International Journal of Engineering Science Invention
ISSN (Online): 2319 – 6734, ISSN (Print): 2319 – 6726
www.ijesi.org Volume 3 Issue 10 ǁ October 2014 ǁ PP.06-12

Heterogeneous Combustion Wave as an X-Ray Source

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ABSTRACT : *By the example of a nanodispersed Ti-B system, it was found for the first time that a heterogeneous combustion wave is an X-ray source with a quantum energy of up to 30 keV. The radiation is generated in individual reaction centers of size 2–5 mm. Among probable causes for the effect can be extreme electron acceleration inside nonequilibrium electric layers formed in the heterogeneous system during high-rate chemical transformation.*

KEYWORDS : *heterogeneous combustion , self-propagating high-temperature synthesis, X-ray*

I. INTRODUCTION

The combustion occurring in a wide range of technogenic and natural systems (burners, rocket engines, fires, etc.) involves electromagnetic radiation in the optical, infrared, and radio-frequency spectral regions [1, 2]. This effect owes to thermal radiation and chemiluminescence of reaction products. The chemical energy released in combustion is normally, on average, no greater than 1–2 eV per product atom. Therefore, the thermal radiation spectrum is bounded from the short wavelength range by the visible spectrum: λ=400–650 nm. In hydrocarbon and hydrogen flames, an intense epithermal spectrum is found spanning into the ultraviolet region up to λ=250 nm [1]. The latter fact suggests that in combustion, nonequilibrium energy concentration on quantum states of individual system particles takes place. What the energy concentration level is and whether X-rays are possible in combustion are the questions that remain open to date. Their study is significant both for deeper fundamental understanding of combustion mechanisms and for development of new systems of direct chemical energy conversion.The systems that hold promise for generation of electromagnetic radiation in the X-ray range are heterogeneous systems that form condensed reaction products. Combustion of these systems, for example, in self-propagating high-temperature synthesis (SHS) and thermal reduction [3, 4], features high density of released energy (10^{13}–10^{15} W/m^3 and more) which is far greater than that in gas flames. Earlier, in thermal explosion of certain heterogeneous systems with condensed products, an intense ultraviolet spectrum at a wavelength λ = 200–400 nm was found [5], and rather recently, soft X-rays were detected [6].In the work, we studied X-ray radiation in wave combustion of a Ti-B system. The chemical reaction was Ti + 2B → TiB_2; the maximum adiabatic combustion temperature was 3190 K [3].

II. RESEARCH TECHNIQUES AND MATERIALS

The reaction system was a powder mixture of composition Ti + 31.5 wt. % B prepared successively as follows: 1) *mixing* of amorphous black boron (B content 98 mass %, particle size d ≤ 1μm) and nanodispersed titanium (Ti content 99.8 mass %, d = 80–100 nm) in isopropyl alcohol; 2) *deposition* of a thin layer of mixture slurry (thickness h = 70 μm) on the surface of aluminum foil (h = 13 μm) by silk screening; 3) *drying* at a temperature of 310 K. The density of the resulting mixture layer was about 0.10–0.12 kg/m^2.The study was performed using the arrangement shown in Figure 1. Combustion of the mixture was initiated by a short heat pulse from the electric spiral located at the specimen face.

