# ON FLAME BALL-TO-DEFLAGRATION TRANSITION IN HYDROGEN-AIR MIXTURES

**Kirillov, I.A.**
**Kurchatov Complex of Transport Nuclear Power Technologies,**
**National Research Center “Kurchatov Institute”, 1, Kurchatov Sq., Moscow, 123182, Russia**
**e-mail: kirillov_ia@nrcki.ru**

## ABSTRACT

Ultra-lean hydrogen-air combustion is characterized by two phenomena: the difference in upward and downward flame propagation concentration limits and the incomplete combustion. The clear answers on the two basic questions are still absent: What is a reason and what is a mechanism for their manifestation? Problem statement and the principal research topics of the Flame Ball to Deflagration Transition (FBDT) phenomenon in gaseous hydrogen-air mixtures are presented. The non-empirical concept of the fundamental concentration limits discriminates two basic low-speed laminar combustion patterns - self-propagating locally planar deflagration fronts and drifting locally spherical flame balls. To understand – at what critical conditions and how the baric deflagrations are transforming into iso-baric flame balls? – the photographic studies of the quasi-2-dim flames freely propagating outward radially via thin horizontal channel were performed. For gradual increase of initial hydrogen concentration from 3 to 12 vol.% the three representative morphological types of combustion (star-like, dendrite-like and quasi-homogeneous) and two characteristic processes of reaction front bifurcation were revealed. Key elements of the FBDT mechanism both for 2-dim and 3-dim combustion are the following. Locally spherical “leading centres” (drifting flame balls) are the “elementary building blocks” of all ultra-lean flames. System of the drifting flame balls is formed due to primary bifurcation of the pre-flame kernel just after ignition. Subsequent mutual dynamics and overall morphology of the ultra-lean flames are governed by competitive non-local interactions of the individual drifting flame balls and their secondary/tertiary/etc. bifurcations, defined by initial stoichiometry.

## 1.0 INTRODUCTION

Hydrogen safety was one of the topical practical problems for the nuclear energy and aerospace applications during the previous nearly three quarters of century. To proactively tackle the potential safety problems in an emerging hydrogen-based economy an improved and more detailed understanding of the different combustion modes, their hazards and appropriate limits is necessary.

The two combustion modes (deflagration (flame) and detonation) were generally distinguished from each other in a waste majority of the works, dedicated to hydrogen safety under severe accident conditions at nuclear power plants [1-8] because they can largely influence the maximum loads from hydrogen combustion sequences and the consequential structural damage. Focus on the so-called “fast flames” and the associated phenomena – flame acceleration, effect of scale, Deflagration-to-Detonation-Transition (DDT), detonation onset, quasi-detonation direct/indirect initiation, – allowed to develop an understanding of phenomenology, mechanisms, critical conditions, qualitative and, in some cases, quantitative criteria for assessing and managing the appropriate high-pressure hazards.

Despite the advances in experimental characterization [9], understanding and modeling [10] of the fast deflagration and the detonation-like phenomena a set of the unresolved problems - practical and scientific - are still exist. One of them is a problem of a justified, non-contradictory and comprehensive identification, characterization, and modeling of the different modes of the “slow” flames and their transition to the “fast” flames. A few simple questions – How many principally different types of the “slow” flames exists, specifically in the ultra-lean hydrogen-air gas mixtures? How and according to what criterion it is possible to distinct different “slow” flames in experiment? What are nature and existence limits (concentration, temperature, pressure, geometrical) of the different “slow” flames? What kind of the “slow” flames can, principally, not accelerate and to transfer into the “fast” flames?

What is a generally accepted and non-contradictory definition of a lower limits for the hydrogen explosions? – are still waiting for a consistent and generally accepted answers.

Report is aimed on the problem statement and the principal research topics of the Flame Ball to Deflagration Transition (FBDT) phenomenon in gaseous hydrogen-air mixtures. The limitations and contradictions of the empirical characterization of the lower concentration limits for the accelerating flames are described. The key theoretical ideas for non-empirical, model-based classification of the slow flames are briefly introduced. Recent experimental results on a driving mechanism of the Flame Ball-to-Deflagration Transition are summarized. A few topics for a future research work on the FBDT are formulated.

