CRITICAL MORPHOLOGICAL PHENOMENA DURING ULTRA-LEAN HYDROGEN-AIR COMBUSTION IN CLOSED HORIZONTAL HELE-SHAW CELL

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ABSTRACT

Free quasi-two-dimensional outward propagation of the ultra-lean hydrogen-air flames was studied in a horizontal, closed flat channel in order to minimize the influences of gravity and natural convection. Experiments were carried out with a sequential change of initial hydrogen concentration in the premixed gaseous hydrogen-air mixtures in the range from 3 to 12 vol. % H_2 under normal pressure and temperature conditions. Two types of critical (in term of concentration threshold behavior) morphological phenomena were observed - formation of a pre-flame kernel and primary bifurcation of the pre-flame kernel and the higher order (secondary, tertiary, etc.) bifurcations of the individual locally spherical and restricted in space flame fronts. For the given initial ambient conditions (channel thickness, initial gas mixture pressure and temperature) variation of initial mixture stoichiometry results in a few substantial changes in overall flame shape. These changes were recorded at the specific concentration limits, which delineate three characteristic macroscopic morphological forms (morphotypes) of the ultra-lean hydrogen-air flame's "trails", – "ray-like", "dendritic", and "quasi-uniform". Transitions between the revealed basic flame morphotypes took place in different ways. The "pre-flame kernel-to-rays" and "rays-to-dendrites" transitions were abrupt and resembled the first order transitions in physics. Transition "dendrites-to-quasi-uniform morphology" were significantly blurred and can be regarded as analogue to the second order transitions.

1.0 INTRODUCTION

One of the specific features of hydrogen-air premixed gaseous combustion under the Earth gravity conditions is a difference between concentration limits for upward (around 4 vol.% H_2) and downward (around 9 vol.% H_2) flame propagation [1-3]. In this concentration range, which can be referred hereafter as an ultra-lean hydrogen-air combustion, the upward propagating flames exist only.

Inspite of a long history [4] of and advances [5] in the experimental and theoretical studies of the ultra-lean hydrogen-air combustion its understanding and quantitative characterization is far from complete up to now.

Results of visual observations of the different overall macroscopic morphologies of the slow-moving flames freely propagating upward in vertical tubes with different cross-sectional sizes (3-30 cm) and heights (up to 4,5 m) and in closed vessels (up to 170 litres) under terrestrial conditions have been described in [6]. Two experimental facts were established. First, overall (at scale of experimental tube or vessel) shape of flames is dependent upon initial chemical composition of hydrogen-air mixture. Authors used the multiple terms for description of the observed flame morphotypes – "caps of flames", "small balls of flames", "vortex rings of flames", "streaky flames", "globular flames", "flame shaped like upright incandescent gas mantles". Second, for a given stoichiometry of initial ultra-lean gas mixture the flames can undergo substantial visible ontogenetic transformations during their development in time and space from formation to quenching, for example - "caps of flames … resolved themselves into … balls of flames".

Internal structure of the upward flames, propagating in vertical tube with 5 cm diameter and 110 cm long in hydrogen-air mixtures within concentration range 4-10 vol.% H_2 at different pressures, has been invasively studied by using different «coloring» admixtures (CO_2, SO_2, $SiCl_4$, SF_6, $Ni(CO)_4$, $Fe(CO)_5$, CrO_2Cl_2) in [7]. It was stressed, the rising flames do not have a uniform continuous burning front. Disintegration of the initial reaction front, aroused around ignition source, into multiple separate individual flamelets took place. The following terms were used for description of the observed ultra-lean hydrogen-air flame morphologies – "threadlike flame" (Flamme mit Fadenstruktur), "isolated flamelets" (Einzelflammechen), "flame head with subsequent flame "tentacles"" (Flammenkopf mit nachfolgenden Flammen-„Tentakeln").

A want to understand the effects of buoyancy on propagation of the ultra-lean flames and the practical needs in development of the space technologies and in assurance of their safety led to the studies under the low-gravity conditions [8 - 10].

In short duration drop tower [11] and later in aircraft microgravity (μg) experiments [12] flame balls were discovered. The following, mainly, qualitative observations were described. First, flame balls can be stationary and stable for at least 500 seconds. Second, progressive dilution of initial hydrogen-air gas mixture results in the following metamorphoses in overall flame shape. "For mixtures sufficiently far from flammability limits, an expanding initial front, formed by ignition source, evolved into cellular deflagration flame, composed of many individual cells. These cells are regularly subdivided to maintain a nearly constant cell spacing. For more dilute mixtures closer to the flammability limits, the cells formed initially did not split but instead closed up upon themselves to form stable spherical flame structures (the flame balls). For still more dilute mixtures all flame balls eventually extinguished".

