EXPLOSION HAZARD OF HYDROGEN-AIR MIXTURES IN THE LARGE VOLUMES

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ABSTRACT
The report deals with the investigation of non-stationary combustion of hydrogen-air mixtures extremely relevant to the issues of safety. Considered are the conditions of its formation and development in the tubes, in the conic element and in the spherical 12-m diameter chamber. The report shows that at the formation of non-stationary combustion in the conic element, in its top the pressure can develop exceeding 1000 atmospheres. It is also shown that in large closed volumes non-stationary combustion can develop from a small energy source, in contrast to detonation for whose stimulation in large volumes significant power influences are required. Simultaneously, in the volume a pressure can be formed by far exceeding the Chapman-Jouguet pressure in the front of stationary detonation.

INTRODUCTION

The present report treats the problems of safety while working with hydrogen. If ignition of hydrogen happens, the major safety goal is minimization of losses and destruction. Work at facilities where hydrogen is used must be regulated by corresponding normative documents, standards and codes. The recommendations stated in these documents have to base on the study of kinetics and gas dynamics of hydrogen-air mixtures combustion.

The nature and size of destruction are determined by the formed mixture combustion regime. The stationary regimes of combustion, deflagration and detonation have found their detailed description in specialized literature. A relatively slow deflagration leads to fires and burnout of facilities or their fragments. In the front of supersonic combustion, i.e. detonation, a 12-15-fold pressure increase takes place, which brings about much more serious destructions of facilities.

The forecasts of emergencies mostly focus on parameters characterizing conditions in the front of stationary detonation as being of primary danger, and these parameters define the requirements regulating the use of hydrogen. Investigations of processes of combustion in large volumes are of great importance for the safety problems. Investigations on combustion of hydrogen-air mixtures in large volumes have been made in a number of studies, e.g. [1–4]. For the most part these studies have been dealt with investigations of possibility of detonation onset under various initiation conditions. However, occurrence of large ruinous pressure is not necessarily related to deflagration to detonation transition. Here we mean, first of all, the regimes of non-stationary combustion that involve the development of high pressures, which in a number of cases tenfold exceed the pressure in the front of stationary detonation. Non-stationary combustion of gas mixtures are secondary centers of combustion and explosion, which were formed as a result of the mixture compression by shock waves generated by primary flame front.

The regimes of non-stationary combustion have been little studied, especially in large volumes representing the most interest for safety. Ways and means of fighting with these regimes have neither been sufficiently developed, nor considered in normative documents on hydrogen safety. However, resolving these questions is extremely significant, especially with the view of accelerated development of hydrogen economy.

Initiation of non-stationary combustion is connected with formation of streams of reaction-capable gases, distribution in them of disturbances, final amplitude waves and shock waves in premises where there are industrial technological lines, power producing units, and large-scale experimental installations. Centers of ignition can result from the formation of stagnant zones and cumulation of waves from sources of various nature and intensity.

The given paper presents the results of the research on initiation of explosions and development of non-stationary combustion and detonation caused by shock waves in hydrogen containing mixtures. Most attention is given to features of the processes accompanying flowing of moderate intensity shock waves in cavities, interaction of waves with surfaces and their propagation in reaction-capable medium.

DEVELOPMENT OF COMBUSTION AND DETONATION BEHIND WAVES IN TUBES

The process of ignition initiated by shock waves is most vivid in the experiments made in shock tubes. Informative and convincing are the experiments visualizing the process of propagating shock waves, formation and development of reaction zone and movement of combustion waves [5-8].

Ignition of hydrogen mixture behind a shock wave normally reflected from the shock tube end is shown in Fig.1. It presents a schlieren photograph of propagation of an incident shock wave (1), reflected shock wave (2), structure of medium behind these waves (3), (4), the centers of ignition (5) and own luminescence of combustion waves (6) extending from ignition centers. Ignition occurred at mixture pressures up to 7 atm. Research were conducted for hydrogen-oxygen mixture.