## 2.0 EMPIRICAL PHENOMENOLOGY OF FREELY PROPAGATING FLAMES

Probably, the first systemic empirical description of the premixed hydrogen-air flames phenomenology was implicitly proposed in [11]. Here a few important concepts, experimental findings and their generalizations were described: 1) flammability “could and should be regarded as a characteristic property of a gas mixture, apart from the precise means used for ignition and from the form of the vessel that might happen to be chosen for experiment”, 2) definition and criterion of the flammability limits, 3) difference in upward and downward flame propagation, 4) combustion incompleteness in the ultra-lean mixtures with initial hydrogen concentration less then 10 vol.%, 5) multiple morphologies (in terms of shape, constituents, sizes) of the flames - “caps of flames”, “small balls of flames”, “vortex rings of flames”, “streaky flames”, “globular flames”, “flame shaped like upright incandescent gas mantles”, 6) minimal concentration thresholds for manifestation of the pressure effects due to combustion – 7,8 vol.% $H_2$ for upward flames and 9,4 vol.% $H_2$ for downward flames, 7) explicit requirements to hardware and procedures for flammability testing.

### 2.1 Fast deflagration flames

Delineation between the “slow” and the “fast” flames, by comparing with the sound speed of the combustion products, had become actively used in the experimental studies of the various turbulent flame and detonation propagation regimes, which have been identified for hydrogen-air mixtures in obstacle-filled tubes [13, 14]: 1) quenching, 2) sub-sonic, 3) choked, 4) quasi-detonation, 5) Chapman-Jouguet detonation regimes. For quantitative estimation of the concentration border between fast deflagration flames and slow flames – limit for effective flame acceleration – a few empirical correlations were proposed [15, 16].

### 2.2 Slow flames

In parallel to the above cited works, aimed at study, in first turn, the fast deflagration flames and detonation-like combustion regimes, the phenomenology, mechanisms and limits of the different types of the “slow”, predominantly, laminar flames were explored under terrestrial and zero-gravity conditions.

#### *2.2.1 Deflagration flames*

In elongated tubes and in spherical vessels the two basic, topologically distinct shapes of the three-dimensional freely propagating deflagration flames with continuous reaction fronts were studied under the Earth gravity conditions – 1) locally-planar deflagration flames, 2) inward/outward expanding spherical flames. Laminar premixed flames are subjected to two modes of instability of a continuous reaction front (plane or spherical) – hydrodynamic and diffusional-thermal ones [10]. Manifestation of these instability is dependent upon initial stoichiometry of hydrogen-air mixture and stage of flame evolution in time and results in the following flame types: 1) “pulsating” flames in near-limit rich hydrogen-air mixtures, for which the Lewis number $Le$ is greater than unity (see ref. in [17]), 2) “cellular” or “wrinkled” flames in lean mixtures with $Le < 1$ [18], and 3) flames with relatively “smooth” front in the near stoichiometric mixtures.

Transition from smooth to cellular deflagration and role of the cellular structure in extinction of the downwardly propagating flames were studied in [19]. In the near-limit (ultra-lean) mixtures (with hydrogen concentration less then 10 vol.%) tendency of the cellular deflagration flames to break-up into separate flamelets was documented in the independent experimental and theoretical studies.

Microgravity (drop tower and aircraft experiments) was used as an attractive environment for studying slow flames with low Lewis number for two reasons: 1) μg enables observation of near-limit flames due to minimization of the natural convection effects, 2) at μg more pronounced cellular structure of flames may occur.