Experiments during the STS-83 and STS-94 Space Shuttle missions [13] revealed the additional facts. First, the ultra-lean hydrogen-air flames exhibit a "rich variety of behavior, including cellular structures, cell bifurcations, concentration limits to cell bifurcation, stationary flame balls". Second, a "longer duration of μg experiments is needed to determine the ultimate fate of flame balls in mixtures which do not exhibit cell splitting". Third, "it would be advantageous to eliminate the "coloring" agent CF_3Br", whose involvement changes structure of the flames and their concentration limits.

Goals of this work – to respond to the questions and problems raised in [6, 7, 11, 13] and to fill the gaps still existing in phenomenological understanding and experimental characterization of the ultra-lean hydrogen-air flames using experimental surrogate of the zero gravity conditions – horizontal closed Hele-Shaw cell.

Two problems have been addressed. First problem was – transformations in overall (macroscopic or integral) structural composition of the ultra-lean flames, governed by chemical composition variation. Second one was - evolution in time and space of the individual microscopic components of the ultra-lean flames at given stoichiometry.

Modification of the Hele-Shaw cell was proposed [15] as an in-expensive and capable supplement and, in some respects, alternative to the drop tower and the aircraft experiments on gaseous premixed combustion.

Focus of the reported here experiments (performed in March-August 2019) was on the critical phenomena, which define mechanisms of the flame shape changes and on the concentration limits between the different morphological flame types. Horizontal allocation of a Hele-Shaw cell with sufficiently small distance between flat walls permits to minimize influence of two effects - gravity and natural convection. Gas tightness of combustion chamber and allocation of spark ignition source at axisymmetric center of Hele-Shaw cell, were selected to facilitate a comparison with the results from the previous reference experiments. Three mentioned features of our experimental setup have been conceived to study an ultimate behavior and external (at the geometric scales from 15 cm to 1 mm) structure of the ultra-lean flames, which it was difficult to explore in the previous μg experiments due to their short duration.

The following specific research questions were posed in our work: 1) What are the critical phenomena, which determine an ontogenetic development of an individual basic constituent of the ultra-lean flames in horizontal Hele-Shaw cell at fixed slot width? 2) How many different ultra-lean flame phenotypes (observable macroscopic shapes and associated behavior) exist due to interactions between the microscopic individual basic constituents? 3) How many transitions exist between the different ultra-lean flame morphotypes? 4) What are the characteristic features of these morphological transitions? 5) What are the archetypical elements (basic constituents), which inherent to all ultra-lean hydrogen-air flames forming, propagating and quenching in the horizontal Hele-Shaw? 6) What are the deterministic and statistical indicators, which can be used for quantitative (measurable or computable) characterization of the overall behavior of the ultra-lean flames? 7) How are these integral characteristics dependent upon hydrogen-air mixture stoichiometry?

2.0 EXPERIMENTAL FEATURES

2.1Apparatus

Combustion was initiated in a closed (gastight) cylindrical chamber with internal diameter d = 150 mm. Two (upper and lower) flat bases of cylinder were aligned horizontally in parallel with each other at distance h = 5 mm (see Fig.1). The cylindrical side wall and the lower base were made of caprolon. Through a hole in this base, the chamber was connected to the gas mixture preparation system, as well as to the system for measuring the static pressure in the chamber. The upper base of the chamber was made of transparent flat quartz glass for visual observation and video recording.

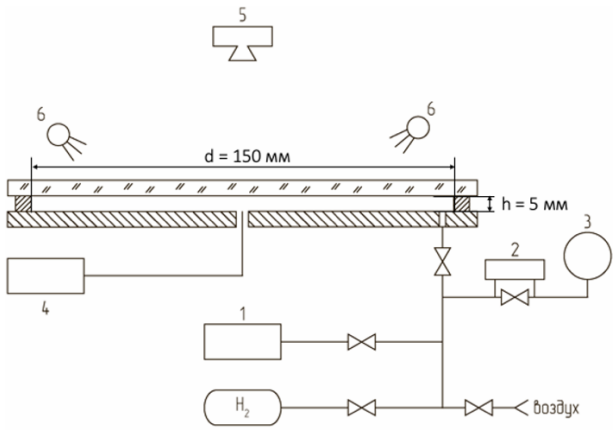

Figure 1. Schematic diagram of the experimental setup: 1 - pump, 2 - differential pressure gauge, 3 - exemplary vacuum meter, 4 - high-voltage discharge generator, 5 - video camera, 6 - backlight.

2.2 Procedures

2.2.1 Hydrogen-air gas mixture preparation

Hydrogen was supplied to the pre-evacuated chamber to a pressure P_{H_2}, then atmospheric air was added to the chamber, the pressure of P_{atm} and the temperature T_0 of which were controlled. Thus, the total pressure of the gas mixture was equal to P_{atm}, and the initial volume concentration of hydrogen $[H_2]_0$ was P_{H_2}/P_{atm}. After a certain exposure, the mixture was ignited. After some time interval, a drop in the static pressure in the chamber was recorded. The value of which was determined by the decrease in the total number of molecules during combustion, as well as the partial condensation of the formed water vapor.

