Buoyant flames in near the lower flammability limit hydrogen–air mixtures

A I Gavrikov, V V Golub, V V Volodin, A Yu Mikushkin and A V Danilin

1 National Research Center “Kurchatov Institute”, Kurchatov Square 1, Moscow 123182, Russia
2 Joint Institute for High Temperatures of the Russian Academy of Sciences, Izhorskaya 13 Bldg 2, Moscow 125412, Russia
3 Bauman Moscow State Technical University, 2nd Baumanskaya Street 5, Moscow 105005, Russia
4 Nuclear Safety Institute of the Russia Academy of Sciences, Bolshaya Tulskaya Street 52, Moscow 115191, Russia

E-mail: gavrikovandrey@yandex.ru

Abstract. The paper presents the results of large-scale experimental and simulation study of the propagation of premixed near the lower flammability limit hydrogen–air spherical flame. Experiments were carried out in a cylindrical volume of 4.5 m$^3$ covered with thin polyethylene film. The effect of buoyancy and stability of the flame ball in the unconfined volume are studied experimentally and numerically. Numerical analysis of the problem shows that the flame ball becomes unstable after some period of stable propagation that qualitatively corresponds with experimental results. The effect of chemistry in a flame ball propagation process is studied numerically. A comparison of experimental results and numerical simulations are presented. A comparison of numerical simulations with and without chemistry is shown. The subject and results of the study are of critical importance for the industrial explosion safety and hydrogen safety of nuclear power plants.

1. Introduction

One of the first papers describing the effect of buoyancy in a flame of depleted hydrogen and air was published in 1948 [1]. It was shown in the paper that the structure of rising hydrogen flames can be made visible by adding chromyl chloride or nickel carbonyl. These additives disintegrate in the flame front with the deposition of solid, thermally glowing substances, so that the flame contours are clearly recognizable. The density, the thermal conductivity, etc of the gas mixture was varied by adding CO$_2$, SO$_2$, SiCu and SF$_6$ as well as by changing the pressure. As previously suspected, in the paper was shown that the rising H$_2$ flames do not have a uniform burning surface, but a structure of the flame front determined by the diffusion and flow processes, which, depending on the concentration ratios, goes through all stages from complete quenching to decay into many individual flame kernels. The number of these kernels was related in elementary terms to their diameter. It was also shown that the upward movement of the individual flame kernel is a turbulent process. In recent decades, a number of experimental researches on buoyant flames were done. They include not only hydrogen–air flames, but also hydrogen–methane–air and methane–air mixtures, also [2–4].
At the same time, it is well known that if hydrogen content is less than 10 vol %, the main mechanism of the flame propagation changes from thermal diffusion to deficient species diffusion into the reaction zone. Ya.B. Zel’dovich was the first who theoretically predicted the possibility of the lean diffusive spherical flames formation in the near the lower flammability limit hydrogen–air mixtures that were called flame balls [5]. Later, some theoretical research on flame balls was done [6, 7]. The smooth flame front in gas mixtures is unstable. The acceleration of the gas flame front at the initial stage after initiation is caused by the curvature of its front [8] and the hydrodynamic stretching of the flame [9]. The flame front curvature is caused by its diffusive–thermal [10] and hydrodynamic [11, 12] instability. The increase in the density of the flame surface [13] is a necessary condition for the acceleration of the front. Hydrodynamic stretching is caused by the presence of walls that change the direction of the gas flow. When the flame front approaching the wall of a volume the gas flow parallel to the flame front. These flows increase the surface of the flame and the flow speed of the combustible mixture along the walls.

The increases in the density of the flame front surface and the dynamic stretching of the flame increase with the degree of expansion of the gas during combustion [14]. The degree of expansion is calculated as the ratio of the density of the fuel mixture to the density of the combustion products and depends solely on the thermodynamic characteristics of the combustion process. The second important characteristic of the flame front, which determines its instability and the speed of the combustible mixture in front of it, is the normal speed of the flame. It is determined by the chemical kinetics of the combustion process and the removal of heat from the volume of combustion products affects it slightly. Thus, it can be assumed that the heat loss on the walls of the volume in which combustion occurs have an impact on the flame propagation through the thermodynamics of the process. All these instabilities can be analyzed both experimentally and numerically. There are a number of recent research works on simulations of buoyant flames. The papers [15, 16] discuss the peculiarities of flame propagation in the ultra-lean hydrogen–air mixture. In the papers, the possibility of the stable self-sustained flame ball existence in unconfined space on sufficiently large spatial scales is shown. The comparison of microgravity flame balls and near-uniform limit flame balls at normal gravity propagating in a tube is presented in [17]. A transition from bubble-like flames to cap-like flames is observed experimentally and numerically for lean limit flames in H₂–CH₄–air mixtures in paper [18]. It is important to mention that flame behavior research in microgravity becomes more and more popular. Premixed conical CH₄–air flames were studied experimentally and numerically under normal straight, reversed gravity conditions and microgravity in [19]. In that paper low- and high-frequency flame-instabilities (oscillations) were explored and demonstrated. Another modern research on numerical analyses of diffusion flame behavior in microgravity was presented in [20]. Two modes of evolution of the partially extinguished flame of ethylene and methane burning in the air were shown.