In experiments under μg conditions [20] the following changes in cellular structure of the deflagration flames were qualitatively observed: 1) "flame fronts in rich H2-air mixtures were smooth and no cellular structure was exhibited", 2) "for sufficiently reactive mixtures, cellular structures resulting from these instabilities were observed and found to spawn new cells in regular patterns. … luminous regions of flame fronts in lean H2-air mixtures are continuous or only slightly broken… The bulk flame front propagates spherically outward but individual cells comprising this bulk front do not increase in size once formed. When the spacing between cells exceeds a critical distance the cell splits into three new cells (which geometrically seems to be the most acceptable value) which quickly grow to the same size and shape as the original cell. The cell structure is regular, and the splitting pattern is consistent from cell to cell...; 3) for less reactive mixtures, cells formed shortly after ignition but did not spawn new cells; instead these cells evolved into a flame structure composed of stationary, apparently stable spherical flamelets… the flame fronts are quite discontinuous and the luminous regions of the cells are very small… the majority of cells are again large but the distribution of cell sizes is bimodal. When viewed in motion, the film records show that the small cells either grow to large cells or occasionally shrink and extinguish whereas the large cells split to form smaller cells."

### *2.2.2 Flame balls*

In mixtures with lowest reactivity (hydrogen concentration is less then 5,5 vol.%), in the drop tower [21] and aircraft experiments [22] the flame balls ("flame bubbles") were discovered. These discrete individual flames were nearly motionless with apparently stationary radius. Experiments during the STS-83 and STS-94 Space Shuttle missions [23] demonstrate that the ultra-lean (hydrogen concentration is less then 10,0 vol.%) hydrogen-air flames exhibit a "rich variety of behaviour, including cellular structures, cell bifurcations, concentration limits to cell bifurcation, stationary flame balls".

Experiments under microgravity conditions were inspired by two seminal works, published a century ago.

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 [11]. 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 [24]. 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").

## 2.3 Basic modes of hydrogen-air combustion and transitions between them

The available experimental data provide solid evidence, that the flammable hydrogen-air gas mixtures can support three basic modes of combustion – flame balls, deflagration and detonation.

Each of the combustion mode possess its own specific morphological (overall shape), structural (reaction front constituents), behavioral characteristics, driving mechanisms and existence limits - in terms of their thermochemical (initial hydrogen-air mixture concentration, temperature, pressure) parameters and geometrical (ambient environment scale and type (open, semi-open, closed, congested)).

Distinction between these basic combustion modes can be made using a few quantitative (measurable or computable) metrics – set of the numbers that give information about a particular combustion process or phenomenon.

From viewpoint of one combustion metric – characteristic velocity of reaction front propagation – the fast flames can exist in form of the two distinctive (from structural viewpoint) combustion types - the turbulent deflagration flames and the Chapmen-Jouguet detonations - and transient ones - quasi-detonations. Phenomenology and mechanisms of the transitions between these individual fast flame types received a great attention in the previous studies. In order to understand the limits of the fast flames existence and to develop the practical methods for avoiding, protection or mitigating explosion consequences the flame acceleration (FA) and Deflagration-to-Detonation-Transition (DDT) phenomena were and is thoroughly studying [5].

Today, it can be assumed (see details in part 3 below), that the slow flames can also exist in two basic forms with substantially different physico-chemical nature. They are – the self-propagating, locally-planar, laminar deflagrations, which can produce substantial baric effects, and, as a other extreme case, the stationary, spherical iso-baric flame balls. Slow deflagration flames have been extensively studied both in the Earth laboratories and under the microgravity conditions.

In comparison with deflagration, a flame ball (FB) is an underdeveloped topic from fundamental science viewpoint now. The same is true for a flame ball-to-deflagration transition (FBDT).

Importance of the FB and the FBDT studies is motivated by the following practical reasons. Specific feature of the hydrogen-air mixtures from explosion safety viewpoint is their substantial stratification in confined (closed or semi-closed) environment under the Earth gravity conditions. In contrast to the fast deflagration flames, which are the direct actors or drivers of the hazardous explosions, the flame balls can be an enabler of explosion. Forming in a place, which is safe from the FA or DDT occurrence viewpoint, it can drift buoyantly upward to the other locations within a stratified hydrogen-air cloud, where hydrogen concentration is higher and, to provoke FA or DDT process. So, experimental characterization, theoretical understanding, numerical modeling and developing of the engineering methods for control and/or prevention of the flame balls in different ambient environments (open, obstacle-laden, gaps) will benefit to enhanced hydrogen safety (standards, procedures, hardware (protection, mitigation), sensors) of emerging hydrogen economy at the Earth and to fire safety of the manned spacecraft missions under the μg conditions.