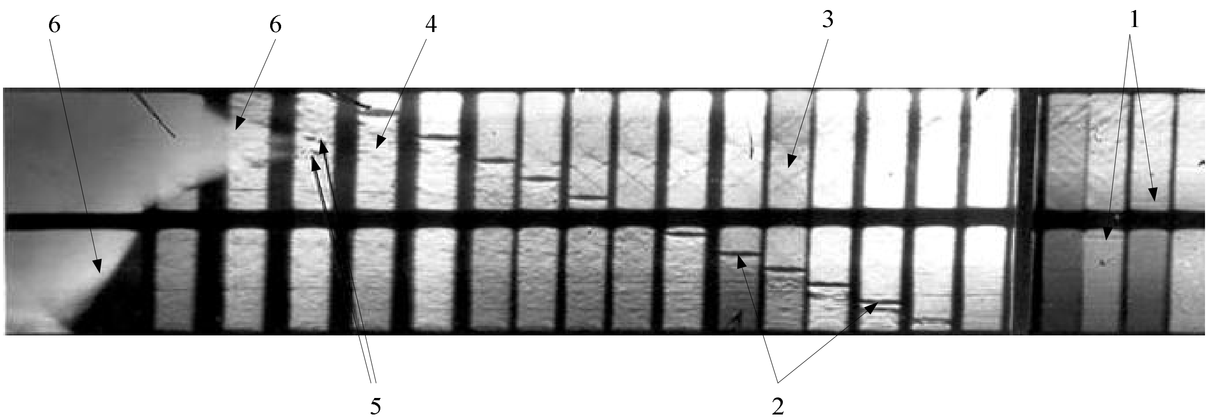


Figure 1. A schlieren photograph of shock waves propagation in the shock tube and combustion development behind the wave reflected from the tube end (tube 4.5 m length with square section 4×4 cm^2).

Basing on the results of the experiments shown in Fig. 1, by the velocity of an incident wave one can calculate the parameters behind the shock wave reflected from the tube end. Quantitative characteristics of the ignition conditions and combustion processes development were received. Values of pressure and velocity of front were measured and calculated. The temperature and density of gas behind the incident and reflected shock waves were estimated, as well as the time of luminescence initiation counted from the moment of reflection, velocity of ignition front movement at the initial stage and velocity of combustion front movement at a later stage.

The treatment of these data shows that the process has a stochastic nature; with the identical initial conditions within the limits of experiment accuracy, qualitative distinctions in process development are registered. Significant time dispersion in delay of the ignition centers formation in relation to the moment of reflection of the wave from the tube end testifies to essential differences in the character of the process development. For a wave with Mach number M = 2.15 (±2%) the dispersion is within the range of 50–300 μs. The nature of the process development after the ignition centers formation is also ambiguous. A sharp (explosive) propagation of reaction is possible: the luminescence front is spread with a velocity exceeding the velocity of stationary detonation right after initiation. It can be a "two-stage" development of combustion: directly from the centers, the luminescence slowly propagates with a practically constant velocity, which then sharply increases (in the picture – the break of luminescence front trajectory), changing to supersonic one.

If the ignition centers are formed close to the tube end, practically right after a wave reflection, the front of intensive luminescence extends into the fresh mixture, merging with the reflected shock wave, while there is no detectable movement of a clearly confined front in the tube end direction and there is only weak luminescence in volume visible. In the regimes characterized by ignition centers initiation at a big distance from the tube end, a clear luminescence boundary can be observed extending both after the reflected shock wave and to the tube end. Herewith the initial data for the regimes are identical, and the formation of the above-described different regimes has a random character.

Here is an estimation of pressure that is formed in similar processes in hydrogen-air mixtures for normal initial conditions. The wave M=3 gives a 10.2-fold increase in pressure, temperature T_2 is 775K; at such temperature the detonation pressure falls 2.65-fold; therefore, behind the wave with M = 3, the pressure of stationary detonation of hydrogen-air mixture will make 60 atm behind the incident wave and 150 atm behind a reflected wave.

FORMATION AND DEVELOPMENT OF COMBUSTION PROCESSES IN CONIC CAVITY

It is obvious that in the volumes with similar linear dimensions on all directions combustion wave propagation is three-dimensional, which creates challenges for both experimental and numerical research. In such volumes, much more complicated become the gas dynamic processes, which play an essential role in the formation of combustion centers, explosions, development and propagation of supersonic combustion waves. The research of non-stationary processes of combustion and explosions in large closed volumes or volumes of the non-conventional form started only recently. In the conditions admitting the ignition centers initiation, the combustion formed by a primary weak source leads to initiation of disturbances that, intensifying at cumulation, can result in explosion initiation. In cumulation zones, high pressure and temperature areas are formed, in which, with slowing down, there may be sufficient-time conditions created for initiation of secondary ignition and explosion centers. These secondary phenomena can cause explosion hazard in natural volumes and premises; in many cases they are characterized by the parameters, first of all pressure, higher than those of the fixed detonation waves.

