

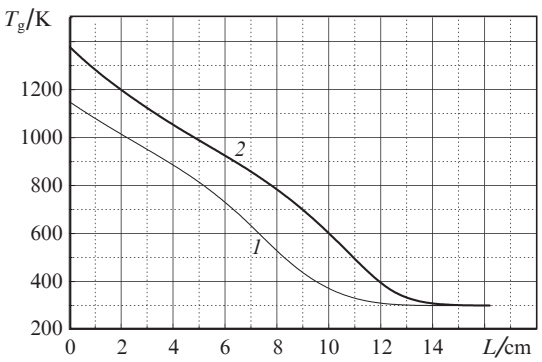

Figure 5. Distribution of the gas temperature T_g along the coordinate L (the axis of the reactor chamber) at the end of the laser pulse for the CO_2 laser radiation energy density 1.6 (*1*) and 2.5 J cm^{-2} (*2*).

By varying the aperture diameter within 18–32 mm, the threshold values of the radiation energy density W_{1thr} and W_{2thr}, at which the 'nonexplosive' ignition of the combustible mixture or the ignition with explosion occurs, respectively, changed nearly in inverse proportion to the aperture area, i.e., the threshold values of the laser radiation energy E_{1thr} and E_{2thr}, providing the abovementioned regimes of the mixture ignition, are independent of the aperture diameter. In the present experiments, we obtained $E_{1thr} = 10.5$ J and $E_{2thr} = 16.5$ J. Thus, the possibility of ignition of the combustible mixture is determined (in agreement with [20]) by the initiation energy.

4. Discussion of the results

As follows from the above experiment, the initiation of ignition of the CH_4–O_2–SF_6 mixture in a closed volume under heating by a pulsed CO_2 laser obeys the same laws that have been found earlier in the experiments using other initiators, namely, strong-current slipping surface discharge [3, 4], microwave discharges [5, 6], laser spark at the surface and in free space [7, 9]. The fast heating up to 1200–1400 K of the gas layer at the reactor entrance by the laser radiation is accompanied by the wave with a relatively weak glow ('primary' wave), propagating from the initiator along the chamber axis. The velocity of this wave is close to $V \approx 10^5$ cm s^{-1}, which essentially exceeds the velocity of deflagration wave in the stoichiometric mixture CH_4–O_2 and is comparable with the velocity of a detonation wave (according to [10], the visible velocity of the combustion wave in methane–oxygen mixture, whose propagation is determined by the diffusion-heat-conduction mechanism, does not exceed 4×10^3 cm s^{-1}).

After the time, long enough to let the 'primary' wave to 'run' through the entire chamber, in the net volume of the latter a bright flash is observed that characterises the processes of fast ('explosive') combustion. The time τ_{ign} of the combustive mixture ignition is counted from the moment of laser pulse action to the moment of attaining the maximum of the glow intensity, recorded by FEU (Fig. 3). The axial velocity of the combustion wave propagation in the present experiment was determined by using the x–t dependences of the position of the glow front in the high-speed photographs, analogous to those presented in Fig. 2.