## 2.4 Lower concentration limits of slow flames

Current empirical understanding of the lower concentration limits for the different slow flame types is summarized in Table 1.

Table 1. Empirical low concentration limits for slow flames.

| Concentration, vol.% H2 | Combustion process / test type | Method/ criterion | Source |
|---|---|---|---|
| 11.0 | Lower Limit (LL) for flame acceleration in horizontal tubes (slow-fast flame transition) | visual/pressure | [16] |
| 10.0±0.8 | LL for flame acceleration in horizontal tubes/channels (slow-fast flame transition) | visual | [25] |
| 9.45 | LL for downward deflagration flame propagation | visual | [26] |
| 9.0 | LL for downward deflagration flame propagation | visual | [11] |
| 8.8 | LL for downward deflagration flame propagation | visual | [27] |
| 8.5 | LL for downward deflagration flame propagation | visual | [28] |
| 8.1 | "lower flammability limit" | pressure | [29] |
| 8.0 | LL for downward deflagration flame propagation | | [31] |
| 8/10 | "explosion severity index" | pressure | [30] |
| 7.7 | LL for downward deflagration flame propagation | visual | [31] |
| 7.5 ± 0.5 | LL for downward deflagration flame propagation | visual/pressure | [7] |
| 7<br>6<br>4 | LL for flammability in vessel 120 l,<br>vessel 20 l,<br>tube | pressure | [33] |
| 5.5 | LL for flame cell splitting in microgravity | visual | [20] |
| 5 | LL for "flame bubbles" existence in microgravity | visual | [20] |
| 4.2 | LL for flammability according to DIN51649-1 | pressure | [34] |
| 4.1 | LL for downward deflagration flame propagation | visual | [11] |
| 3.8 | LL for flammability according to ASTM E681-01 | pressure | [34] |
| 3.75 | LL for flammability according to EN1839(B) | pressure | [34] |
| 3.6 | LL for flammability according to EN1839(T) | visual | [34] |
| 3.35 ± 0.05 | LL for flame ball existence in microgravity | visual | [22] |

Usage of the empirical methods for assessment of the slow flame concentration limits, shown in Table 1, and discriminating the different flame types have the following flaws and limitations. First, absence of a unified criterion, accepted by all stakeholders and reflecting the essential features of the phenomena. For example, the differences (around 15%) in the lower flammability limit values (from 3.6 to 4.2 vol.% $H_2$), estimated according to the ASTM, DIN, or EN technical standards, cannot be reconciled [35] within empirical framework only. Second, variability (around 30%) in the concentration limit values for downward flame propagation (from 9,4 to 7,5 vol.% $H_2$) is, probably, related with differences in shape, scale, material of the test vessels/tubes and/or criterion, selected for assessment in the laboratory-scale studies. This rather high uncertainty demonstrated even in well controlled experimental conditions hinder comparison of the results obtained at lab and large-scale experiments and studies of the transient phenomena. Third, overlapping of the concentration ranges, associated with the different slow flame phenomena. Fourth, absence of understanding of hierarchy and relations between different slow flame concentration limits.

In a concentration range (3 – 9.45 vol.% $H_2$), which can be referred hereafter as an ultra-lean hydrogen-air combustion range, both the slow upward propagating deflagration flames (in labs at the Earth) and the flame balls (in drop tower and aircraft experiments) were observed. For this concentration range a lot of the unanswered fundamental questions exit: What is a reason and what is a mechanism for difference in upward and downward reaction front propagation in ultra-lean hydrogen-air mixtures? What are the basic types of the flame balls, which can be distinguished in zero gravity and in terrestrial conditions? What is a driving mechanism for transition from flame balls to cellular flames? At what critical conditions and how the baric deflagrations are transforming into the nearly iso-baric drifting flame balls?

From practical viewpoint (in the hydrogen safety perspective) it will be reasonable to clarify – what kind of the new studies (physical or computational ones) does it necessary to prepare and to perform for accurate identification and characterization of the slow flames?

