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Acoustic instabilities in hydrogen-air mixtures in the closed reactor at the central spark initiation

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ABSTRACT

It was shown that inhomogeneities detected in light emission that arise after contact of a flame front with the walls of cylindrical reactor can be correlated with occurrence of acoustic waves by the example of combustion of hydrogen-air mixtures containing 30 and 15 % of hydrogen. It is revealed that flame velocities in stoichiometric hydrogen-air mixtures at central spark initiation do not depend on the material of inner reactor surface but on its shape.

KEYWORDS

Hydrogen;
Flame;
Propagation;
Surface;
Acoustic;
High-speed cinematography.

INTRODUCTION

As is known a combustion process is a source of acoustic vibrations. The sound generated by combustion plays an important role when ensuring stable functioning of engines, turbines, etc.[1] because, in particular, intensive pressure fluctuations lead to undesirable loads of walls of a combustion device. On the other hand, it is known[2] that in fluctuating flows there is an essential acceleration of various heat mass exchange processes (mixing, evaporation, heat transfer from gas to walls, heat exchange at flow interaction to heated bodies etc.), agglomeration of particles and coagulation of aerosols; in addition vibrational combustion slows down NO_x[3] content in combustion products.

Acoustic fluctuations in combustion are caused by instabilities arising at flame front propagation[4]. These instabilities are shown in various forms in combustion processes and can be classified as follows. Thermal diffusion instability is observed in the flames in which velocities of heat transfer and diffusion differ markedly, i.e. if $Le \neq 1$ (Le is Lewis number = D/χ, where D is diffusivity of the component determining the velocity of combustion, χ is heat diffusivity[5, 6]. Hydrodynamic instability is determined with a difference in densities of the burned-down and initial gas, which is associated with thermal expansion of reaction products[4]; this instability in certain conditions can lead to occurrence of cellular flames[7]. Thermoacoustic instability is related to the feedback between a non-stationary mode

of thermal emission and acoustic fluctuations in the reactor[4]. Rayleigh[8] gave the criterion of occurrence of this instability. It reads that acoustic fluctuations are sustained if the maximum quantity of heat is transferred to fluctuating gas at the time of its maximum compression. In[9] this hypothesis is formulated as following: if phase shift between fluctuations of pressure and fluctuations of velocity of heat application is less than $\pi/2$ on absolute value, fluctuations are sustained; if phase shift ranges from $\pi/2$ to π, the fluctuations are suppressed.

Pressure fluctuations at combustion in closed vessels accompanied with sound are usially associated with thermoacoustic instability[10], however up to now the conditions of occurrence of acoustic vibrations are not quite clear[10]. In[11] acoustic fluctuations in rich pentane - air mixes were detected, however in rich benzene - air mixes acoustic fluctuations were not observed. In[12] acoustic vibrations in combustion of hydrogen-air mix at $\phi = 0.42$ (15 % H_2) were detected however in combustion of hydrogen - air mix ($\phi = 0.60$; 20 % H_2) acoustic vibrations were missing (ϕ is the coefficient of excess of fuel). The possibility of spontaneous ignition in the course of flame propagation in a bomb of constant volume was considered in[13]. High speed of change in thermal emission at such spontaneous ignition or at flame acceleration[5] can induce pressure oscillations and acoustic fluctuations. The experiments performed in[10] showed that at ignition of rich isooctane-air mixes and lean hydrogen air mixes strong pressure fluctuations occur. The origination of acoustic fluctuations in[10] is related to development of hydrodynamic instability at flame propagation. However, pressure oscillations in combustion of stoichiometric hydrogen- air mix was not observed in[10] though it is known that combustion of the mix in cylindrical reactors is accompanied by a characteristic sound[12].

The non-uniform wave equation for acoustic pressure with a thermal source is used for modeling of the thermo acoustic phenomena[3, 14, 15]:

$$\frac{1}{c_0^2}\frac{D^2P(x,y,z,t)}{Dt^2} - \nabla^2 P(x,y,z,t) = \frac{\partial}{\partial t}\left[\frac{\gamma-1}{c_0^2}Q_t\right] \quad (1)$$

where c_o is the speed of sound, P is pressure, γ – the relation of thermal capacities at constant pressure and constant volume, *dQ/dt* is the velocity of thermal emission.