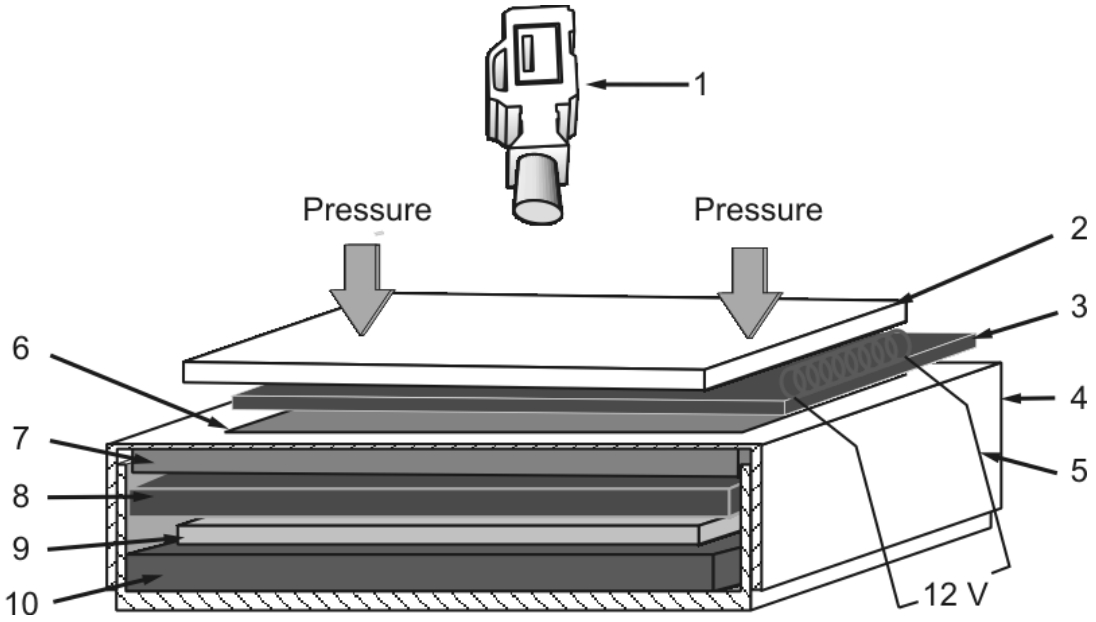

Fig. 1. Schematic of the experimental arrangement. 1 – high-speed video camera; 2 – optical glass; 3 – reaction system specimen; 4 – container; 5 – igniter spiral; 6 – radiation input window; 7 – titanium foil; 8 – black paper; 9 – X-ray film; 10 – copper plate

In the study, Kodak RAR-2497 X-ray film was placed on the surface of a heat-removing copper plate inside a lightproof steel container equipped with an X-ray input window. For protection of the X-ray film from heat, infrared and optical radiation in wave combustion, the window was covered with protective shields: titanium foil (h = 20 μm) and black lightproof paper (h = 120 μm). In individual experiments, for control of the X-ray spectral composition, a plane nickel mesh (h = 20 μm, cell size 400 × 400 μm, fiber width 140 μm) was placed directly on the surface of the photosensitive film layer. Above the container window, a reaction system specimen and a sight quartz glass were arranged and pressed against the shields. The combustion was controlled through the sight glass with a Motion Pro X3 high-speed video camera. The experiments were performed in air at normal pressure.

The experiments demonstrated that during the combustion, the protective shields preserve their integrity and are a barrier to optical and infrared radiation of the specimen. According to thermocouple measurements, the X-ray film in the reaction experiences short-term heating to less than 360 K. The heating, according to simulation of thermophysical conditions with an external electric heater, hardly affects the film photosensitivity.The X-ray intensity J was estimated from the degree of exposure of the X-ray film by photometric methods [7]. For this purpose, an image on the X-ray film (after standard cycles of chemical development and fixing) was converted to a digital form. Computer processing of a negative format of the digital image was used to measure the average brightness of the site of interest (i) providing that $i < i_n$, where i_n is the saturation brightness of exposed X-ray film. To allow for the background, the brightness of an image on unexposed X-ray film (i_o) was also determined.

Considering that the image brightness is proportional to the irradiation dose,

$$J \approx k_r (i - i_o),$$

where k_r is a parameter dependent on the exposure time, photon energy, and film properties. Within a rather small space region, it can be taken that $k_r \approx$ const.

III. RESEARCH RESULTS

According to the video filming data, the combustion front propagates in the mixture layer with an average linear velocity $V_c \approx 0.2$ m/s. In the combustion front, local reaction centers emerge stochastically (figure 2a). During the incubation period 2–5 ms, as the maximum brightness temperature is reached, the centers

produce gas dynamic bursts of condensed reaction products (scattering rate $V_F \approx 0.5$ m/s). X-ray diffraction analysis shows that the combustion end product consists mainly of TiB_2 compound.