To answer on the mentioned questions an appropriate theoretical framework is necessary.

## 3.0 THEORETICAL FLAME CLASSIFICATION

### 3.1 Two originating models

From theoretical (analytical modeling or computational simulation) viewpoint, a diversity of the premixed slow flame phenomena (within the known empirical hydrogen-air limits (4.1-75 vol.% H2)) can be derived from two basic theoretical models [42].

The Zeldovich-Frank-Kamenetskii model [36, 37] of the self-sustained, frontal deflagration flames (also known as ZFK model) is a critical generic component of the ample set of the different specific (subsidiary or daugther) models [38, 39] for laminar (curved, cellular, etc.) and turbulent flames. The ZFK model considers the deflagration flames as a self-similar propagation of reaction fronts (locally plane). Distinctive feature of the deflagration flames is that they can propagate outwardly (to source of ignition) indefinitely in the open (not confined by walls or obstructed by obstacles) space and can occupy all available volume, filled in by flammable gas mixture. Due to density variation across the reaction front, the deflagration fronts can be accelerated and act as the “gas pistons”. In other word, deflagration flames can produce different baric effects.

The Zeldovich model [40] of a stationary spherical flame (aka Flame Ball (FB) model) is ancestry for multiple models [41] for ball-like flames, whose reaction fronts are confined in space, both under Earth gravity and under zero gravity conditions. Key differences in flame behavior, simulated by the ZFK and the FB models, are

- deflagration flames have tendency to self-spreading from ignition point and, as a result, enlarging (in case of absence of external confinement) surface area of reaction front in contrast to space limited reaction front behavior in the flame balls,

- deflagration flames are self-propagating with normal flame propagation velocity in contrast to the flame balls, which can be a stationary (zero velocity) in the uniform gas mixtures or can drift in the ambient non-uniform fields (concentration, density, temperature, etc.),

- deflagration flames produce baric effect in contrast to spherical flame balls. Heating of the combustion products in non-confined deflagration reaction front results in acoustic disturbances, which can, in favorable conditions, results in substantial baric effect. Due to spherical topology of the flame ball-like flames baric effects are absent or substantially diminished in comparison with deflagration flames.

### 3.2 Basal taxa – deflagration flames and flame balls

In [42] it was assumed, that theoretical classification of a complete variety of the observable slow flames can be built, using the planar deflagration flames (DF) and the spherical flame ball (FB) as the two basal taxa. First two levels in a hierarchy of the proposed basic slow flame types are shown at Fig.1.

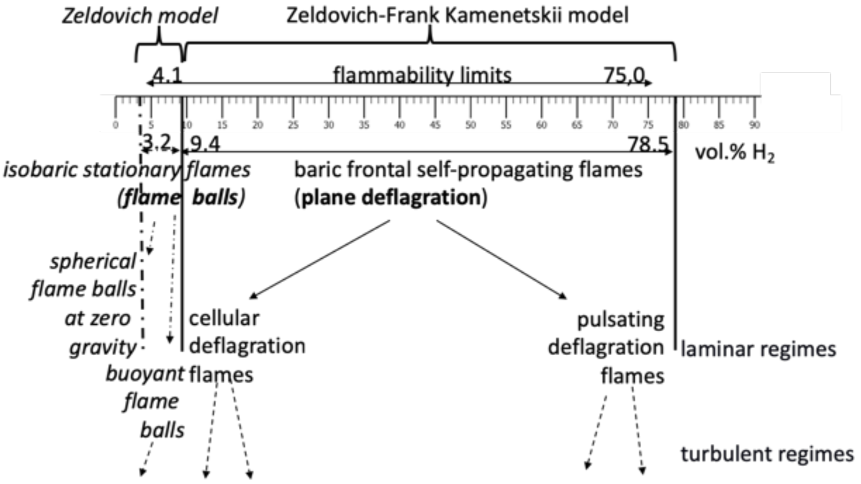

Figure 1. Theoretical taxonomy of the concentration limits for the basic laminar combustion regimes in premixed hydrogen-air gas mixtures under normal conditions (1 atm, 298 K). Cited from [43].