In the Joint Institute for High Temperatures, Russian Academy of Science, they carry out research in focusing of the blast waves propagating in the reaction-capable mixture filling large volumes.

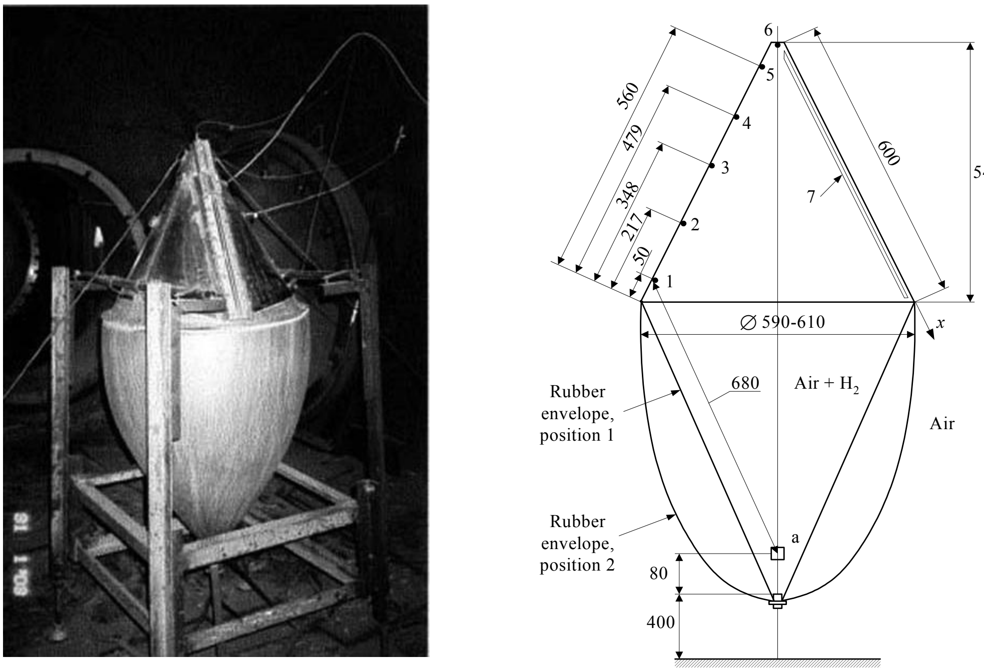
The results of the research on occurrence of combustion centers and explosions with cumulation of blast waves have been partially published by the authors in [9, 10]. In [9, 10] there are stated the results of research on combustion and explosion development of hydrogen-air mixtures filling a conic volume when entered by a blast wave from an initiating source of small energy. In the conditions of our experiments (Fig.2, a and b) at initiation of the process by explosion of a RDX charge whose mass is less than m = 1.5 g the combustion front from the primary center (an initiation source) does not reach the cone top. With the charge mass $1<m<1.5$ g for stechiometric mixtures in the cone top there can be a combustion center, and herewith the pressure rises to 20-30 atm. In hydrogen impoverished mixtures with the mass of an initiating charge under 1g, there is no combustion observed in the cone top at the moment of maximum pressure registration, but the pressure rises to 6-8 atm. The processes taking place near the cone top are characterized by the interaction with the cone of pressure waves and the streams arising with the explosion of an initiating charge and propagation of a primary flame in the subcone volume. Wave propagation in reaction-capable and reacting mixture with the initiating charge less than 1.5 g leads to the fact that with their cumulation the pressure in the top becomes 5-12-fold higher than with the cumulation of the waves from an explosion of a similar energy charge in a neutral gas – air. It is observed with the implementation of such regimes when a primary wave of combustion does not reach the top, and no secondary ignition centers appear in the subcone area.

Improving experimental base and expanding the range of defining parameters of physical experiment make it possible to receive some new information for realizing of tendencies of process development.