2.2.2 Flame "trail" visualization in reflected light

Condensation of water vapor in the combustion zone near the upper flat optically transparent wall of the chamber (see Fig. 1) made it possible to visualize the flame evolution process by scattering light from an external source on the condensed water microdroplets. Video capturing was made by a Digma DiCAM 400 camera (resolution 1080p@30fps) in reflected light. On the one hand, it ensured a non-invasive nature of the observation. On the other hand, it was possible to record the regions with extremely low luminosity of the ultra-lean hydrogen-air flames.

2.2.3 Ignition

The premixed hydrogen-air gas mixtures were ignited by a spark discharge on the surface of the ceramic dielectric at the lower caprolon base of the chamber (see Fig. 1). The interelectrode distance, which determines the discharge length, was 2 mm.

2.2.4 Fraction of hydrogen burnt determination

Incompleteness of hydrogen combustion was estimated from the decrease in the static pressure in the chamber after the combustion pulse. In this case, the calculations took into account the relative humidity of atmospheric air, as well as the value of the temperature-dependent pressure of saturated water vapor. Condensation contributes to the drop in the static pressure of the mixture if the partial pressure of water vapor in the chamber exceeds the saturated vapor pressure. Otherwise, the drop in static pressure is determined only by the stoichiometry of the combustion process.

3.0 RESULTS

3.1 Critical phenomena and their concentration limits

Three critical phenomena, which are dependent upon ultra-lean mixture stoichiometry, can be observed in the horizontal Hele-Shaw cell. Below they are described in sequence of their appearance during flame evolution from central axisymmetric ignition via internal self-extinguishing to external quenching at side wall. Numerical values of concentration limits, characterizing the critical phenomena, are given below for inter-plate distance h = 5 mm.

3.1.1 Pre-flame kernel formation

Electric spark, initiated by discharge generator, led to formation of a circular-shaped, white-blue-violet flash (Fig. 1a) with $R_{flash} \sim 0{,}01\ m$ in all the studied gas compositions (from 3 to 12 vol.% H_2).

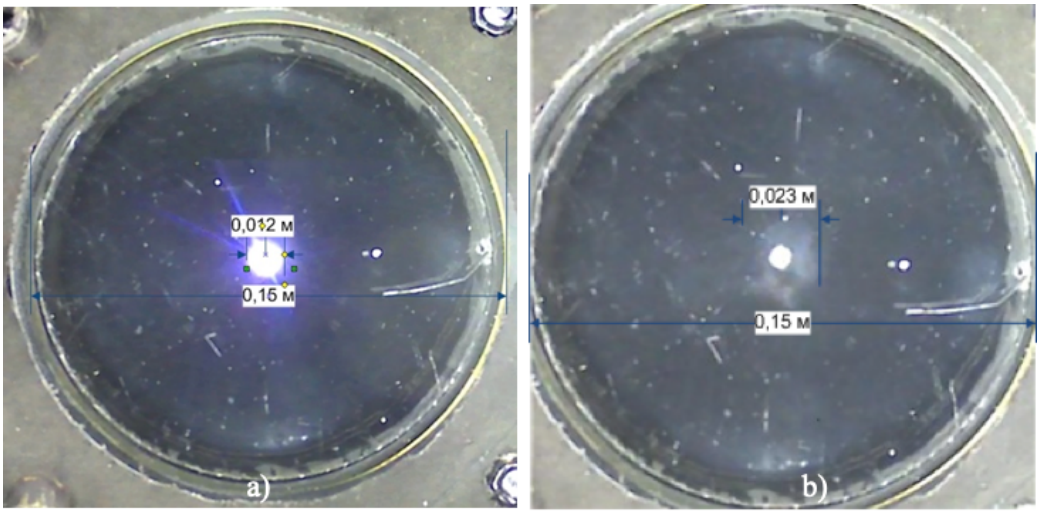

Figure 1. Flash just after spark initiation (a) and subsequent pre-flame kernel formation (b).

After disappearance of the flash, a pre-flame kernel (Fig. 1b) with characteristic scale $R_{pfk} \sim 0{,}02\ m$ was visible by eye as a pale white "cloud" around electrodes. Kernel was recorded in the gas mixtures with hydrogen concentration higher than threshold $c_{H_2} \geq c_{kernel}^{prim} = 5{,}55 \pm 0{,}05$ vol.% H_2 only. It was absent in the gas mixtures with a leaner stoichiometry $c_{H_2} < c_{kernel}^{prim}$.

3.1.2 Primary bifurcation of continuous reaction front in pre-flame kernel

As soon as a radius of outward expanding pre-flame kernel attain a lower critical value $R_{crit}^{min}(c_{H_2})$ (see Fig. 2 with negative photo), a primary bifurcation (see Fig. 3a) of a continuous reaction front occurred. Expansion of the pre-flame kernel was stopped, when its radius attains an upper critical value $R_{crit}^{max}(c_{H_2})$ (see Fig.3b). Self-fragmentation of the pre-flame kernel into a few "rays" was recorded in all gas compositions, where it is exists - $c_{H_2} \geq c_{kernel}^{prim} = 5{,}55 \pm 0{,}05$ vol.% H_2. For example, at 6,7 vol.% H_2 six "rays" were observed, at 7,1 vol.% H_2 – 15 ones. Since number of the "rays" N_r was proportional to c_{H_2} - $N_r \sim (c_{H_2})^1$, - the primary bifurcation of pre-flame kernel can be named "linear".