If we assume a feedback mechanism representing *dQ/dt* as a linear function of pressure[14], it is possible to show[15] that the equation (1) represents an acoustic oscillator which is excited or is damped out depending on Rayleigh's criterion.

In the work the results of experiments on features of combustion of hydrogen - air mixes in a cylindrical bomb of constant volume with central ignition are presented. The special attention is paid to influence of material and a shape of an internal surface on the velocity of flame propagation and conditions of occurrence of acoustic fluctuations.

EXPERIMENTAL

Experiments were performed with stoichiometric hydrogen - air mixes at atmospheric pressure and initial temperature T_0 = 298 K in horizontally located stainless steel cylindrical reactor of 15 cm in length and 13 cm in diameter. The reactor was supplied with an optical quartz window on one of end faces. Electrodes of spark ignition the distance between which made up 0,5 mm were placed in the center of the reactor. To change the material and the shape of internal surface of the reactor 0.1 mm thick Ti foil or 0.1 mm thick Ta foil covering all internal lateral surface of the reactor and 12×6 cm 0.3 mm thick Pt foil were placed in the reactor in various experiments. In a number of experiments 0.1 mm thick Ti foil 12 cm in length covering internal lateral surface of the reactor with three longitudinal flanges 1cm high, and located at distance of 4 cm from each other was positioned in the reactor. We will specify that placing of Ti foil in the reactor can be considered as treatment of internal reactor surface with titanium dioxide TiO_2 because titanium is always covered with a thin oxide layer. In a number of experiments speed filming was carried out via neutral NS-1 filters for reduction of light intensity. With the same purpose in some experiments, the optical glass was covered with a thin layer of talcum powder.

Experiments were performed in the following sequence. The pumped reactor was first filled with

CCl_4 (if needed), then with hydrogen to nesessary pressure, then air was added up to the atmospheric pressure so that H_2 content in mix made up 30% of 15%. The gas was maintained for 15 min in the reactor for completeness of mixing and then spark initiation was carried out. Speed filming of ignition dynamics and flame front (FF) propagation was carried out through the optical window by means of a color high-speed digital camera Casio Exilim F1 Pro (frames frequency 60–1200 s^{-1}). The video file was stored in computer memory and its time-lapse processing was performed. The pressure change in the course of combustion was recorded by means of a piezoelectric gage synchronized with the discharge.

Carbon tetracloride (CCl_4) was added to some mixes for visualization of hydrogen flame. Notice that the additive is practically inert and does not show noticeable inhibiting action on hydrogen combustion[12]. Before each experiment, the reactor was pumped out with the use of a forvacuum pump 2NVR-5D. Pressure in the reactor was also controlled with a model vacuum gage. H_2, carbon tetrachloride were chemically pure, the purity of foils made up: Ti – 99,9%, Ta – 99,99%, Pt – 99,99%.

RESULTS AND DISCUSSION

It was shown that the features of luminescence at afterburning of stoichiometric H_2 -air mix in conditions of central spark initiation don't depend on the material of internal reactor surface (stainless steel, TiO_2, Ta, Pt), but depend on its shape. Normal velocities of hydrogen flames also do not depend on surface material. Experiments showed also that acoustic fluctuations caused by combustion are excited after achievement of pressure maximum, however in combustion of 15% H_2 -air mix the fluctuations are excited later, than in stoichiometric 30% H_2 -air mix.

Results of high speed filming of process of FF propagation in 30% H_2 -air mix are presented in Figure 1 at shots frequency 600 frames/s.