The experimental volume (whose scheme and general view are shown in Fig. 2) is made of a 52-liter metal cone and a thin rubber film "bag" attached to it, which is schematically presented in two variants: position 1 and position 2. In position 1, the general capacity of the experimental volume is 113 liters; in position 2, it is 190 liters at the normal initial conditions (the energy of the mixture with volume of 190 liters is equivalent to 130 g TNT).

It can be estimated that, at the ignition of the mixture by RDX charges with the mass no more than 3.5 g, the contribution of initiation energy made 1-4% of the total heat of the hydrogen-air mixture that filled the experimental volume. Hence, it is not the energy contribution of the initiating source with the RDX mass increase from 1 g to 3.5 g that brought about a significant change of the pressure on the cone surfaces and in the subcone areas. It was a change in gas dynamic structures of the streams caused by an initial explosion and a subsequent combustion. The intensity of the wave, which was formed at the primary explosion of the initiating charge and the volume of combustible gas adjoining to it, differs from the wave initiated in the inert environment not only by the regime of its initiation, but also by the conditions of propagation. The waves in reaction-capable mixture are considerably more intensive, as there is additional charging of waves by the energy emitted in the reaction zone and generated by the combustion front disturbances.

In the experiments, the pressure sensors *1-6* (Fig. 2) registered the information of the pressure disturbances arrival time and the value of this pressure. By means high-speed photography, along a window-slot on a cone generatrix, luminescence propagation is registered, which allows to determine an average value of visible velocity of the combustion zone movement.



a b

Figure 2. General view (a) and scheme of experimental conic volume (b).
1-6 – pressure sensors, 7 – window-slot for high-speed photography
(all dimensions are in millimetres)

Check experiments on cumulation of blast waves in the neutral environment (air) are made at exploding 1.5 g and 3.5 g of RDX. The pressure on the first sensor located near the cone edge is 1.5-2 atm. In the subtop area, the pressure sharply increases to make 7-9 atm on sensor 5 and 18-20 atm on sensor *6*. An average velocity of disturbance propagation from the moment of RDX explosion in point "a" before the registration of a signal by sensor *6* makes 450 km/s ("average Mach number" 1.3).

If we admit that with the same degree of cumulation of reaction-capable gas in the cone top stationary detonation of Chapman-Jouguet will take place, the pressure increase will be 170 atm. Measured on sensor 6, the maximum pressure values in the reaction-capable mixture exceed this value by 5-8 times.

It is worth mentioning the fact of a non-monotonous change in maximum pressure along the cone generatrix. Practically in all experiments, the maximum registered pressure varies from sensor to sensor: on the first transducer, its value is not minimal, while on the second and third ones it usually drops. It is obvious that in the experiments non-stationary gas dynamic processes take place and non-stationary combustion wave propagation is realized. Figure 3 shows typical oscillograms of signals of pressure sensors.