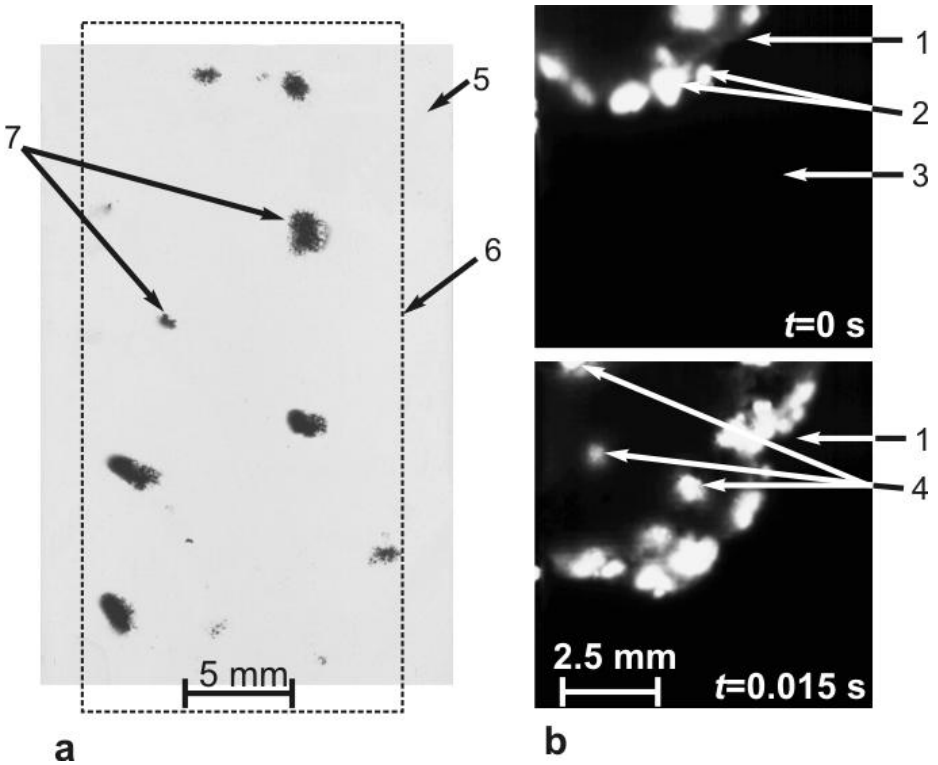

Fig. 2. X-ray film flaring during the propagation of the combustion wave in the Ti-B powder system (a) and frames of filming the process (b). 1 – combustion front; 2 – reaction center; 3 – initial mixture; 4 – reaction products; 5 – X-ray film; 6 – location of the reaction system specimen; 7 – flare spots

The reaction process results in flares of size up to 5 mm on the X-ray film (figure 2b). The latter fact points to the presence of X-rays in individual sites of the combustion wave. The flare spots are inhomogeneous (figure 3a), and this is explained by the presence of primary radiation sources of size no greater than 100–200 μm. During the combustion in open air environment, ozone smell was felt indirectly confirming the presence of ionizing radiation. The radiation of the combustion wave leaves an imprint of the absorber contour on the X-ray film (figure 3a). Photometric measurements of local brightness of the image (areas of 80 × 200 μm) near the boundaries of the nickel mesh (in the shadow zone formed by the mesh fiber, i_r; outside the shadow, i_{ro}) allows estimating the radiation transmittance:

$$\beta_r = J_r/J_{ro} \approx (i_r - i_o) / (i_{ro} - i_o),$$

where J_{ro}, J_r are the X-ray intensities before and after transmission through the nickel layer, respectively.

The study shows that the values of β_r measured at different sites of the absorber contour differ greatly from each other, and this is indicative of a broad X-ray energy spectrum. From statistical data processing of β_r over 100 measurements (figure 3b) and known absorbing properties of nickel (figure 3c) it follows that the radiation in the combustion wave contains a spectrum of X-ray quanta from 4.5 to 30 keV in which about 28 % of the flow is due to quanta of energy more than 18 keV.