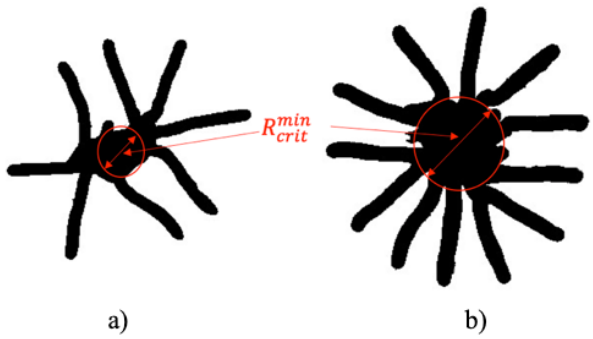

Figure 2. Formation of the ray-shaped nuclei at the outer edge of the pre-flame kernel during primary bifurcation of its continuous reaction front: a) 6,8 vol.% H_2 - 9 nuclei, b) 7,0 vol.% H_2 – 14 nuclei.

3.1.3 Higher order bifurcations of isolated, limited in space reaction fronts

In the gas mixtures with hydrogen concentration higher then $c_{H_2} \geq c_{rf}^{hob} = 7{,}05 \pm 0{,}05$ vol.% H_2 the multiple higher order bifurcations of the isolated (discrete), locally spherical reaction fronts, formed during the primary self-fragmentation of the pre-flame kernel, have been recorded. The secondary, tertiary, etc. bifurcations of a selected discrete flame front are shown in the rainbow colors at Fig. 3b.

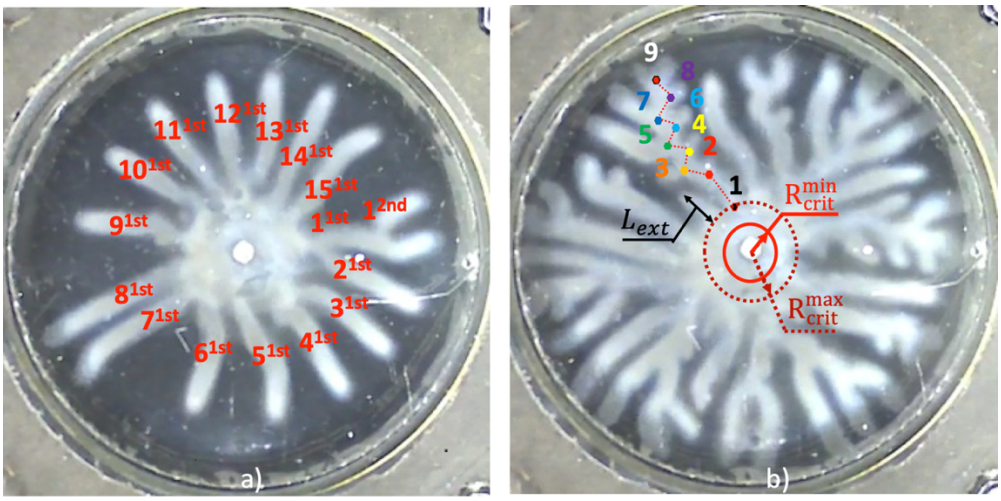

Figure 3. Two types of reaction front disintegration:

a) primary bifurcation of continuous pre-flame kernel into 15 discrete flame fronts at 7,1 vol.% H_2 and b) multiple (eight) higher order bifurcations of a selected discrete flame front at 7,2 vol.% H_2.

A sequential increase in the hydrogen concentration led to a nonlinear increase in the number of the leading discrete reaction fronts, therefore, self-fragmentation of the discrete reaction fronts in the gas mixtures with hydrogen concentration $c_{H_2} \geq c_{rf}^{hob} = 7{,}05 \pm 0{,}05$ vol.% H_2 can be called as "nonlinear" or "branching" self-fragmentation.

3.2 Phenotypes of ultra-lean flames

The two above-described critical morphological phenomena – the primary bifurcation of pre-flame kernel and the higher order bifurcations of the isolated reaction fronts – are characteristic for ontogenetic evolution of the microscopic constituents of the ultra-lean flames, manifested themselves as the isolated, locally spherical flame fronts for a given chemical composition of the hydrogen-air mixture.

Below the following phenotypic traits – observable and measurable properties - of the ultra-lean quasi-two-dimensional flames are described from three viewpoints: three distinct flame morphotypes (overall flame shapes or macroscopic visual appearance), behaviour of their elementary microscopic constituents (isolated flame fronts), and unified quantitative deterministic thermochemical and statistical topological characteristics of the ultra-lean flames.