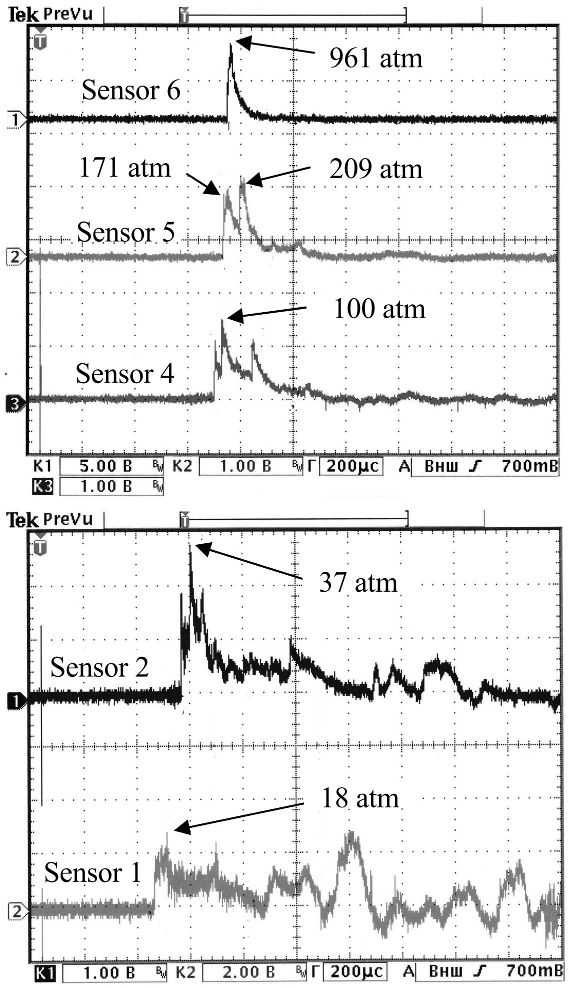


Figure 3. Oscillograms of signals of pressure sensors (cone volume, 30% H_2, 3.5 g RDX).

Table 1 demonstrates examples of pressure change along the cone generatrix for mixtures of different composition. There are several values of pressure and registration time of this pressure corresponding to consecutive pressure peaks on oscillograms in some table cells. For the experiments reflected in lines 1-3 (for the stechiometric composition mixture) the energy of RDX charge with mass of 3.5 g is supercritical, while for the data in line 4 (15% H_2) and line 5 (9.3% H_2) this energy is significantly below critical.

Table 2 shows the results of measurements executed for the stechiometric hydrogen-air mixture at the initiation of the process by the explosion of a RDX charge with the mass of 3.5 g. Figure 4 presents a photosweep characteristic of these regimes of the mixture combustion process made through the window along the cone generatrix.

Table 1. Typical examples of pressure change along the cone generatrix.

RDX mass, g	Concentration H_2, %	Pressure value *P*, atm, & registration time *t*, μs*					
		Sensor, No.					
		1	2	3	4	5	6
3.5	29	41.4 (346)	18.2 (402)	62.1 (446) 37.7 (450)	37.7 (498)	169.5 (535) 109.7 (569)	830 (551)
3.5	29	–	61 (380) 34.5 (382)	44.2 (428)	49.0 (474)	36.2 (524) 70.3 (553)	767 (530)
3.5	29	56.7 (331)	35.9 (391)	53.5 (432) 46.9 (610)	39.6 (478) 46.7 (573)	40.3 (525) 76.6 (555) 82.9 (579)	1028 (534)
3.5	15	6.8 (728)	2.83 (944)	2.85 (1020) 14,02 (1380) 12.9 (1680)	3.01 (200) 9.17 (1344) 10.79 (1584)	8.17 (1388) 18,37 (1480)	44.28 (1408) 38.25 (1464)
3	9.3	3.35 (1040, 1050)	4.88 (1260,1375)	3.06 (1250, 1595) 2.87 (1830) 3.71 (2060)	3.02 (1490, 1845) 3.72 (2018) 3.57 (2295)	9.2 (1450,1990) 7.9 (2210)	48.7 (1970, 2040)

**The time of pressure registration is in the brackets. Note that for data shown in 1-4 lines time rise of signal is less than 5 μs; for data shown in line 5 the first value corresponds to the advent of a signal at the sensor, the second one –- to achievement of the maximal pressure.*

Table 2. The pressure registered by sensors 1 and 6 for the stechiometric hydrogen-air mixture and the realization time (process initiation by explosion of 3.5 g of RDX).

Parameter	Experiment #							
	1	2	3	4	5	6	7	8
Time t_1 of wave arrival to sensor *1*, μs	327	310	331	331.5	320	335	–	346
Pressure P_1, registered by sensor *1*, atm	56.7	42.2	56.7	40.0	47.8	51.2	–	41.4
Time t_6 of wave arrival to sensor *6*, μs	530	519	534	531	539	537	530	551
Pressure P_6, registered by sensor *6*, atm	625	515	1028	810	582	978	766	830

Related to the experiments discussed, there is a significant disperse of experimental data, which is generally characteristic of combustion processes in quickly burning mixtures. In [11] an opinion is quoted concerning the features of hydrogen- and acetylene- containing mixtures, which says that in studying combustion processes there appears "velocity of flame propagation (in the tubes) essentially changing from experiment to experiment despite all attempts to maintain identical experiment conditions".

Apparently, non-stationary explosion processes and combustion processes at their formation stage have even more stochastic character.

While discussing safe use of hydrogen and developing regulatory documents on hydrogen safety, it is necessary to take into account that the received pressure values implemented in hydrogen-air mixtures with normal initial parameters were extremely high.