In this paper, large-scale experiments and numerical simulations of premixed near the lower flammability limit hydrogen–air spherical flame propagation are shown. Two possible flame propagation regimes: stable cup-shaped flame propagation and unstable ligament type flame propagation regime are examined for the first time both experimentally and numerically. “flame ball” and “thermal ball” behavior was compared numerically and theoretically.

2. Experiment details
The experimental part of the study was carried out on an installation consisting of a cylindrical shell with a diameter of 1.5 m and a height of 2.4 m, made of high-density polyethylene with a thickness of 0.1 mm bounded by a metal frame. The top was closed with a thin (0.1 mm) rubber shell. The lower part of the structure was covered with a solid aluminum plate. The installation was located inside the VBIK-2 (from Russian “vzryvnaya barokamera”—explosion vacuum chamber), which is part of the Moscow Regional Explosive Collective Use Center at the
Russian Academy of Sciences. The shell was filled with a hydrogen–air mixture with a hydrogen content of 5–6 vol % at normal atmospheric pressure at a temperature $T \approx 293$ K. The filling procedure was carried out as follows. The required amount of hydrogen (0.205–0.245 m$^3$) was added to the shell, which was initially filled with air. The gas in the shell was mixed using a fan with a diameter of 200 mm, providing a flow rate of 5 m/s. Stirring took place within 1 hour, after which the fan stopped. The relative error in the composition of the mixture was about 1%.

After 0.5 hours after the fan stops, the mixture was ignited by the electric spark between two steel electrodes with a diameter of 4 mm in the lower part of the cylinder. During ignition, the mixture was motionless. The energy released in the spark was 1 J. In all experiments, the distance from the ignition point to the surface was 100 mm. To register the sequence of positions and shapes of the flame front in time, an ir camera InfraTek ImageIR 8320 with a spectral range of 2.0–5.7 $\mu$m was used. The water formed during the oxidation of hydrogen emits with a characteristic maximum at a wavelength of 2.8 $\mu$m. Thus, an object captured by an ir camera is the combustion products.

3. Numerical algorithm

Simulations were performed with the CABARET-SC1 computer code. In order to approximate the equations of motion, the CABARET approach is used, the fundamentals of which were developed and elaborated in [21–23]. The main properties of the approach include conservatism, the second order of approximation in time and space on non-uniform grids, low numerical viscosity, and the resolution of shock waves by two computational cells. The low-Mach form of the CABARET method [24] is used in this work for computation of the motion of a multicomponent gas mixture. Specific heat capacities, internal energy, and enthalpies of formation are calculated according to the thermochemical database [25]. Chemical kinetics data was based on the contemporary kinetic scheme presented in [26]. Numerical simulations were done in two-dimensional (2-D) geometry. The ignition of the mixture was modeled with the instantaneous energy release into the small region corresponded to the local mixture heating up to 1500 K. Cartesian grid spacing was 0.2 mm. Two sets of simulations were done. The total number of cells was 750$\times$2500 square cells for a first set and 1500$\times$2500 cells for a second set.

4. Results and discussion

Experimental results showed that there are two different regimes for near the lower flammability limit hydrogen–air spherical flames propagation. The first one is stable cup-shaped flame propagation and another one is unstable ligament type flame propagation. Sequences of ir photographs of a propagating flame were obtained in each experiment. Individual photographs are shown in figure 1 for unstable ligament type flame for 5 vol % hydrogen in the air mixture.

![Figure 1. Infrared images sequence of 5 vol % hydrogen–air flame propagation at time moments of 80, 245, 365, 505 and 695 ms.](image)
Figure 2. Infrared images sequence of 6 vol % hydrogen–air flame propagation at time moments of 95, 205, 405, 565, 685 and 985 ms.

and in figure 2 for 6 vol % hydrogen in air mixture for the cup-shaped flame. In some experiments for 6 vol % hydrogen in air mixture ligament type flame propagation was seen, but it was not so precisely visible like for 5 vol % hydrogen. The accuracy of initial mixture preparation and premixing plays important role here and the resulting precision of mixture composition and homogeneity in our experiments is the key of different combustion regimes for 6 vol % hydrogen in air mixture. The reason for the ligament type behavior is the development of instabilities and still under question.

