The formation of ignition centers before the front of spherical flame in hydrogen–air mixtures under intense initiation

V A Petukhov, N P Bublik, P A Gusev, L D Gutkin and O I Solntsev
Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia
E-mail: petukhov@ihed.ras.ru

Abstract. Studied is the influence of volume of hydrogen–air mixture, its composition and energy of its initiation on the formation of ignition centers before the front of spherical flame. During the experiments the mixture was placed into thin rubber envelopes of spherical shape having the initial volume of 7 and 40 m³. The mixture was initiated in the center of the reaction volume by the energy which was several times less than the critical energy of direct initiation of detonation. With increasing the initial volume of the mixture with the identical composition, registered is a decrease of the initiation energy at which the ignition centers before the front of spherical flame are formed.

1. Introduction
Propagation of the spherical flames of gas mixtures has a significant place in the gas dynamics of combustion [1]. However, the evolution of the spherical flames in hydrogen–air mixtures has not been adequately studied. One of the reasons is the necessity to carry out experiments in large volumes (dozens of cubic meters) as opposed to the mixtures in which oxygen is the oxidizer [2–4]. Having at our disposal a spherical explosive chamber with inner diameter 12 m, designed for a blast up to 1000 kg TNT, enables us to study spherical flames of the hydrogen–air mixtures.

In the few pieces of research of spherical flames of the hydrogen–air mixtures, the two limiting situations of mixture initiation were mainly studied, namely, a weak initiation by the energy up to 1 J and by the energy at which a direct initiation of detonation occurs [5]. Investigating the influence of initiation energy of intermediate values on the evolution of spherical flames in hydrogen–air mixtures revealed the formation of ignition centers before the primary front of the flame when the nearly-stoichiometric hydrogen–air mixture was initiated by the energy equal to 15% of the critical energy of direct detonation initiation [6]. Previously the formation of ignition centers was observed while compressing the hydrogen–air mixture in one or another way (impact, adiabatic) [7, 8].

In this paper, we study the influence of some factors, namely, the composition and the volume of the hydrogen–air mixture and the initiation energy, on the formation of ignition centers before the front of the spherical flames in hydrogen–air mixtures.
2. Experimental conditions

Investigations were carried out on the facility whose scheme is shown on figure 1. A combustible mixture was in a thin rubber envelope placed in the explosion chamber 13Ya3 (figure 2). We used sounding balloons as the rubber envelopes which, when filled with the gas mixture, became near-spherical. Hydrogen and air were fed into the envelope in the quantities needed to obtain the needed mixture composition. We measured the amount of the gas entering the reaction volume by means of the PROMSS 80F 08 and PROWIRL 72F 15 high-precision mass flow meters. While the hydrogen and air were being fed into the reaction volume, the mixture was blended by a fan; after the end of feeding the mixture was blended for 40 minutes more. Then, we turned off the fan and the mixture was being aged for some 30 min to damp the gas movement inside the reaction volume and to level the composition further.

We investigated hydrogen–air mixtures with the initial volume equal to 7 and 40 m$^3$ and pressure of 10$^5$ Pa. The mixture was ignited in the center of reaction volume. The initiation energy was equal to 2.3, 4.6, and 15.5 kJ. This was achieved by explosion of 0.4, 0.8, and 2.7 g spherical PETN charges, respectively. While the mixture was burning, the envelope was expanding until the rupture when the flame front reached it. As this took place, the difference between the pressure inside and outside the envelope due to its elasticity did not exceed 100 Pa, which had been proved by a special experiment.

Because of a large volume of the explosion chamber (equal to 910 m$^3$), during the combustion of the gas mixture, which had the initial volume of 40 m$^3$, the pressure inside the chamber by the time of the formation of ignition centers increased maximum by 15%, or (no more than) by 1.5 $\times$ 10$^4$ Pa. This increase in pressure cannot cause auto-ignition.