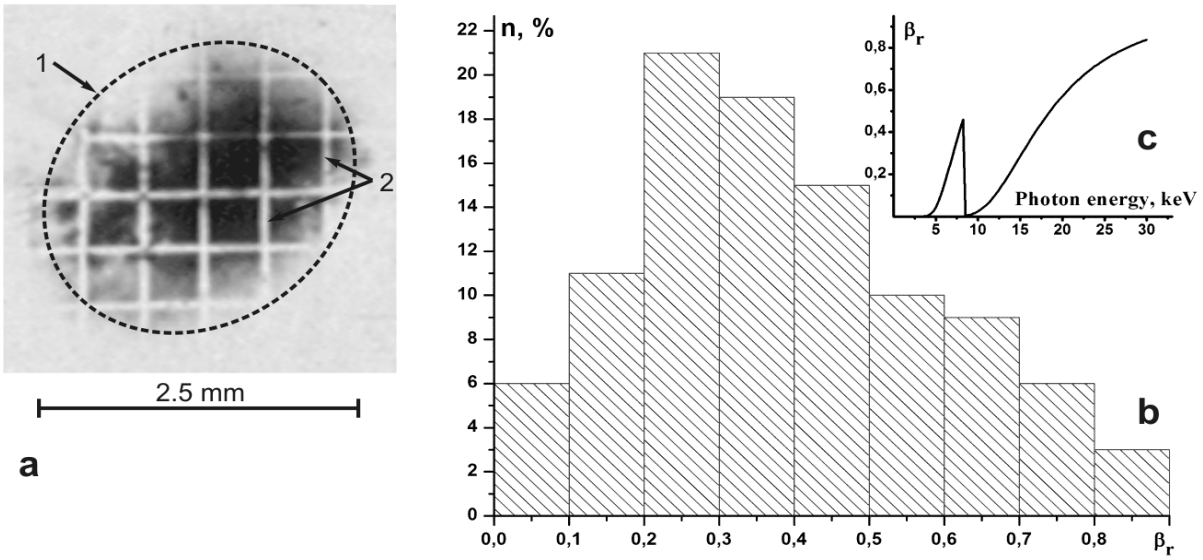

Fig. 3. Distribution of β_r (b) over local flaring sites of the exposed film (a) and dependence of β_r on quantum energy (c) (from [8]). n – relative number of sites corresponding to specified β_r; *1* – flare spot; *2* – imprint of the nickel mesh contours

IV. DISCUSSION

The generation of X-rays under our experimental conditions is yet to be strictly explained. From general consideration it is clear that production of X-ray quanta of energies tens of kiloelectron-volts requires fast electrons of appropriate energy, and acceleration of electrons requires high electric fields. It is known that in a nanosecond breakdown of various gases at increased pressures, runaway electron beams and X-rays arise [9, 10]. In natural conditions, for example, in a cloud-to-ground discharge, this is attainable [11].In a series of physical processes, the generation of high-energy electrons and quanta was observed with no external electric field as well. In 1953, V.P. Karasev *et al*. studied the electron emission on separation of eupolymer film from glass in vacuum [12]. According to the experimentally substantiated electrical theory of adhesion, an electric double layer is formed at the "film–substrate" interface. The researchers demonstrated that on fast separation of the film at low pressures, intense electron emission and acceleration of electrons to an energy of 10^3–10^4 eV takes place. Based on this effect, nanosecond X-ray bursts were realized in atmospheric pressure air in rewinding of peeling tape from reel to reel [13]. In fragmentation of a LiF single crystal, electron emission with an energy spectrum more than 100 keV was detected, and this was explained in the context of triboelectric charge separation and electron acceleration between crack walls [14]. As shown recently, splitting of ionic crystals results in an electric field of strength ~10^8 V/cm between the formed surfaces where runaway electrons can be generated even at atmospheric pressure [15]. According to [16], on short pulsed laser irradiation of synthetic opal crystals, a narrow-band flow of X-ray photons is initiated. In the authors' opinion, the effect is due to shock-induced electron acceleration in the field of electroacoustic waves of the crystal.

In view of the available data, it can be supposed that the generation of X-rays in combustion of the Ti-B system occurs by the following mechanism. The pattern of physicochemical transformations of the powder mixture layer in the combustion wave, according to the known concepts of combustion of heterogeneous systems with condensed products [3], is represented by stages of heating, capillary coalescence of melted Ti and B particles, mixing (convective, diffusion) of reagents, and chemical reaction between the components with the formation of TiB_2 crystalline compounds. The initial components and reaction products possess electronic conductivity; therefore, noticeable piezo- and triboelectrical effects in the process are hardly probable. It can be hypothesized that on microlevels of the reaction system during fast physicochemical transformations, energy of extreme density is released and, as a consequence, electrons are accelerated to energies of about several and tens of kiloelectron-volts ("hot" electrons). These electrons in their interaction with the reaction system or with the protective shields produce the X-rays observed in the experiment.

The foregoing hypothesis agrees with the well-known effect of nonequilibrium emission of electrons and positive ions from the surface of condensed particles in combustion and thermal explosion of a series of powder systems. The gas plasma formed above the surface of particles due to the emission features a high electron temperature (up to 80000 K [17] in Ti-Si systems) and a rather large fraction of hot electrons with an energy of up to 60-150 eV (Ni-Al and Mo-B systems [18]); the plasma contains atomic pairs of reaction components of first and second ionization degrees (Ti-B and Zr-B systems [5]). The nonequilibrium emission results in electric polarization of the reaction system: the surface of condensed particles accumulates a charge of one sign and the surrounding gas medium accumulates a charge of opposite sign. Reasoning from charge energy conservation in Coulomb interaction, the polarization can be estimated by the relation $E_e \approx \Delta\varphi_c \cdot e$, where e, E_e are the charge and the kinetic energy of emitted electrons, $\Delta\varphi_c$ is the maximum potential difference between the surface of condensed particles and the gas medium. We observed electric polarization in combustion of a Ti-B-Al model system in air. The system was a twist of Ti and Al wires covered with an amorphous boron layer (figure 4a). The combustion was initiated by preliminary electric current heating of the twist to a temperature of 1100÷1300 K. At a certain critical temperature, a self-accelerating exothermal reaction between the components began in one of the twist loci. Thereafter, the twist was spontaneously separated into two pieces, the current in the circuit cut off, and a self-sustained chemical reaction as a combustion wave occurred in each pieces of the twist (figures 4b, 5).