3.2.1 Basic morphotypes of free 2-dim outward propagating ultra-lean flames

With a sequential increase of the hydrogen concentration (from 3 to 12 vol.% H_2) in hydrogen-air gas mixtures, three characteristic morphotypes of a free, quasi-two-dimensional, outward cylindrical propagation of the ultra-lean flames in a narrow horizontal channel were observed: 1) "ray-shaped" (see Fig.4a), 2) "dendritic" (see Fig.4b), 3) "quasi-continuous" (see Fig.4c).

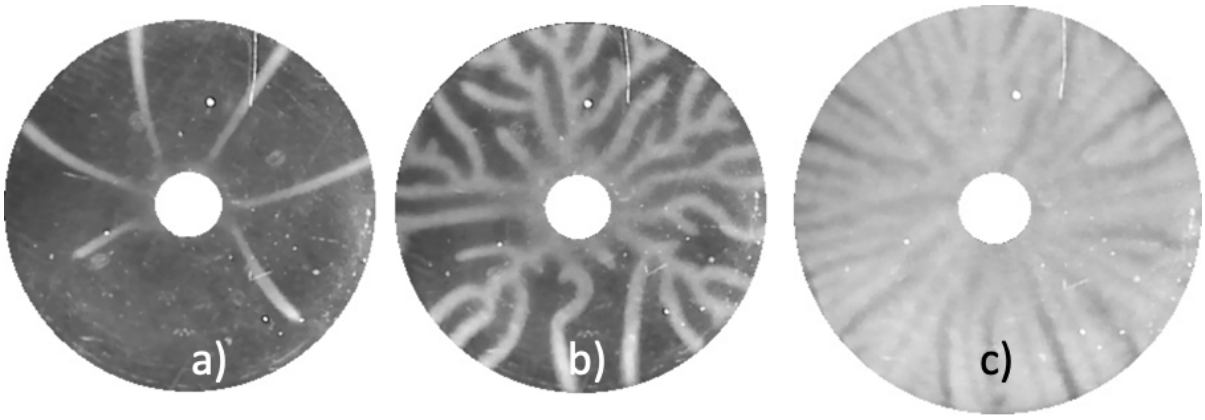

Figure 4. Three characteristic morphotypes of the ultra-lean hydrogen-air flames in a narrow flat horizontal channel, induced by central axisymmetric ignition:
a) "ray-like" (6.3 vol.% H_2), b) "dendritic" (7.2 vol.% H_2), c) "quasi-continuous" (9.0 vol.% H_2).

The observed morphotypes of the ultra-lean flame are distinct from structural viewpoint (overall shape, constituents and their sizes). Their key difference is in the overall visual appearance at macroscopic scale, defined by a size of Hele-Shaw dimensions. Each of the revealed ultra-lean flame morphotype has its own concentration range of existence.

The "ray-like" shape of the ultra-lean flames were observed in hydrogen concentration range from $c_{kernel}^{prim} = 5{,}55 \pm 0{,}05$ vol.% H_2 to $c_{rf}^{hob} = 7{,}05 \pm 0{,}05$ vol.% H_2. For this flame morphotype N_r - number of the visible rays (in fact – discrete, locally spherical flame fronts, which leave the ray-like "trails" of water vapor) - can be used as an observable shape characteristic. For flames with "ray-like"

morphotype, dependence of $N_r \sim (c_{H_2})^1$ upon initial hydrogen concentration is linear, due to "linear" nature of the primary bifurcations of pre-flame kernel.

The "dendritic" flames exist in all gas mixtures, where hydrogen concentration is higher, than $c_{H_2} \geq c_{rf}^{hob} = 7{,}05 \pm 0{,}05$ vol.% H_2. Their visual appearance can be characterized by N_d - number of the leading reaction fronts at outer edge of the dendritic structure. Appearance of the "dendritic" flame morphotype was nonlinearly sensitive to initial stoichiometry variation $N_d \sim (c_{H_2})^{\alpha_d}$ with $\alpha_d > 1$ due to nonlinear growth (or branching self-reproduction) of the discrete leading reaction fronts (see in 3.2.3 description of analogy with chain reactions kinetics).

Global morphologies of the "ray-like" and "dendritic" flames were robust in the repetitive experiments under the same initial conditions. The overall shape, number of the main constituents, their characteristic sizes were stably reproduced. Only direction of the rays and core dendritic components were stochastically changed.