In Ref. [21] the ignition of methane–oxygen mixture at $P_0 = 1.65$ atm in a 32-mm-long tube with the radius 5 mm was investigated by numerical modelling. The setting of the problem, namely, the formation of a fast combustion wave by a heat source localised in space, was close to the conditions of our experiments, and the results of calculations [21] qualitatively agree with the data, obtained by us. In particular, the formation of a fast combustion wave is observed (whose velocity approaches 2×10^4 cm s^{-1}), which is due to intense gas-dynamic processes, initiated by heat generation in the closed volume. It is worth noting that in the calculations [21] the initial temperature of the gas in the region of ignition was 4000 K, while in our experiments the initial temperature T_g did not exceed 1400 K, as was already mentioned above. The values of the ignition time $\tau_{ign} \leqslant 500$–600 μs, observed in our experiments, are small enough for methane–oxygen mixtures (at $T_g \leqslant 1400$ K and the pressure 200 Torr the ignition time for the stoichiometric CH_4–O_2 mixture is greater than 6 ms [22]). However, under the considered conditions one should take into account the presence of SF_6 in the combustible mixture and the possible dissociation of these molecules by the CO_2 laser radiation. In Ref. [23] the results of measuring the induction times for the mixture CH_4:O_2:SF_6 = 7:14:1 at P = 100 Torr, excited by two sequential CO_2 laser pulses, are presented. The first pulse provided heating of the mixture and the second one caused its ignition. It was shown that at the initial temperature $T_g = 1400$–1430 K the ignition time for the considered mixture is 200–600 μs (depending on the degree of gas heating in the first pulse), which agrees with the results of our measurements. The induction time was determined as the time interval between the end of the second laser pulse and the flash of emission from the site of the pulse action, recorded by the FEU. Moreover, based on the comparison of the induction times for the secondary pulses with similar radiation energy density but different duration, the conclusion is drawn about the possible influence of chemically active particles, produced by the laser action, on the ignition of the considered mixture.

One of the most interesting results obtained in the present experiment and not demonstrated so apparently in the preceding work [3–9] using other systems of electric-discharge and laser initiation is the attainment of detonation regime by the arising combustion wave under the condition that the energy of laser radiation exceeds a certain threshold value. As already mentioned above, the velocity of the combustion wave, formed under the considered conditions ($V \approx 10^5$ cm s^{-1}), considerably exceed the velocity of sound, which, according to the existing concepts [24–31], provides premises for transforming the deflagration wave into a detonation one (deflagration-to-detonation transition, DDT). This transition manifests itself by substantial reduction of the glow duration of the combusting mixture and the increased intensity of this glow (see Figs 2b and 3b), as well as by the explosive destruction of the reactor chamber. According to the results of special studies, at the initial pressure $P_0 \leqslant 200$ Torr of the gas mixture the destruction of the silica reactor requires fast increase in pressure by nearly 80–100 times. In our opinion, under the present conditions only a detonation wave may provide such a fast and intense increase in pressure, because such a pressure jump is possible at the front of a shock wave with the Mach number $M \geqslant 7$ and cannot be implemented under the adiabatic heating of the gas. The attainment of detonation at relatively small energy deposit into the initiation region (less than 20 J) seems to be a rather unusual and hardly implementable phenomenon [26].

A considerable number of theoretical studies (see, e.g., [27–32]) and experimental work (see [33, 34]) are devoted to the physics of the transition from combustion to detonation. The models presented in [27–32] involve different mechanisms of acceleration of the combustion wave up to the velocities, exceeding the velocity of sound, with subsequent fast transition of the combustion wave into a detonation one.

Among the considered mechanisms of acceleration of deflagration waves, which at the moment of birth have the velocity much smaller than that of sound, the following ones are worthy of special attention. First of all, this is the mechanism of a fast combustion wave formation in a gas medium with axial temperature gradient. This model, proposed by Zel'dovich et al. [27] and developed numerically in [28], predicts the formation of combustion waves that have the character of 'phase' waves. At a certain distribution of the gas temperature these waves can acquire the velocity exceeding that of sound. In Refs [30, 31] the processes, taking place in the near-wall region and promoting the increase in the combustion wave front area and the relevant acceleration, are considered as mechanisms that determine the acceleration of

deflagration waves in the cylindrical chamber. Finally, the authors of Refs [29, 30] emphasise the essential role of shock waves, formed in the region of ignition initiation, whose reflection from the walls and multiple interaction with the combustion site leads to acceleration of the deflagration wave up to the velocities that may exceed the velocity of sound.