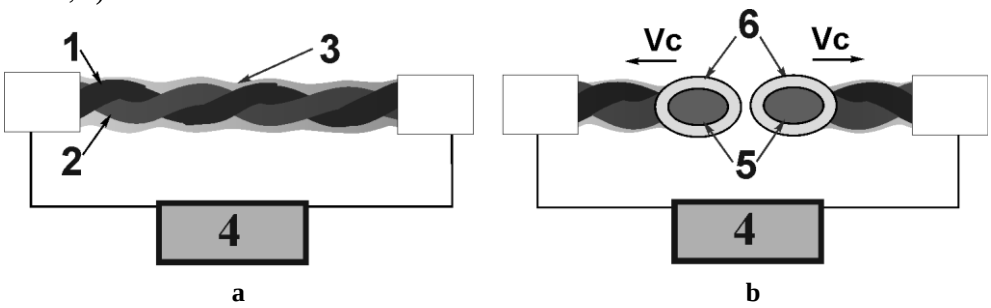

Fig. 4. Experimental arrangement for combustion of the Ti-B-Al model system. a – initial state (1, 2 – Ti and Al wires, 3 – boron layer, 4 – power supply), b – combustion process (after cutoff of the current), 5 – reaction zone, 6 – gas plasma layer.

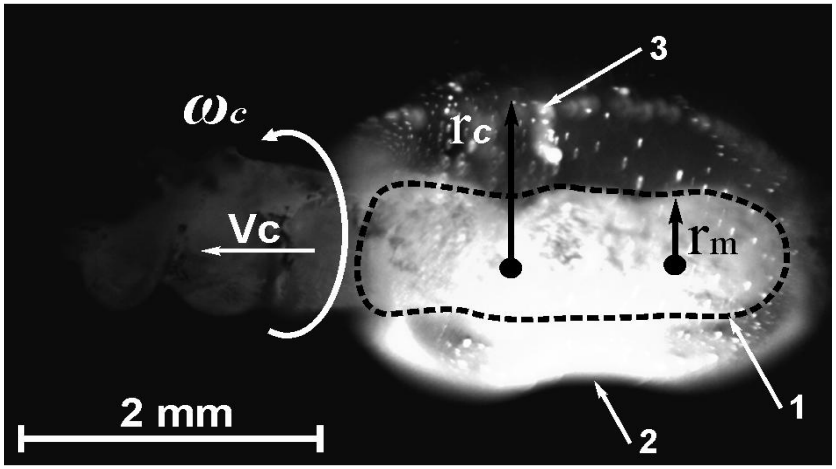

Fig. 5. Frame taken in high-speed filming of combustion of the Ti-B-Al model system. 1 – reaction zone, 2 – gas plasma, 3 – condensed particle.

According to the data of high-speed video filming, the combustion gives rise to a glowing gas plasma zone containing a suspension of fine condensed particles above the surface of the reaction zone. The suspension presumably consists of amorphous boron particles or reaction products which are ejected from the twist surface by local gas-dynamic flows. During the reaction, the plasma (with the suspension) rotates about the reaction centripetal force responsible for rotational motion is likely to be Coulomb interaction of unlike electric charges

distributed on the twist surface and in the plasma zone.The observed polarization of the system can be estimated in the following model approximation. Let the reaction zone of the twist in combustion emit an electron flow. In this case, a positive charge is accumulated on the surface of the twist reaction zone and a negative charge is accumulated in the plasma zone. The electric potentials of suspension particles and surrounding plasma are taken equal. The latter means equal average charge densities in the particles and in the surrounding plasma.