### 3.3 Fundamental Limits

For searching and studying the transition from flame balls to deflagration flames it will be reasonable to use the fundamental concentrations limits. In the non-empirical theoretical estimations (see Table 2), term “fundamental concentration limit” means [44, 45] - an inherent physico-chemical property of a combustible mixture, independent of external influences, associated with or defined by specific experimental setup (type, scale, size, material of test vessel), particular measurement procedure or empirical observation criterion for a given combustion regime (flame ball, deflagration, detonation, etc.).

Table 2. Theoretical concentration limits for slow flames (deflagrations and flame balls).

| Lower Limit, equivalence ratio (vol.% H2) | Upper Limit, equivalence ratio (vol.% H2) | Slow flame type | Source |
|---|---|---|---|
| - | 10.1 (80,9) | plane deflagration | [44] |
| 0.533 (15) | | planar deflagration | [47] |
| 0.294 (11) | - | self-sustained outward expanding spherical deflagration flames | [47] |
| 0.251 (9.51) | - | plane deflagration | [48] |
| 0.247 (9.40) | 8.697 (78.5) | plane deflagration | [43] |
| 0.165 (6.5) | 0.294 (11)<br>dynamic range | self-extinguishing outward propagating spherical flame | [47] |

|  |  |  |  |
|---|---|---|---|
| 0.0863 (3.5) | 0.264 (10.7)<br>static range | stationary FB | [46] |
| 0.0863 (3.5) | 0.165 (6.5) | stationary FB in zero g | [47] |
| 0.298 (11,1) | - | planar deflagration | [51] |
| - | 0.285 (10,7) | FB stability limit | [51] |
| 0.0866 () | 0.34 (12,5) | stationary flame ball | [50] |
| 0.06 (2.4) | - | drifting flame ball | [49] |

Each families of the fundamental concentration limit values have their own uncertainties (aleatoric or epistemic). For example, now the non-empiric calculations of ZFK flame propagation have been made for plane fronts only. These estimations do not take into account cellular structure of deflagration flames in ultra-lean (< 12 vol.% $H_2$) hydrogen-air gas mixtures and associated effects of preferential diffusion and Soret on internal structure of the deflagration reaction front and appropriate concentration limit.

From comparative analysis of the empirical (Table 1) and the fundamental (Table 2) concentration limits the following hypotheses can be made.

First, the estimations made in [48] and [42] provide nearly the same value for the lower concentration limit for deflagration propagation. Small variation is, probably, related with usage of the different values for Chaperon coefficient (third body efficiency) for trimolecular chain termination reaction. This lowest (among non-empirical estimations) value (9,4 vol.% $H_2$) correlate with a highest (among empirical estimations) limit for downward propagation of the deflagration flame and can be used as an upper bond to flame ball-to deflagration transition (FBDT).

Second, in [20] it was made a qualitative description of the morphological changes in overall shape of cellular flames under μg conditions during gradual diminishing of the hydrogen concentration in mixture: cellular flames with cells, spawning new cells -> separate cells, which do not spawn new cells -> stationary, stable spherical flamelets. However, due to short duration of the μg experiments (both in drop tower, in aircraft, in STS missions) and usage of invasive visualization method ($CF_3Br$ as "coloring" agent) the "ultimate fate of flame bubbles in mixtures which do not exhibit cell splitting" was not studied. To overcome mentioned limitations of the previous reference experiments another experimental setup and another visualization method is required for more detailed and, probably, quantitative study of the driving mechanism of the Flame Ball-to-Deflagration Transition (FBDT).

## 4.0 EXPERIMENTS ON 2-DIM FLAME BALLS-TO-DEFLAGRATION TRANSITION

### 4.1 Zero-gravity simulator

Experimental setup (zero gravity simulator) and video recording in reflected light were used in experiments [52, 53], aimed at systematic study of flame ball-to-deflagration transition in quasi-2-dim free propagation of the ultra-lean flames.

Horizontal allocation of a Hele-Shaw cell with sufficiently small distance between flat walls permits to minimize joint 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 3-dim reference experiments [20]. Video capturing of flames were made in reflected light. Three mentioned features of experiments have been conceived to study an ultimate quasi-2-dim behaviour 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.