We registered the burning process by means of a high-speed video camera through the illuminator in the upper hatch of the explosion chamber with rates of 300 and 600 frames per second (figure 1). We controlled the filling of the rubber envelope by gas mixture using a video camera located inside the explosion chamber.
### Table 1. Experimental parameters.

| Experiment No. | $H_2$ (vol %) | Initial volume ($m^3$) | Initiation energy (kJ) | Effect $^a$ |
|----------------|---------------|------------------------|------------------------|-------------|
| 1              | 30.0          | 40.0                   | 2.3                    | +           |
| 2              | 30.0          | 40.0                   | 2.3                    | +           |
| 3              | 30.0          | 40.0                   | 0.001                  | –           |
| 4              | 20.0          | 40.0                   | 2.3                    | –           |
| 5              | 25.0          | 40.0                   | 2.3                    | –           |
| 6              | 30.5          | 7.0                    | 2.3                    | –           |
| 7              | 30.5          | 7.0                    | 4.6                    | +           |
| 8              | 28.0          | 7.0                    | 4.6                    | –           |
| 9              | 22.0          | 7.0                    | 15.5                   | –           |
| 10             | 24.0          | 7.0                    | 15.5                   | +           |
| 11             | 26.0          | 7.1                    | 15.5                   | +           |
| 12             | 28.0          | 7.0                    | 15.5                   | +           |

$^a$ “+”—ignition centers occurred; “−”—ignition centers did not occur.

### 3. Experimental results

The composition of hydrogen–air mixtures under study, the initial volume and the initiation energy are shown in table 1. Many of the mixtures studied had a composition close to that of a stoichiometric one, i.e. containing 29.6 vol % of hydrogen.

Figure 3a shows the frames obtained by means of the high-speed video camera (600 frames per second) in experiment 1. The mixture was initiated by the energy of 2.3 kJ, which equals to 15% of the critical energy of the direct detonation initiation of the stoichiometric mixture [9]. The frames clearly show the formation of ignition centers growing in size.

We repeated this experiment with the same initial parameters, but with higher requirements to homogeneity of the mixture. To do this, hydrogen and air were fed inside the reaction volume through a special mixer. Then, after the end of feeding, the mixture inside the rubber envelope was blended by a fan for ~1.5 hours. However, the centers were still occurring (experiment 2). For comparison, figure 3b shows the frames obtained for the hydrogen–air mixture having the same initial composition and volume, which was initiated by the energy 1 J (experiment 3). In this case, no ignition centers occurred before the flame front.

The products of explosion sources could not be the cause of ignition center formation because the impact of the explosion product on the gas mixture ceases at the distance of 10–12 charge radii [10]. In the present investigation, the radius of the spherical 0.4 g PETN charge equals to 4 mm and the impact by the explosive products ceases at the distance of 40–48 mm, which is more than one order less than the distance at which ignition centers occur.

In experiments 4 and 5, when the initial volume and initiation energy were the same as in experiments 1 and 2 and hydrogen content in the mixtures was lower, no ignition centers occurred.

Further experiments were carried out in the volume of 7 m$^3$.

In the mixture with hydrogen content of 30.5 vol %, initiated by the energy of 2.3 kJ (experiment 6), ignition centers did not occur. In the mixture with the same initial volume and hydrogen content (7 m$^3$ and 30.5 vol %) ignition centers occurred when the initiation energy increased up to 4.6 kJ (experiment 7, figure 4). In leaner mixtures, initiated by the energy of 4.6 kJ, ignition centers did not occur (experiment 8).
Figure 3. Video frames of the 30 vol % hydrogen–air mixture: (a)—initiation energy 2300 J, frame rate 600 f/s (experiment 1); (b)—initiation energy 1 J, frame rate 300 f/s (experiment 3). The arrows indicate the ignition centers.
**Figure 4.** Video frames of the hydrogen–air mixture burning: 30.5 vol % H$_2$, initial volume 7 m$^3$, frame rate 300 f/s, initiation energy 4.6 kJ.

**Figure 5.** Video frames of the hydrogen–air mixture burning: 24 vol % H$_2$, initial volume 7 m$^3$, frame rate 300 f/s, initiation energy 15.5 kJ.
At the same time, at initiating of hydrogen–air mixtures by the energy of 15.5 kJ in the volume of 7 m$^3$ ignition centers occurred in the mixtures with 24 vol % H$_2$ (figure 5, experiment 10) and richer (experiments 11, 12) right up to the beginning of detonation, which with such initiation energy comes when the hydrogen content is equal to 29 vol %. The energy of 15.5 kJ equals to 25% of the critical energy of the direct initiation of detonation for the mixture containing 24 vol % of hydrogen [9].

4. Discussion
The formation of the ignition centers in hydrogen–air mixtures is connected with the increase in its temperature to the auto-ignition temperature.

A number of factors affects this increase: the composition of the mixture, an explosive wave from the source of initiation, the waves generated by the expanding turbulent front of the flame, the waves reflected from the rubber envelope, and there is a small contribution of compressing the mixture by an expanding flame (for the experiments with the initial volume of 40 m$^3$ the contribution is equal to 15 K).