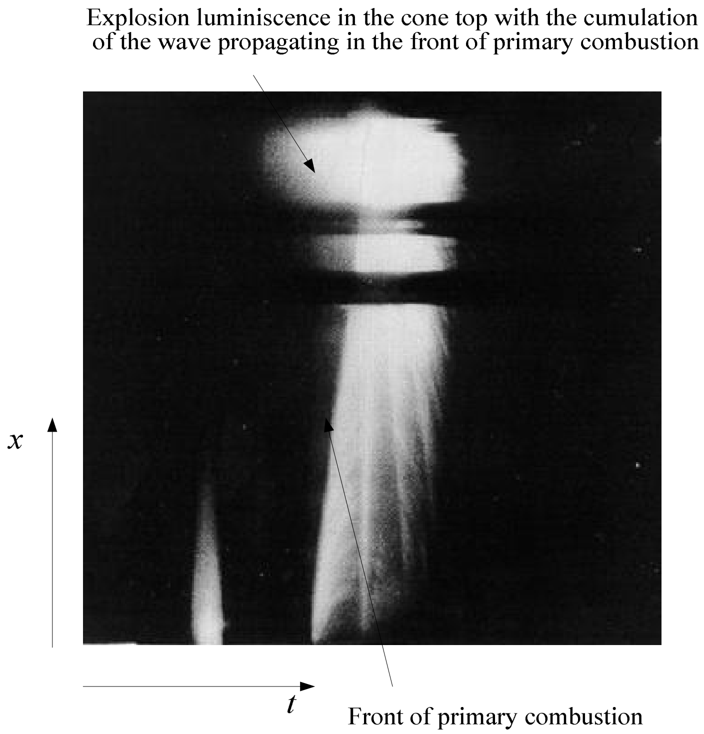


Figure 4. Luminescence propagation of a in the focusing zone of the cone

EXPERIMENT IN A SPHERICAL CHAMBER OF LARGE VOLUME

An experiment performed in a spherical chamber with the volume of 910 м^3 (Fig. 5) revealed a crucial influence of gas dynamic processes on the formation of extreme situations of explosion in gas inflammable mixtures. The chamber case having 12 m in diameter is designed for the explosion in neutral gas 1000 kg TNT. The chamber is made of 10 cm thick armored steel and has been tested for static pressure of 150 atm.

Figure 5. General view of the explosive chamber

The experimental volume is schematically presented in Fig. 6 showing the arrangement of ionizing sensors i1 – i4 and crusher pressure sensors kI – kIV. Fig. 6 demonstrates the arrangement of the constructions having periodic structure and simulating a possible option of blocking up the volume in natural conditions. These constructions were wooden and fastened on steel tubes 60 mm in diameter. The chamber interior before the experiment is shown in Fig. 7.

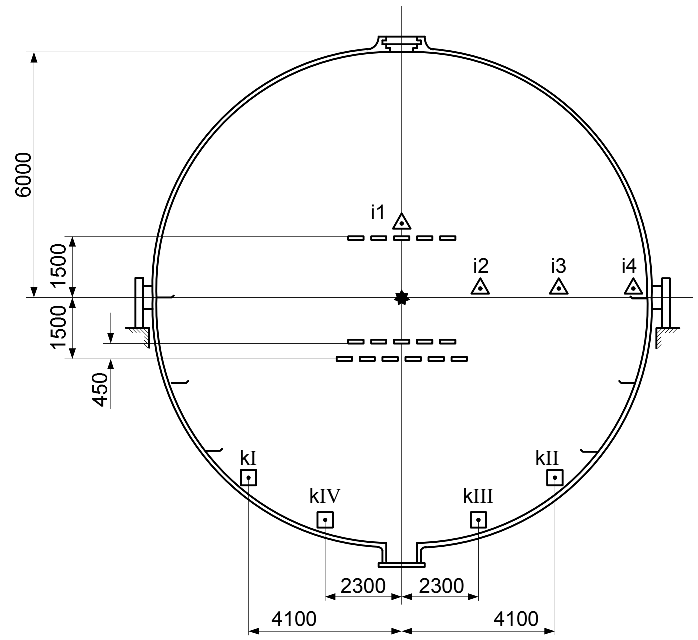


Figure 6. Scheme of sensor arrangement in the explosive chamber (all dimensions are in millimetres)