In the approximation of cylindrical system symmetry, the balance condition for Coulomb and centrifugal forces in circular motion of individual suspension particles can be represented in the form:

$$E(r_c)\cdot\delta(r_c)=\rho_c\cdot\omega_c^2\cdot r_c, \qquad (1)$$

where r_c, ω_c, ρ_c are the radius of circular motion, the angular velocity, and the density of a particle, respectively; $E(r_c)$, $\delta(r_c)$ are the electric field strength and the charge volume density in the plasma where the particle moves.

According to the laws of electrostatics, in a cylindrical coordinate system with the circular axis r originating at the twist center, the potential difference $\Delta\varphi_c$ between the surface of the twist reaction zone and the particle is determined by the equation:

$$\Delta\varphi_c=\int_{r_o}^{r_c}E(r)dr\,, \qquad (2)$$

where $E(r)=\left(\tau-\int_{r_o}^{r}2\pi\cdot r\cdot\delta(r)dr\right)/\left(2\pi\cdot\varepsilon\cdot\varepsilon_o\cdot r\right)$ is the distribution of the electric field strength in the plasma zone, r_0 is the coordinate of the twist surface; ε, ε_0 are the dielectric permittivity of the medium and the electric constant; $\delta(r)$ is the charge volume density distribution function in the plasma zone; τ is the linear charge density on the surface of the twist reaction zone.

In view of (2), condition (1) can be represented as follows;

$$\Delta\varphi_c=\omega_c\int_{r_o}^{r_c}E(r)dr\cdot\sqrt{\rho_c\cdot r_c/\left(\delta(r_c)\cdot E(r_c)\right)} \qquad (3)$$

From the law of charge conservation in the system follows the relation:

$$\tau=\int_{r_o}^{r_*}2\pi\, r\delta(r)dr, \qquad (4)$$

where r_* is the coordinate of the outer boundary of the plasma zone, $\delta(r > r_*) = 0$.

It would appear reasonable that $\delta(r)$ is a continuous function with a maximum in the range $r_0 \le r \le r_*$. Let $\delta(r)$ be represented in the following linear approximation:

$$\delta(r)=\begin{cases}a(r-r_o); r_o\le r\le r_m\\ b(r_*-r); r_m\le r\le r_*\end{cases}, \qquad (5)$$

where a, b are constants and r_m – is the coordinate of the maximum of $\delta(r)$.

In view of relation (4), function (5) takes the form:

$$\delta(r)=\left\{\begin{array}{l}\dfrac{\tau\cdot(r-r_o)}{\pi(x_1+\alpha\cdot x_2)};\ r_o\le r\le r_m\\ \dfrac{\alpha\cdot\tau\cdot(r_*-r)}{\pi\cdot(x_1+\alpha\cdot x_2)};\ r_m\le r\le r_*\end{array}\right\}, \qquad (6)$$

where $\alpha=\frac{(r_m-r_o)}{(r_*-r_m)}$; $x_1=\frac{2}{3}\cdot r_m^3-r_o\cdot r_m^2+\frac{r_o^3}{3}$; $x_2=\frac{2}{3}\cdot r_m^3-r_*\cdot r_m^2+\frac{r_*^3}{3}$

Joint analysis of equations (3, 6) with variation of the parameters r_m, r_* shows that under the experimental conditions ($\omega \approx 170$ rad/s; $r_o \approx 5\cdot10^{-4}$ m; $r_c \approx 1.4\cdot10^{-3}$ m; $\rho_c \approx (1\div3)\cdot10^3$ kg/m^3; $\varepsilon \approx 1$), the minimum value of $\Delta\varphi_c$ sufficient for circular motion of a suspension particle is $\Delta\varphi_c(\min) \approx (2.1\div3.6)\cdot10^3$ V. Representation of $\delta(r)$ by other approximating functions (for example, by power functions of different orders) little changes the estimate of $\Delta\varphi_c(\min)$.The obtained data suggest that the spontaneous electric polarization of the reaction system in combustion reaches a kilovolt level. This is possible if the reaction zone of the wave emits a flow of hot electrons with energies higher than several kiloelectron-volts.The proposed hypothesis does not exhaust other

explanations of the X-ray generation. Therefore, further research is required to develop a strict model of the effect.

V. CONCLUSION

The obtained data suggest that the spontaneous electric polarization of the reaction system in combustion reaches a kilovolt level. This is possible if the reaction zone of the wave emits a flow of hot electrons with energies higher than several kiloelectron-volts.The proposed hypothesis does not exhaust other explanations of the X-ray generation. Therefore, further research is required to develop a strict model of the effect.

ACKNOWLEDGEMENTS

The publication of this paper was supported by the grant from the Russian Science Foundation, project №14-29-00052.

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