On the structure and stability of ultra-lean flames

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Abstract. The present study discusses features of flame propagation and stability in ultra-lean hydrogen–air mixtures under terrestrial gravity conditions. Transient flame dynamics, flame structure and the temperature of the combustion products are analyzed numerically. Obtained results allowed us to distinguish the role of natural convection in the development of superadiabatic ultra-lean flame structure and its stability. It is shown that at significantly low hydrogen content in the mixture even superadiabatic temperature of the combustion products could not maintain the stability of the flame ball rising under the buoyancy action. The convective flows break the flame ball already on the early stage, so there is no opportunity for stable flame ball to be formed. The corresponding value of hydrogen concentration defines the lean concentration flammability limit which is estimated as \( \approx 5.25-5.5\% \) and occurs to be larger than one estimated from spherically symmetric calculations (3.45%).

1. Introduction
Issues related to the combustion processes in lean gaseous mixtures are paid close attention by both industrial and scientific communities. Solutions for many fundamental combustion theory problems such as determining concentration limits, studying mechanisms of flame development in the lean mixtures, and obtaining criteria for its stable propagation can be easily applied in many branches of industry and energy sector. Fire and explosion safety problems are traditionally of the greatest concern [1]. However, known features of the lean combustion, such as low emission of the nitric oxides and other greenhouse gases, small thermal and dynamic loads, define possible benefits of utilizing lean combustion for propulsion devices and power plants [2].

Today combustion of hydrogen-based mixtures is one of the most relevant topics due to great prospects of hydrogen use as a renewable and non-polluting energy source. However one of the acute issues on the way towards the successful daily use of hydrogen as a replacement for fossil fuels is a development of the reliable fire and explosion safety equipment. According to the most probable emergency situation scenario, hydrogen-based combustible mixture can be formed as a result of accidental hydrogen emission into surrounding air atmosphere [3]. Ejection of hydrogen into surrounding air can occur as a result of fuel tanks or pipelines depressurization. During emergency on the nuclear power plant, a similar process of the hydrogen gas accumulation in the atmosphere under containment takes place as a result of zircaloy oxidation reaction [4]. For large confined spaces with the source of a hydrogen leak, such as garages, hangars or depots, volumetric fuel fraction can be small so the combustible mixture can have lean or even ultra-lean composition with equivalence ratio close to the lean flammability limit. Such mixtures have relatively low burning velocities and the main mechanism of the flame propagation is the transport of the hot combustion kernels due to convective forces. Despite moderate pressure and
thermal loads caused by lean mixtures burn out, the convective drift of the flame kernels can be a serious threat, as the temperature of combustion products is still high enough to initiate combustion processes in more reactive media or to cause equipment breakdown.

It is known [5, 6] that, in near-stoichiometric and lean hydrogen–air mixtures with hydrogen fraction larger than 10%, the combustion develops in a classic deflagration regime. The main mechanism of such combustion wave propagation is heat transfer from the hot combustion products to the fresh mixture. As hydrogen fraction decreases lower than 10% the mixture becomes ultra-lean and the diffusion of deficient species, which is hydrogen for lean hydrogen-based mixtures, into the reaction zone becomes the leading mechanism of the combustion reaction support. Herewith temperature of the combustion products for such flames exceeds the adiabatic value. In zero or microgravity conditions flame kernels in ultra-lean flames have a specific spherical shape, so-called flame balls. Previously stability and structure of the non-adiabatic “flame balls” in microgravity was thoroughly studied both experimentally [7, 8] and theoretically [9–11].