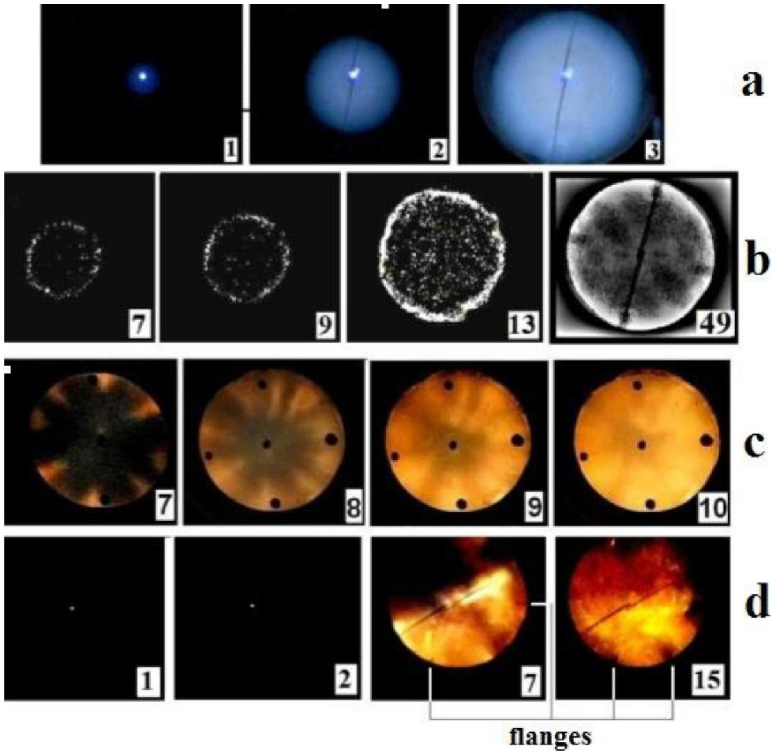

Figure 1 : High-speed filming of process of spherical FF propagation. The figure on a shot corresponds to a shot number during the shooting, T_0 = 298 K, E_0 = 1.5 J; a) – $(H_2+air)_{stoich}$ + 2% CCl_4, 600 frames/s; b) – 14.5% H_2 + air + 3% CCl_4, 300 frames/s; c) – $(H_2+air)_{stoich}$, the NS-1 filter, an optical window is treated with talcum powder, surface – stainless steel, 600 frames/s; d) – $(H_2+air)_{stoich}$, 2 NS-1 filters, Ti foil with flanges (noted on shots 7 and 15) is located in the lower part of the reactor, 600 frames/s

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The frames of FF propagation in 30% H_2 -air mix containing 5% CCl_4 are presented in Figure 1a. As is seen in Figure 1a FF has a spherical shape without perturbations. Results of filming of process of FF propagation in 15% H_2 + air + 3% CCl_4 mix are shown in Figure 1b. It is seen in the figure that FF consists of small cells; in the course of FF propagation long-wave perturbations occur. Oscillograms of pressure change at ignition of gas mixtures and 15% H_2 + air + 3% CCl_4 are presented in Figure 2. As is seen in Figure 2 acoustic fluctuations in 30% H_2 + air are excited after achievement of a pressure maximum, and in 15% H_2 + air these fluctuations are excited later than in the stoichiometric mix. However, in[10] acoustic fluctuations in the spherical reactor were observed only in lean hydrogen-air mixes and before achievement of a pressure maximum, but in the mixes close to stoichiometric one acoustic fluctuations were not observed at all (Figure 3[10]).

As is seen in Figure 1b, the perturbations of spherical FF caused by thermal diffusion instability[5, 6] in combustion of the lean mix in the cylindrical reactor, develop until a contact of FF with lateral walls of the reactor. However, these perturbations do not lead to initiation of acoustic fluctuations. However, in[10], in which the diameter of a spherical bomb made up 38,4 cm (see Figure 3[10], curve ϕ=0,4) acoustic fluctuations were observed at FF propagation. Seemingly, amplitude of perturbations at the stage of spherical FF propagation in this work isn't sufficient for initiation of acoustic fluctuations due to smaller diameter of the reactor (13 cm) as compared to the reactor used in[10]. In the cylindrical reactor acoustic instabilities, as is seen in Figure 1 a,b and Figure 2, are excited after the time of contact of FF with lateral walls of the reactor. The data obtained are the argument in favor of existence of large-scale effect at occurrence of this type of instability.

We have revealed[7] cellular flames originating in diluted n-pentane-air mixes after the contact of FF with lateral walls of a cylindrical reactor caused by occurrence of gasdynamic instability. It means that pressure fluctuations (Figure 2) can be caused by the gasdynamic instability arising after FF contact with lateral walls[7] because both gasdynamic instability and acoustic one are experimentally observed in the same time interval. Verification of this assumption demands further research.