The influence of the explosive wave can be seen comparing the results of experiments 6 and 7 with the same initial volume and composition of the mixture: at initiation the mixture by the energy of 4.6 kJ the ignition centers occurred, but at 2.3 kJ they did not occur.

The temperature of auto-ignition depends on the content of hydrogen in the hydrogen–air mixture.

The influence of the mixture composition can be seen from comparing experiments 1, 2 with experiments 4, 5. The initiation energy for all these experiments was equal to 2.3 kJ.

In experiments 1 and 2 the mixture was close to stoichiometric one having lower auto-ignition temperature in comparison with the leaner mixtures [11]. The formation of ignition centers was detected in experiments 1 and 2, and was not in experiments 4 and 5. The same conclusion can be drawn from the comparison of experiments 7 and 8, as well as experiments 9, 10, 11 and 12.

The heating of the mixture by the waves generated by the expanding turbulent front of the flame and the waves reflected from the rubber envelope depended on the time the mixture was exposed to it. The larger the mixture volume, the longer the exposure.

Therefore, the ignition centers occur when the mixture with the initial volume of 40 m$^3$ and hydrogen content of 30 vol % is initiated by the energy of 2.3 kJ (experiments 1 and 2), and the centers do not occur when the mixture with hydrogen content of 30.5 vol % is initiated by the same energy of 2.3 kJ, but in the initial volume equal to 7 m$^3$ (experiment 6).

Propagation of spherical flames in hydrogen–air mixtures is closely connected with their burning in large volumes, which is of great interest for safety in atomic and hydrogen power engineering.

Intensification of burning leads to more devastating consequences, because devastating effect of a shock wave at burning of gas is determined by the time of the conversion of the initial mixture to combustion products [10]. The extreme regimes of intensification are detonation and even a more destructive regime–non-stationary combustion [12]. The formation of ignition centers before the front of spherical flame leads to more intense burning of the hydrogen–air mixture.

As noted above, with the increase in the mixture volume the initiation energy decreases at which the ignition centers occur. Because of this, in large industrial areas (for instance, internal volume of the containment of a nuclear power plant reaches 70,000$^3$) the formation of ignition centers in case of emergency inflammation is possible at lower initiation.

5. Conclusion
Given are the results of the study of influence of various factors (the composition and the volume of the hydrogen–air mixture and the energy of its initiation) on the formation of ignition centers.
Their formation leads to more intense combustion of large amounts of hydrogen–air mixtures, which may cause an increase in the loads on the structural elements in case of emergency inflammation of hydrogen in industrial facilities.

Acknowledgments

This work is supported by the Russian Science Foundation (grant No. 14-50-00124).

References

[1] Zel’ dovich Ya B, Barenblatt G I, Librovich V B and Mahviladze G M 1980 Matematicheskaja Teorija Gorenija i Vzryva (Moscow: Nauka)
[2] Voevodsky V V and Soloukhin R I 1965 Proc. 10th Symp. (Int.) Combustion (Pittsburg: Combust. Inst.) pp 279–283
[3] Oppenheim A K 1985 Phil. Trans. R. Soc. A 315 471–508
[4] Vermeer D J, Meyer J W and Oppenheim A K 1972 Combust. Flame 18 327–336
[5] Makeev V I, Gostincev Yu A, Stroganov V V, Bohon Yu A, Chernushkin Yu N and Kulikov V V 1983 Fiz. Goreniya Vzryva 5 16–18
[6] Petukhov V A, Bublik N P, Gusev P A, Gutkin L D and Solntsev O I 2016 High Temp. 54 92–98
[7] Gel’fand B E, Khomik S V, Medvedev S P, Polenov A N and Bartenev A M and Groenig H 1998 Dokl. Phys. Chem. 359 97–101
[8] Gel’fand B E, Bartenev A M, Medvedev S P, Polenov A N and Khomik S V 2001 Zh. Vses. Khim. Obsch. im. D I Mendeleeva 45 5–14
[9] Gel’fand B E, Popov O E and Chaivanov B B 2008 Vodorod: Parametry Goreniya i Vzryva (Moscow: FIZMATLIT)
[10] Orlenko L P (ed) 2002 Fizika Vzryva vol 1 (Moscow: FIZMATLIT)
[11] Gamburg D Yu and Dubovkin N F (eds) 1989 Vodorod. Svojstva, Poluchenie, Hranenie, Transportirovanie, Primenenie (Moscow: FIZMATLIT)
[12] Petukhov V A, Naboko I M and Fortov V E 2009 Int. J. Hydrogen Energy 34 5924–5931