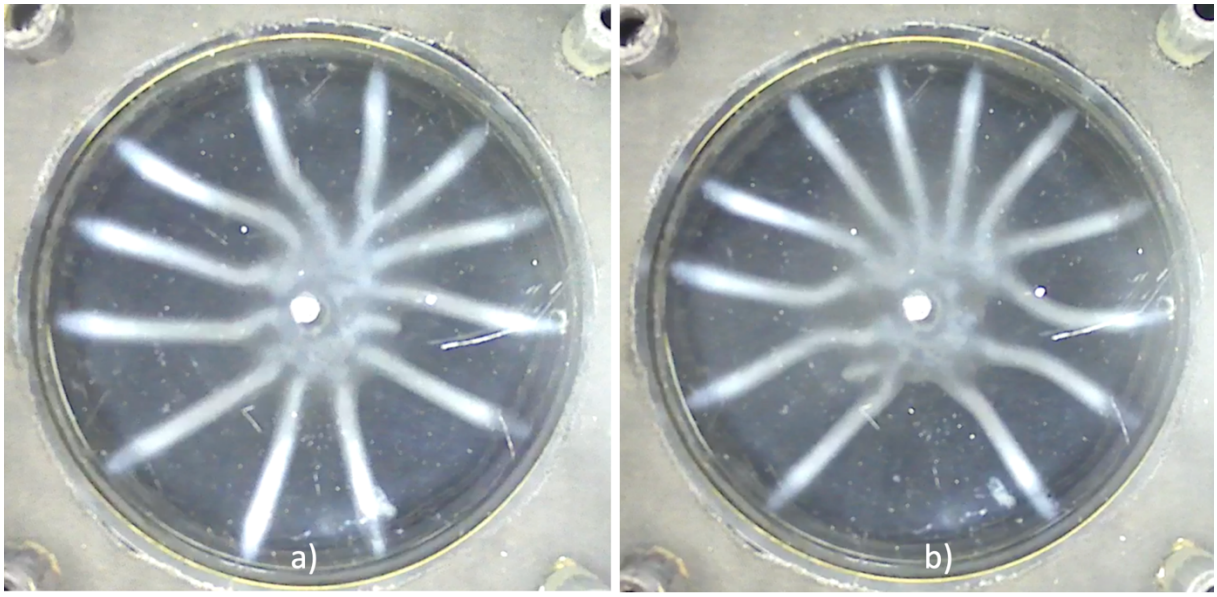

Figure 5. Topological robustness of the "ray-like" flame morphology in the two repetitive tests at 6,9 vol.% H_2: total number of "rays" (12) is preserved, their direction in space is stochastically changing.

A transition from "ray-like" to "dendritic" morphotype was abrupt (a small variation - 0.1 vol.% H_2 – in initial hydrogen concentration results in a drastic shape changing) and resembled the first order transitions in physics.

Morphological transition from "dendritic" to "quasi-uniform" flames is significantly blurred. It occurs in the range [8.1 - 9.1] vol.% H_2 and can be regarded as analogue to the second order transitions in physics and chemistry. In fact, the "quasi-uniform" morphotype is a subset of "dendritic" morphotype with a high density of the elementary constituents.

3.2.2 Ultra-lean flames as a system of the multiple discrete, locally spherical flame fronts

Three-dimensional low-speed, self-sustained combustion of hydrogen-air mixtures with sufficiently high hydrogen concentration in open space or in pipes/vessels spreads as locally plane flames (deflagrations). Specific feature of the self-propagating deflagration flames is, in majority cases, a continuity of their reaction fronts. Level of continuity is dependent upon stoichiometry of hydrogen-air gas mixture. In the rich and near-stoichiometric mixtures reaction front is smooth. In the lean mixtures, where diffusion-thermal instability of plane front takes place, reaction front of the deflagrations is

wrinkled and has cellular structure. However, depending on the ignition method and geometry of ambient environment the laminar deflagration flames are topologically equivalent to a single plane (in tubes) or single sphere (in open space or at earlier stages of outward spherical deflagration propagation in vessels).

In the narrow horizontal channel used for the reported series of experiments, quasi-two-dimensional ultra-lean flame propagation occurred in a different (namely - discrete) topological mode – as an evolution of a system of the multiple, locally spherical combustion foci (leading centers), which interact with each other. During their movement, these leading centers left a "trail" of combustion products (water vapor) on the transparent upper base of the chamber, which was recorded by a video camera. In the concentration range from 6.7 to 9 vol. % H_2, the leading combustion centers were characterized by the following averaged (over set of the test runs) parameters: 1) the characteristic transverse sizes of the foci were in the range from 1 ± 0.5 mm to 5 ± 0.5 mm, 2) the average velocity of the leading centers drifting was in the range from 4 ± 0.7 cm/sec to 25± 0.7 cm/sec.

For all initial and boundary conditions (pressure, temperature, slot width) under study, we could not fix any case of the existence of an evolving flame with single, continuous reaction front (neither stationary nor self-propelling outward).

After self-fragmentation of the pre-flame kernels, all ultra-lean flames consisted of multiple discrete, locally spherical flame fronts in all the gas mixtures under study. Number of these archetypical elements (basic constituents) – which can be regarded as the drifting flame balls [16,17] (in contrast to the stationary flame balls [18]) - and magnitude of interaction between themselves is, mainly, governed by initial hydrogen-air mixture stoichiometry and stochastic fluctuations in space and time.