The results of speed video filming of combustion of stoichiometric H_2 + air mix over time interval, corresponding to occurrence and development of pressure oscillations for various materials of reactor surface (stainless steel, TiO_2 with flanges Figure 1; Pt and Ta, Figure 3) are presented in Figure 1 c, d and Figure 3 a, b. As is seen in the figures after FF contact with reactor walls the luminescence of FF occurs not uniformly in the vertical section of the reactor; the symmetric picture of afterburning can thus be observed. It is seen in these figures that in case of smooth internal reactor surface experimentally observed character of afterburning (shot 8 Figure 1 c, shot 6 Figure 3a, shot 8 Figure 3 b) doesn't depend on the material of reactor surface.

Non-uniform currents of gas obtained by introduction of Ti foil with three longitudinal flanges into the reactor (see Experimental) were provided in the

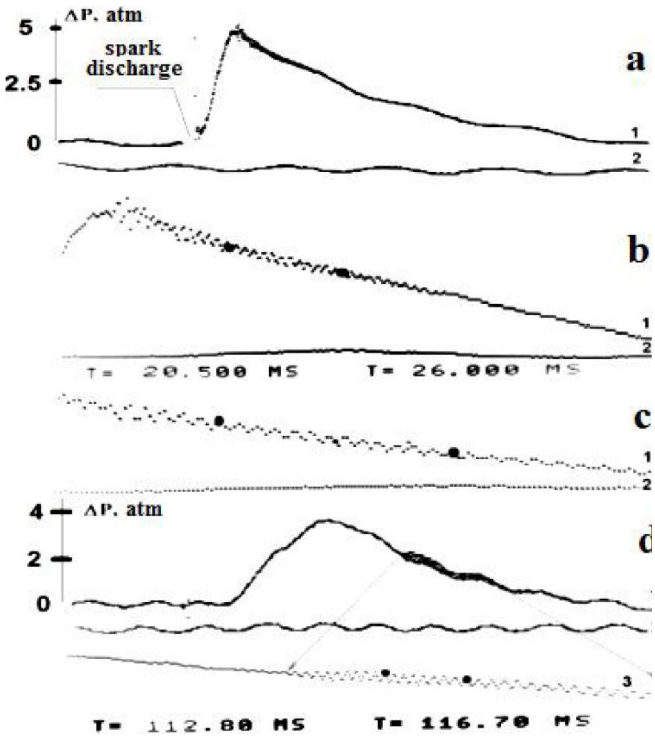

Figure 2 : Oscillograms of pressure change in combustion ignition of gas mixtures; 1 – signal at the initiated ignition; 2 – comparison signal, large black points – time interval; 3 – the stretched time interval; a,b,c – $(H_2+air)_{stoich}$, various time intervals; d – 15% H_2+air + 3% CCl_4