Figure 7. Chamber interior before the experiment

The chamber checked for air tightness was filled with hydrogen-air mixture. The mixture pressure in the chamber was 1.4 atm, and the mixture had stechiometric composition, containing 29% (vol.) of hydrogen. However, this concentration determined by a volumetric ratio of constituents was averaged. Measuring hydrogen contents in the mixture, which was carried out repeatedly in the course of 100 hours during which the mixture was maintained, showed that in the bottom part of the chamber, where tests for analysis were taken, the hydrogen concentration steadily kept the value of 25.4%. Measurements of composition of mixtures under consideration were carried out by gas analyzer GT-201 (Gas Tech, Inc., USA).The mixture must have been stratified, and in the top part of the chamber the hydrogen concentration could have reached 32.6%.

The initiation of the reaction in the mixture was performed by burning a copper fine wire and the discharged energy was 6–8 J. After the mixture initiation, an explosion took place in the chamber, a strong vibration of the chamber case was observed, and depressurization happened accompanied by a powerful emission of combustion products. On an internal surface of the chamber the pressure of 190 atm was registered.

Table 3 shows ionizing sensor location, and time lapse between flame front arrival to these sensors and the ignition as well as the results of estimation of average and maximum flame front velocity based on these data. In this table S is the distance between sensor and the point of ignition, t_i is time lapse between the advent of signal of ionizing sensor and the moment of ignition, $U(0\text{-i})_{av}$ is average velocity of flame front propagation from the initiation point to the location of i-th sensor, $U(\text{i-i+1})_{av}$ is average velocity of flame front propagation between sensors, and $U\text{i}_{max}$ is maximal calculated value of velocity in the vicinity of i-th sensor. One can see from the table that velocity of front propagation changes non monotonically, sharply increases nearly sensor No.2 and significantly decreases (more than sevenhold) on approaching the chamber wall.

Table 3. Ionizing sensor locations, time lapse between flame front arrival to sensors, and average and maximum flame front velocity.

Sensor number	S, m	t_i, ms	$U(0\text{-i})_{av.}$	$U(\text{i-i+1})_{av.}$	$U\text{i}_{max}$
			m/s		
1	2.0	56.5	35.4		70.8
				160	
2	4.0	69.0	57.9		249.5
				143	
3	5.9	83.0	72.3		34.3
4	1.7	47.0	36.1		72.0

A considerable acceleration of flame must be accompanied by its turbulization and formation of shock (instead of transsonic) waves, which noticeably change the parameters of the environment before the flame. Due to a higher velocity, these waves repeatedly interact with the wall. With the transsonic velocity, in 56 ms the wave will cover the distance from the center to the wall of the chamber and back (12 m) two times, and when Mach of the wave is equal 2 the number of interactions is doubled. Disturbances in the medium and its heating not only cause the change of flame propagation regime, but also create conditions for initiation of ignition centers and explosion before primary flame front, which is similar to what was observed in subtop area of the conic cavity and the tube.

Random gas dynamic structure of the environment leads to formation of asymmetrically located – in relation to the center of the chamber – secondary centers of ignition and, hence, asymmetric influence on the chamber of explosions developing from these centers, which can serve as the cause of above mentioned construction vibrations.

Significant strengthening of shock waves is possible as a result of their interaction with combustion zone. The development of processes of interaction between the combustion front and a shock wave in detonation tubes was considered in works [12, 13]. A result sample of registration of such interaction is shown in Fig. 8, borrowed from [12]. In [12] considered was a shock wave passing through combustion front of hydrogen mixture, which was formed as a result of flame propagation from an initiation source located in the middle of the tube to its ends. Fig. 9 illustrates the dependence of the factor of shock waves amplification on the initial pressure of stechiometric hydrogen mixture when they are passing through the flame.

Based on the work results [12], rough estimates of wave amplification in the conditions of our spherical chamber can be made.