No experiments aimed at the investigation of the process of transformation of deflagration into detonation, based on the gradient mechanism by Zel'dovich [27], are known to us. Their absence may be related to the difficulties of preparing a gas temperature profile with prescribed parameters in the combustible mixture. From this point of view, the scheme described in the present paper may be considered as one of very few possibilities to test experimentally the Zel'dovich mechanism of the deflagration-to-detonation transition. Here the axial gradient of the initial temperature can be, in principle, varied within large enough limits via the variable content of SF_6 that absorbs the radiation of the CO_2 laser. However, in the discussed experiment only one composition of the gas mixture was used with the corresponding initial temperature distributions, presented in Fig. 5. Using this distribution, the phase velocity of the deflagration wave, which can be evaluated as $v_{\text{def}} \approx \partial z/\partial \tau_{\text{ind}}$ [z being the axial coordinate and τ_{ind} being the induction time at the given value of $T(z)$], appears to be equal to $\sim 10^2$ cm s^{-1}, which is essentially smaller than observed in the experiment.

It should be noted that, strictly speaking, the Zel'dovich model [27] deals with the gradient of the induction time of combustible mixtures rather than with the temperature gradient. The induction time gradient formation is affected both by the profile of the gas temperature and by the profile of concentration of chemically active particles. This was the case implemented in our experiments, since the action of high-power radiation of CO_2 laser on methane–oxygen mixtures with some SF_6 added causes not only heating of the mixture, but also SF_6 dissociation, leading, as already mentioned, to the production of chemically active radicals.

The mechanism of deflagration wave acceleration, considered in [29–32] and associated with strong gas-dynamic perturbations, accompanying the electric-discharge or laser initiation of the gas mixture ignition, seems to be closest to the experiments, described in the present paper (and also in Refs [3–9]). Recognising the apparent necessity of new experimental data, specifying the mechanism of deflagration wave acceleration, we again draw attention to the fact of obtaining very high velocities of the combustion wave at very small (from a few centimetres to ten centimetres) distances from the ignition initiation site. Comparing the present experiment with the preceding ones [3–9], we conclude that the use of CO_2 laser radiation provides the greatest ($\sim 10^5$ cm s^{-1}) velocities of the combustion wave at the smallest distances from the initiation site. The result obtained in the present experiment, namely, the formation of a detonation wave, completely corresponds to the requirement of the DDT model that implies the growth of the velocity of initially slow deflagration wave up to supersonic values.

In conclusion, we draw attention to the fact that the weak glow waves, preceding the bulk ignition, can be considered to be deflagration waves rather tentatively. Actually, these are waves, to which a more adequate definition given in [3–9] applies, namely, the waves of 'incomplete combustion'. However, even at this stage the temperature behind the front of the 'incomplete combustion' attains 1500–2000 K [22], which is significantly lower than the temperature of combustion for the stoichiometric methane–oxygen mixture, but, apparently, quite enough for transforming the combustion into detonation.

5. Conclusions

In the reported experiments we carried out the ignition of the combustible gas mixture (CH_4–O_2–SF_6) using the radiation of a CO_2 laser. As in the preceding works with high-power electric-discharge initiation, we observed a fast wave of 'incomplete combustion' propagating from the initiation site into the chamber and, finally, producing the bulk ignition of the gas mixture in the reactor. The velocity of this wave exceeds that of sound, which creates the prerequisites for its transformation into a detonation wave. At the threshold value of the CO_2 laser radiation energy $E_{2\text{thr}} = 16.5$ J the deflagration wave turns into a detonation one, which, in particular, is detected by the explosion and destruction of the reactor chamber. Thus, in the experiment the possibility to implement the process of transformation of combustion into detonation at relatively low (compared to the DDT threshold [26]) energy deposit into the region of the ignition initiation (equal to or less than 20 J) is demonstrated.

Acknowledgements. The work was partially supported by the Russian Foundation for Basic Research (Grant No. 11-02-00465) and the Program of the Presidium of the Russian Academy of Sciences 'Fundamental problems of mechanics of interactions in technological and natural systems'.

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