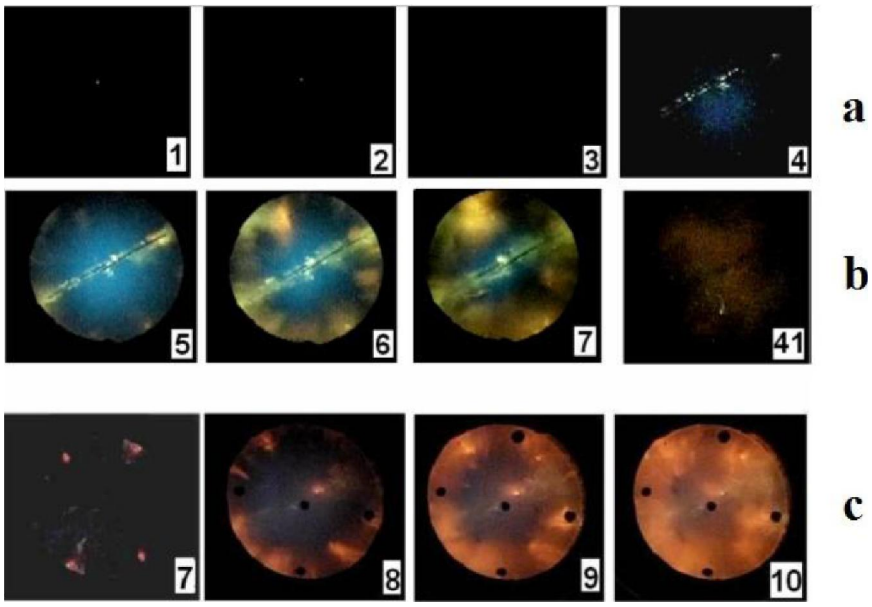

Figure 3 : High-speed filming of the process of spherical FF propagation. The figure on a shot corresponds to shot number during the shooting, T_0 = 298 K, E_0 = 1.5 J, 600 frames/s; a – (H_2+air) $_{stoich}$, 1 NS-1 filter, Pt foil is located in the lower part of the reactor; b – (H_2+air) $_{stoich}$, 2 NS-1 filters, Tà foil is located in the lower part of the reactor; c – (H_2+air) $_{stoich}$, 1 NS-1 filter, optical window is treated with talcum powder, Ti foil is located in the lower part of the reactor

following series of experiments. It was established (Figure 1, TiO_2 foil with flanges) that in case of rough internal surface of the reactor the most intensive afterburning of gas mixture occurs over the area of non-uniform currents of gas namely over the region of flanges.

It is of interest to compare the frequencies of the oscillations observed directly on pressure oscillograms (Figure 2) and frequencies of normal modes of vibrations of the cylinder filled with gas. For determination of normal modes of the cylindrical reactor, it is enough to put the right term of equation (1) equal to zero. Assuming P (x, y, z, t) = exp (iwt) v (x, y, z) we get the well-known wave equation[15] :

$$\nabla^2 v(x,y,z) = \frac{w^2}{c_0^2} v(x,y,z), \qquad (2)$$

where w – the frequency of normal mode of vibration.

The solution of the problem was carried out by means of the finite elements method. For calculation of normal modes of vibration of the cylinder filled with inert gas with motionless walls and height equal to diameter was performed by means of the software package FlexPDE 6.08 (A Flexible Solution System for Partial Differential equations, 1996-2008 PDE Solutions inc.[16]) in which this problem appears as an example.

As combustion takes place in conditions close to adiabatic ones, one can estimate the temperature in the reactor at given timepoint from a ratio $\frac{\Delta P}{P} = \frac{\Delta T}{T}$ according to pressure oscillogram. For instance, for the timepoint t = 21 ms in Figure 2 ΔP = 4, P = 1 we get ΔT = 273•4 ≈ 1000 °C, for this temperature C_0≈ 000 m/s[17], and for the timepoint t = 27 ms ΔP = 3.5, P = 1, we get ΔT = 273•3.5 ≈ 900 °C; for this temperature c_0 ≈ 900 m/s[17]. In Figure 4 the shot 7 of video filming shown in Figure 1c, the shot 41 from Figure 3b, and the calculated normal modes of vibrations of air in the cylinder: the 36th mode and 4th mode are presented. As is seen in Figure 4 the calculated normal modes correlate qualitatively well with experimentally observed features of afterburning for stoichiometric H_2 - air mix.

Taking into account the length of the cylinder (15 cm) we get for the 36th vibrational mode w = 1.11 kHz; for C_0 = 900 m/s (T ≈ 900 °C) we obtain for the fourth vibrational mode w = 0.7 kHz. From pressure oscillograms in Figure 2b over time interval 20.5 - 26.0 ms and in Figure 2d over time interval 112.5 - 116.7 ms we obtain oscillation frequency ~