Peculiarities of the ultra-lean flame dynamics subjected to the natural convection because of the buoyancy of the hot combustion products were not so comprehensively analyzed as stationary flame balls in microgravity conditions. However, it is known that convection can play a crucial role in flame propagation in ultra-lean mixtures determining its stability or on the contrary causing flame quenching depending on the conditions [12, 13]. In turn, the intensity of the convection is governed by the temperature of the hot combustion products that is superadiabatic for ultra-lean flames. Number of recent papers are devoted to the analysis of the ultra-lean CH₄–air flame balls in terrestrial gravity conditions [14–18]. However, those papers study flame-balls stabilized in external flow inside confined channels. In this case, the stability of flame balls is maintained by different mechanisms in contrast to freely propagating flame ball in terrestrial gravity. External flow together with confinement restricts flame lateral extension and reduces the effect of convection. In the present paper, we provide results on numerical simulations of the ultra-lean hydrogen–air superadiabatic flame propagation dynamics in unconfined space and analyze the influence of the convective motion on flame stability under terrestrial gravity conditions.

2. Problem setup and numerical method

Let us consider following problem setup. Initially, semi-unconfined space is filled with the ultra-lean hydrogen–air mixture with 5–9% hydrogen content under normal conditions (T = 300 K, p = 1 atm). The bottom wall is isothermic and its temperature is equal to the initial ambient temperature of the mixture (T_{wall} = 300 K). The ignition of the mixture occurs near the bottom wall in the small region initially heated up to 1500 K. The cell size of the Cartesian numerical grid is 0.2 mm. That value was determined from one-dimensional flame velocity convergence tests for the considered combustible mixture compositions.

Mathematical model on the basis of the Navier–Stokes equations in the low-Mach number approximation was used to reproduce dynamics of flame ball propagating through the ultra-lean hydrogen–air mixture under the action of the buoyancy force. To achieve detailed description of the simulated processes the thermal conductivity, multicomponent diffusion and energy release due to chemical transformations were also taken into account. Chemical kinetics was calculated according to the contemporary kinetic scheme presented in [19]. Governing equations are the following:

\[
\frac{\partial \rho}{\partial t} + \frac{\partial \rho u_j}{\partial x_j} = 0, \quad (1)
\]

\[
\frac{\partial \rho Y_k}{\partial t} + \frac{\partial \rho Y_k u_j}{\partial x_j} = \frac{\partial \rho Y_k V_{k,j}}{\partial x_j} + \rho \left( \frac{dY_k}{dt} \right)_{chem}, \quad (2)
\]
In a leaner mixture, e.g., with 5% hydrogen content, the mixture reactivity is not sufficient to overcome flame stretch induced by the vortical structure. Flame surface breaks off in the upper (leading) point of the flame, so the flame kernel reorganizes into the spatially separated regions (in figure 1(b), in planar cross-section, it looks like the formation of two independent flame kernels). Being located near the center of the vortical motion temperature inside these kernels is supported by the flow kinetic energy dissipation. However spatial scales of these flame structures are not enough to maintain stable combustion so finally reaction quenches. Figure 2(a) demonstrates the evolution of the flame ball size (area in two-dimensional representation) normalized by the ignition-source area (i.e., the area of initially heated region). Local regions of
Figure 1. Flame structure for time instant 180 ms after ignition: (a) mixture with hydrogen content 6%; (b) mixture with hydrogen content 5%. Streamlines are illustrated by the solid lines with arrows.

Figure 2. (a) Flame area normalized by the area of initially heated region: red line—mixture with hydrogen content 6%; blue line—mixture with hydrogen content 5%. (b) Flame structure for time instant 270 ms after ignition in mixture with hydrogen content 5%.

flame area decrease on the blue line (case of 5% hydrogen–air mixture) correspond to the flame surface breakup events. The breakup event starting at $\approx 90$ ms results in the formation of the structure represented in figure 1(b). Here it should be noted that such a structure occurs to be stable for $\approx 120$ ms (from 140 to 260 ms). Subsequently burning kernels breakup in smaller ones, figure 2(b). These burning kernels of smaller scales occur to be less stable, and finally, the global quenching of the combustion takes place.