3.2.3 Basic types of drifting flame balls

Analysis of the video records permits to delineate three types of the drifting flame balls (see Table 1), which are different in terms of their behaviour in time and space.

Table 1. Basic types of the drifting flame balls in horizontal Hele-Shaw cell.

	Discrete reaction front type	Abbreviation	Characteristic for ultra-lean flame morphotype	Behaviour	Concentration limit, vol.% H_2	
					lower	upper
1	self-extinguishing drifting flame balls	SE DFB	ray-like	transient	5.5	6.8
2	self-sustaining drifting flame balls	SS DFB	ray-like	steady	6.8	7.0
3	self-branching drifting flame balls	SB DFB	dendritic	transient	7.1	8.0-9.0
	self-branching drifting flame balls	SB DFB	quasi-continuous	transient	8.0-9.0	12.0

Life-cycle of a self-extinguishing drifting flame ball (SE DFB) consists of the following stages: 1) formation during the primary bifurcation of continuous reaction front at the outer edge of the pre-flame kernel, 2) transient outward propagation, 3) self-extinguishing at some distance $L_{ext}(c_{H_2})$ (see Fig.3b) from the pre-flame kernel, which is dependent on initial hydrogen concentration in gas mixture. At the near-limit but super-critical concentrations $c_{H_2} \sim c_{kernel}^{prim}$ the drifting flame balls disappeared not far - $(L_{ext} - R_{crit}^{max})/R_{crit}^{max} \ll 1$ - from the outer edge of the pre-flame kernel.

Increasing of initial hydrogen concentration (up to approximately 6.8 vol.% H_2 for 5 mm slot width) changes behavior of the drifting flame balls from transient (self-extinguishing) to a sustained mode. After formation at pre-flame edge, the self-sustaining drifting flame balls (SS DFB) propagate steadily to side cylindrical wall and quench there.

Main peculiarity of an individual pathway for a self-branching drifting flame ball (SB DFB) in the mixtures with $c_{H_2} > c_{rf}^{hob} = 7{,}05 \pm 0{,}05$ is a recurrent sequence of the following events (similarly to the chemical chain branching reactions): 1) formation during a primary bifurcation of continuous pre-flame kernel reaction front (chain initiation), 2) propagation at distance $L_{br}(c_{H_2})$ (chain propagation), 3) secondary bifurcation of the discrete drift flame ball into one of three states: a) internal self-quenching due to shortage of available fuel in the near filed (chain termination), b) external quenching at chamber wall (chain termination), c) formation of the only two "daughter" drifting flame balls (chain branching), d) repetition of the cycle.

Total number $N_{br} \sim (c_{H_2})^{\alpha_{br}}$ of the higher order bifurcations (acts of nonlinear branching of the DFB) along the pathway or an overall length of the pathway - $L_{br}^{tot} = L_{br} \cdot N_{br}$ - can be used as the quantitative metrics for characterization of the individual pathways of the "primary" drifting flame balls, formed at pre-flame kernel surface.

Using analogy with kinetics of the chain chemical reaction, behavior of the self-branching drifting flame balls can be attributed as a degenerate chain branching. Inspite of degenerate character of drifting flame balls reproduction, its rate is substantial and results in a "closing" or "overlapping" of the adjacent pathways from the nearby DFB. Two factors - this "overlapping" and "broadening" of the drifting flame balls' size - are responsible for a "quasi-continuous" visual appearance of the ultra-lean flames. Transition from "dendritic" to "quasi-continuous" morphotype occurs in 8 – 9 vol.% H_2 concentration range.

Identification of the three drifting flame ball types can be regarded as an expected, long-awaiting, direct experimental evidence of theoretical prediction on the existence of non-transient self-drifting balls [16] and systematic experimental description of the realistic 2-dim drifting flame ball phenomenology, which expands the existing contemporary theoretical and numerical understanding [17, 19, 20] of the drifting flame balls.

3.2.4 Thermodynamic and topological characteristics of the drifting flame balls

All the 2-dimensional ultra-lean flames can be characterized by the two (at least) following quantitative integral characteristics (indicators), uniformly applicable throughout a whole range of their existence.

Fraction of the hydrogen burnt characterizes an overall reactivity of the 2-dim ultra-lean flames (as well is in case of 3-dim flame propagation) from thermodynamic viewpoint.

Topological dimension of the interface between the combustion products (in our case – water vapor "trails", recorded by video) and the unburnt initial reagents characterizes overall (macroscopic) morphology (shape, constituents, sizes, behavior) of the 2-dim ultra-lean flames. We made two assumptions.

First assumption is - for the geometrical scale region under study – from 1 mm ("microscopic" drifting flame ball scale) to 15 cm ("macroscopic" or "global" scale, restrained by experimental chamber size) – the geometric figures representing the results of combustion, namely, the two-dimensional field of concentrations of water vapor, deposited on the transparent upper base of the chamber, can be considered as physical quasi-fractals [21].