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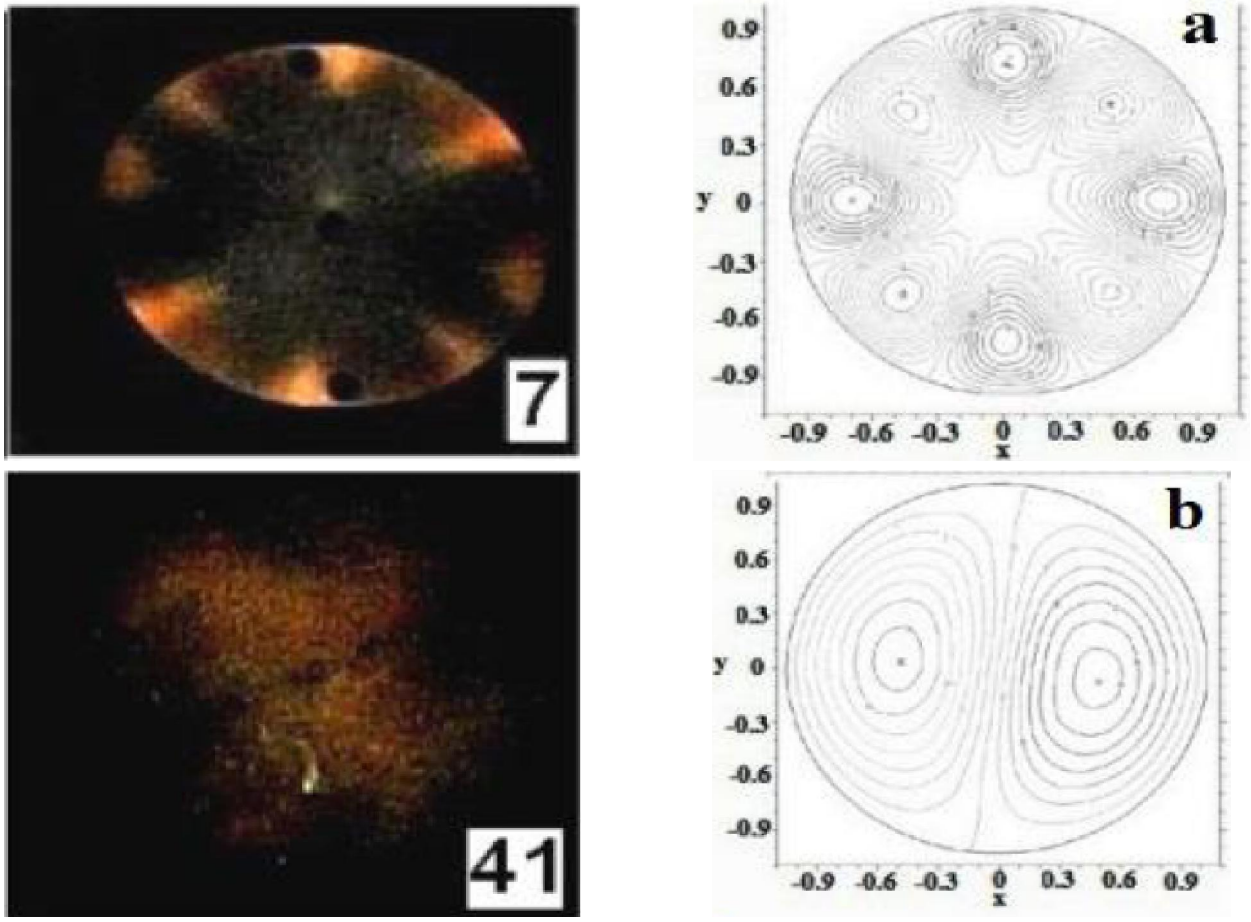

Figure 4 : Frames of video filming from Figure 3c and Figure 3b, and calculated normal modes of vibrations of air in the cylinder, a – the 36th mode, b– the 4th mode

1.9 kHz which correlates well with the value w = 1.11 kHz, calculated from the equation (2) on the basis of the algorithm[16]. As is also seen in Figure 2, at the end of the process of afterburning, on the portions of pressure oscillograms corresponding to the reactor cooling-down, the oscillation frequency considerably decreases and reaches 1 kHz on the oscillogram 3 in Figure 2. This value also is in agreement with the calculation for lower vibration mode w = 0.7 kHz.

Notice that the comparison performed above has only illustrative character and does not allow interpreting more complicated mode structure of acoustic vibrations in combustion of 15% H_2 -air mix (Figure 1, shot 49).

CONCLUSIONS

It has been established that velocities of flame propagation in stoichiometric hydrogen-air mix under central spark initiation don't depend on the material of internal reactor surface (stainless steel, TiO_2, Ta, Pt), but depend on the shape of the internal surface. By the example of combustion of hydrogen-air mixes (30 and 15% of H_2) it is shown that the inhomogeneities of luminescence at afterburning can be associated with occurrence of acoustic waves. It is revealed that acoustic fluctuations in combustion of hydrogen-air mixes at atmospheric pressure are excited in the cylindrical reactor after achievement of pressure maximum. In 15% H_2 -air mix these fluctuations are excited later than in stoichiometric mix unlike the spherical reactor of the larger size, where acoustic fluctuations were observed only in combustion of lean hydrogen-air mixes and before achievement of the pressure maximum.

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