Similar combustion mode in the ultra-lean mixture was obtained in paper [28] where radiation was incorporated in the mathematical model while gravity was not taken into account. In this study authors demonstrated that stretch and local break off of initially slightly disturbed planar flame induced by the thermal-diffusion instability enhanced by the radiation can result in the formation of similar isolated circular flame kernels. However, in zero-gravity conditions, those flame kernels are shown to be stable. To compare conditions of stable flame propagation in
microgravity conditions and in terrestrial gravity we performed calculations of the temperature of combustion products for different mixture compositions in both cases (figure 3). It is known that equality condition between crossover temperature, at which the induction time is equal to the exothermal reaction time, and the maximum flame temperature is required for stable flame propagation. According to the classic combustion theory, stable deflagration wave can be formed only at hydrogen content higher than 9% (see the intersection between green and black lines determining the critical conditions for stable deflagration). For near limit flames, the considered criterion is still valid, although the maximum temperature in these flames is no longer adiabatic due to the switch to the diffusive mechanism of combustion. Therefore, the stable combustion could proceed even at lower hydrogen content in the mixture ($< 9\%$).

Considering two conditions—micro- and terrestrial gravity—one can see that concentration combustion limit in both cases is much lower than that estimated for adiabatic flames. But it should be noted that other physical mechanisms become to play the leading role in the limitation of flame propagation. Due to this, one can observe flame quenching even at superadiabatic temperature. In particular the presence of gravity results in the gas-dynamical breakup of the flame ball on the early stages of its formation, so even at high enough superadiabatic temperature of combustion products there is no stable flame already at 5.25% content of hydrogen. Thus, the critical composition for the ultra-lean flame in terrestrial gravity conditions is that with hydrogen content $\approx 5.25–5.50\%$, whereas in microgravity lean combustion limit equals to 3.45%. Obtained
value for microgravity lean combustion limit is in a good accordance with previously reported numerical investigations (in [29] obtained lean combustion limit is 3.4%) and experimental data (in [30] measured lean combustion limit is 3.6 ± 0.1%). Lean flammability limit for hydrogen–air flames in terrestrial gravity conditions was reported in [31] as 6 ± 0.3%.

4. Conclusion
Many physical mechanisms determine the stability of the flame kernels, which develop in ultra-lean hydrogen–air mixtures. It is well known that in microgravity conditions combustion in form of stationary spherical flame balls with a temperature of combustion products greater than adiabatic can be stable even in mixtures with much leaner composition than lower concentration limit for freely propagating planar flames. According to the results provided in present study stable superadiabatic ultra-lean combustion can be also obtained in terrestrial gravity conditions where stability of flame kernels is mainly determined by convective motion that establishes already on the early stage of flame development. Convective flows of the gaseous mixture in the form of three-dimensional vortical structure interact with the flame kernel and determine stretch of the flame surface and as a result the subsequent dynamics of the flame kernel. In particular, it is shown that this effect can cause a local breakup of the flame surface and eventual flame quenching. So, even super-adiabatic flame ball occurs to be unstable and can quench in terrestrial conditions. Due to this, the concentration limits for stable flame ball formation occur to be narrower compared with the case of microgravity conditions but still wider than for adiabatic planar flames. The particular value of lean concentration limit obtained in the framework of this research is 5.25–5.50% of hydrogen in the mixture. For authors knowledge, obtained value correlates well with available experimental data [31]. It should be however noted that contrary to the experimental works the calculations are carried out for the spatially unconfined case, and the results are not affected by the factor of confinement that is rather difficult to realize in the laboratory scale experiment. In view of this, the obtained value of lean flammability limit can be expected as one applicable for large industrial buildings. In real conditions in mixtures with higher hydrogen content stable upwardly propagating kernels can lead to much more severe sequences as it was shown recently in [27]. That fact together with the value of lean concentration limit estimated here should be taken into account in the development of fire and explosion safety measures for nuclear plants and facilities of hydrogen energy.

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