Semi-unconfined space filled with the near the lower flammability limit hydrogen–air mixture with 6 vol % hydrogen content at normal initial conditions was modeled. Two sets of simulations were performed. The idea of the first simulation set was to compare “flame ball” and “thermal ball” behavior. For that purpose, two simulation cases with and without chemical reactions were done. There is a possibility to turn chemical reactions “on” or “off” in any desired time moment in a numerical code. Initially, both cases started with chemistry switched “on”. The chemical reactions were switched “off” after 0.5 s of mixture combustion in the first case and only thermal evolution of combustion products took place later on. The second case continued with simultaneous combustion and thermal evolution after 0.5 s. The initial stage of 0.5 s of mixture combustion when chemical reactions are turned “on” in both cases is needed for the initial development of instabilities and the formation of an essential amount of hot combustion products. When the process will develop further the combustion products and the flame front
Figure 3. Results of numerical simulations of the 6 vol% hydrogen–air flame propagation at different time moments (left and right parts—simulations with and without chemical reactions, respectively). Density distribution is shown in kg/m$^3$.

will grow further and soon will be very close to the computational domain borders. This process takes approximately one second. The influence of borders plays a significant role in numerical
Figure 4. Results of numerical simulations of the 6 vol% hydrogen–air flame propagation at different time moments. Density distribution is shown in kg/m$^3$. Unstable flame buoyancy.

results. So, the time of 0.5 s was selected as the moment when we turned chemistry “on” or “off” to have a compromise between already developed instabilities and hot products from one side and to minimize the influence of borders for further simulations from another side. Half-
space was modeled with the assumption of centerline symmetry for that set. The results of the simulations are shown in figure 3.

The comparison of simulations showed that flame ball and thermic flow (in the case when the chemical reactions were turned off) behavior is quite different. One conclusion is quite obvious that the velocity of the flame ball and thermic propagation is different and flame ball velocity is higher. We consider the difference in densities is the main reason for differences in the velocity. The ratio between the average buoyancy rates of the flame ball and of thermic propagation is nearly 1.4. At the same time density ratio is nearly 1.3. It means that the buoyancy force makes a significant contribution to the ascent acceleration. It was shown in [16] that “the terminal rising velocity can be accurately assessed by the relation for the terminal rising velocity of the closed bubble rising in liquid”. Here it is shown that there exists a significant diversity in the propagation of the chemically reacting gaseous cloud and an ordinary one.

The idea of the second simulation set was to analyze the stability and behavior of the “flame ball”. The second set of simulations was performed with an ignition point located on a central line of mesh to analyze the possible asymmetry of flame propagation. The difference with the first set of simulations is that the computational domain is not half-space but the full-space, so the computational domain is two times larger in the second set of simulations. The results of the simulations are shown in figure 4. One can see that flame is not stable and the so-called cup-shape flame structure becomes unstable after 1100 ms and split on ligaments. Such a behavior looks qualitatively similar to what was seen in experiments (see figure 1), but time mark is different in experiments and simulations. Looking at experiments on figure 1 one can see that flame behavior changed at 245 ms. It is important to mention that only 2-D simulations were performed but the effects of buoyant flame stability are essentially three-dimensional (3-D) and only general trends of flame behavior can be estimated from 2-D simulations. Another important question is the detailed chemistry used in simulations. Detailed chemistry for near the lower flammability limit hydrogen–air mixtures is still a big problem. Even non-dimensional detailed chemistry calculations of reaction delay times or laminar flame speed have eliminated many difficulties. The work will be extended with new simulations in real 3-D geometry and with other detailed chemical kinetics mechanisms.

5. Conclusion

The results of experiments showed that the buoyant flame behavior near the lower flammability limit in hydrogen–air mixtures demonstrate two distinguished cases: stable and unstable. The border between these two regimes is approximately 6 vol% of hydrogen in air, and is very close to a lower flammability limit that is known to be 4.5 vol% of hydrogen in air. The simulations also showed that flame ball behavior becomes unstable after 1100 ms of propagation and similar flame behavior was seen at 245 ms in the experiment. The attempt to analyze the influence of chemistry on flame propagation and stability was done. Here it is shown that there exists a significant diversity in the propagation of the chemically reacting gaseous cloud and an ordinary one. All the simulations were done in 2-D geometry. 3-D simulations will be performed.

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