At initiation of a mixture in the center of the chamber, there are weak shock waves formed which, being reflected from the wall, three times interact with the flame front and amplify due to reaction intensification. Having accepted amplification as a result of a single interaction act equal 1.3, we shall receive a wave the pressure behind which increases by 1.96 times, M=1.36. At the subsequent stages of the process development, by the time moment of 78 ms, 5 ms before the arrival of the flame front at the wall, the wave can amplify up to M=2.8. The pressure behind such a wave increases by 6.05 times, the temperature – by 1.97 times compared to the condition before the wave. The wave with such parameters is reflected from the wall before a wave of primary combustion reaches the wall; secondary combustion is initiated, turning into an explosion similar to that observed in the shock tube (see Fig. 1).

Conditionality of quantitative values of the given estimation is obvious, as we are discussing a flat wave and the figures received for a specific regime in the tube, but qualitatively the considered script is possible.

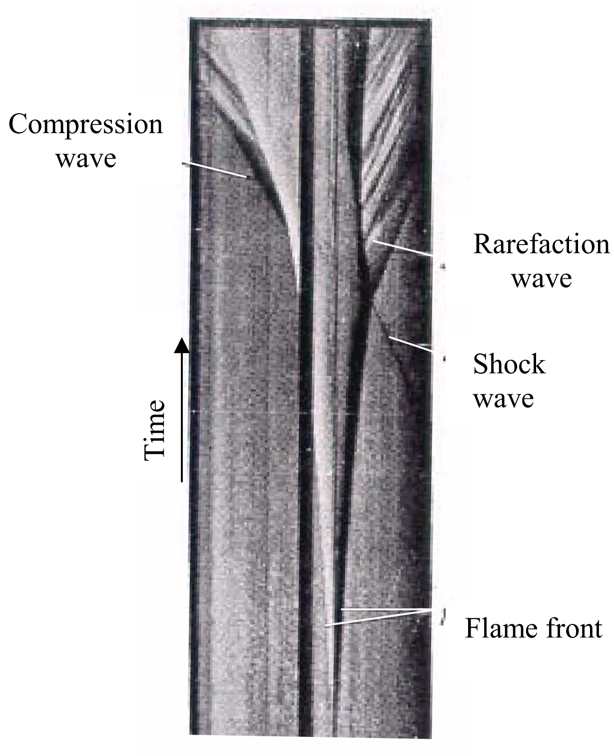


Figure 8. Interaction of shock wave with the flame front

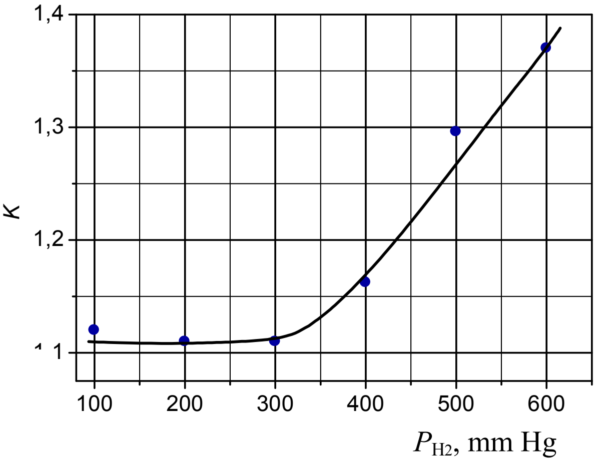


Figure 9. Influence of pressure of stechiometric hydrogen mixture P_{H2} on factor of shock waves amplification at interaction with the flame front

CONCLUSION

There was a study carried out of the processes of formation and development in the various form and geometry volumes of non-stationary combustion of hydrogen-air mixtures that have much importance for the issue of hydrogen safety.

There were considered conditions of non-stationary combustion formed in the tubes, in the conic element and in the spherical chamber having 12 m in diameter.

It was shown that non-stationary combustion regimes were most dangerous and significant in terms of their power effect on construction elements. Instability of non-stationary combustion front results in forming disturbances, waves and streams before the front. In closed and cumulating volumes wave intensification creates secondary combustion centers – explosions whose parameters exceed the values predicted by the Chapman-Jouguet conditions for stationary detonation (with normal initial conditions approximately fivefold).

It is demonstrated that in large closed volumes non-stationary combustion can develop from a source of small energy, in contrast to detonation for whose stimulation significant power effect is required. Simultaneously, in the volume there can be a pressure by far exceeding the Chapman-Jouguet pressure in the front of stationary detonation.

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