Second one is – in a first approximation a fractal dimension D_F can be used for quantitative topological characterization of all the observed 2-dim ultra-lean flame morphotypes. The dependencies of the

hydrogen burnt fraction ξ_b and the fractal dimension D_F upon initial concentration of hydrogen in hydrogen-air gas mixtures are shown at Fig. 5.

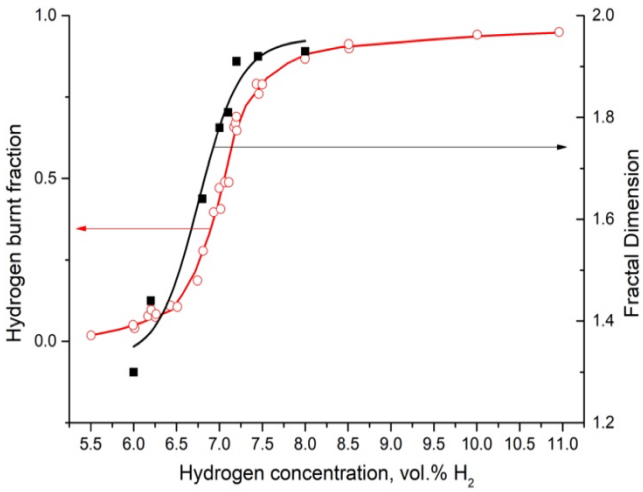

Figure 5. Integral thermodynamic and topological characteristics of the 2-dim ultra-lean flames vs initial hydrogen concentration: hydrogen burnt fraction (red line), fractal dimension (black line).

The inflection point (around 7.0 vol.% H_2 for 5 mm slot width) at the dependencies of the hydrogen burnt fraction ξ_b (c_{H_2}) and fractal dimension D_F (c_{H_2}) upon the initial concentration of hydrogen can be interpreted from two viewpoints. From "macroscopic" viewpoint this inflection point corresponds to a transition from the "ray-like" to the "dendritic" morphotype of the 2-dim ultra-lean flames. From "microscopic" viewpoint it marks the drastic change in drifting flame balls behavior – from "self-sustaining" to "self-branching".

4.0 ACKNOWLEDGMENTS

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CONCLUSIONS

1. It was proposed to use horizontal Hele-Shaw cell and video recording in reflected light to fill the gaps still existing in phenomenological understanding and non-invasive experimental characterization of ultimate behavior and structure of the ultra-lean hydrogen-air flames, which it was difficult to explore in the previous μg experiments due to their short duration.

2. In concentration range under study (3 - 12 vol.% H_2) two critical morphological phenomena were revealed: 1) primary self-fragmentation of the continuous reaction front at outer edge of pre-flame kernel, 2) higher order (secondary, tertiary, etc.) bifurcations of the discrete, locally spherical reaction fronts, birthed during primary bifurcation. The detected critical phenomena define a mechanism of the morphological changes (overall shape, number of constituents and their sizes) of the ultra-lean flames during their free, outward propagation in horizontal Hele-Shaw cell under variation of initial gas mixture stoichiometry.

3. From macroscopic visual appearance viewpoint, any water vapor "trail" from quasi-2-dimensional ultra-lean flames belongs to one of the three characteristic morphotypes: 1) "ray-like", 2) "dendritic", 3) "quasi-continuous". Each identified ultra-lean flame morphotype has its own concentration limits. Transitions between the morphotypes took place in different ways. The "pre-

flame kernel-to-rays" and "rays-to-dendrites" transitions were abrupt and resemble the first order transitions in physics. Transition "dendrites-quasi-continuous" morphology were significantly blurred and can be regarded as analogue to second order transitions.

4. From a microscopic structural viewpoint, all ultra-lean flames consisted of the multiple discrete, locally spherical flame fronts, which interact with each other. Magnitude of interaction between themselves is, mainly, governed by initial hydrogen-air mixture stoichiometry and stochastic fluctuations in space and time of unburned reagents chemical composition. For all initial and boundary conditions (pressure, temperature, slot width) under study, we could not fix any case of the existence of an evolving 2-dim ultra-lean flame with a single, continuous reaction front (neither stationary nor outward self-propelling). Proposed instrumental tools (horizontal Hele-Shaw cell and video recording in reflected light) permit to provide a direct experimental evidence, that drifting flame balls are the archetypical elements (basic constituents) of the 2-dim ultra-lean flames. Three basic types of the drifting flame balls were identified: 1) self-extinguishing, 2) self-sustaining, 3) self-branching.

5. Two integral quantitative indicators for 2-dim ultra-lean flames characterization were proposed: 1) deterministic - hydrogen burnt fraction, and 2) statistical - fractal dimension. It was observed that their dependencies upon initial hydrogen concentration in hydrogen-air mixture are symbatic. The inflection point on both curves corresponds to the microscopic transition from the self-sustaining to the self-branching drifting flame balls and to the macroscopic transition from "ray-like" to "dendritic" morphology.

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