### 4.2 Mechanism of 2-dim flame ball-to-deflagration transition

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.

*4.2.1 Two types of reaction front fragmentation*

Two types of critical (in term of concentration threshold behavior) morphological phenomena were observed - formation of a pre-flame kernel and primary bifurcation of reaction front at its outer edge and the higher order (secondary, tertiary, etc.) bifurcations of the individual drifting flame balls (DFB). Term drifting is used to delineate the moving quasi-spherical flame balls from stationary, spherical ones. These critical phenomena, driving by thermal-diffusional instability of the curved (non-planar) reaction fronts, define both the macroscopic (overall shape) and microscopic (behavior of the individual constituents) features of the ultra-lean flames.

*4.2.2 Behavior of individual drifting flame balls*

An ultimate lower concentration limit for appearance of pre-flame kernel, which was visible as a pale white “cloud” around electrodes was $c_{H_2} \geq c_{kernel}^{prim} = 5{,}55 \pm 0{,}05$ vol.% $H_2$. In mixtures between 5,5 and 6,8 vol.% $H_2$ only self-extinguishing drifting flame balls (SE DFB) recorded, which were formed after primary bifurcation of the pre-flame kernel, moved outwardly and disappear at some distance from pre-flame kernel. In mixtures between 6,8 vol.% $H_2$ and $c_{rf}^{hob} = 7{,}05 \pm 0{,}05$ vol.% $H_2$ the self-sustained drifting flame balls (SS DFB) propagated outwardly up to side wall of test chamber and quench there. In mixtures with hydrogen concentration higher than $c_{H_2} \geq c_{rf}^{hob}$ self-branching drifting flame balls (SB DFB) were recorded. Outward propagation of the SB DFB resembles an avalanche, where at its leading front the self-branching of the drifting flame balls support a steady “density of the FB per unit length” of avalanche perimeter.

All three phases, qualitatively described in [20] for 3-dim FBDT in μg experiments, were observed also in 2-dim FBDT under zero-gravity surrogate conditions too. Quantitative information on the life-cycles of all three types of the drifting flame balls - birth, propagation, quenching – were collected.

*4.2.3 Morphotypes of 2-dim ultra-lean flames*

Competitive interaction of the individual drifting flame balls results in three visual morphotypes (overall shape) of the trails [53] (condensed water vapor) during and after the free, 2-dim propagation of the ultra-lean hydrogen-air flames in Hele-Shaw cell: 1) "ray-shaped", 2) "dendritic", 3) "quasi-continuous". Transitions between the revealed basic flame morphotypes took place in different ways. The “pre-flame kernel-to-rays” and “rays-to-dendrites” transitions $c_{kernel}^{prim} = 5{,}55 \pm 0{,}05$ vol.% $H_2$ and $c_{rf}^{hob} = 7{,}05 \pm 0{,}05$ vol.% $H_2$ were abrupt and resembled the first order transitions in physics. Transition “dendrites-to-quasi-uniform morphology” were significantly blurred, took place within 8-9 vol.% $H_2$ range and can be regarded as analogue to the second order transitions.

## ACKNOWLEDGMENTS

Author is truly thanks to Hans Pasman and Vadim Simonenko for vital humor, multiple professional discussions, useful critics and wise advices.

## CONCLUSIONS

The available direct experimental evidences were reviewed for the Flame Ball-to-Deflagration Transition (FBDT) in the ultra-lean premixed hydrogen-air gas mixtures. With sequential increase of hydrogen concentration in initial mixture the overall morphologies of the slow flames, the structures and behaviour of their constituents are changing in a regular and repeatable manner. This combustion phenomenon is a one more specific feature of the premixed ultra-lean hydrogen-air gas mixtures along with the other well-known ones - difference in the lower concentration limits for upward and downward flame propagation, incompleteness of combustion, sub-adiabaticity of the combustion product pressure.

Qualitative description of the 3-dim FBDT under microgravity conditions were firstly described for the independent sets of the combustion experiments in the drop towers, aircrafts and space flights.

Recent experiments in horizontal Hele-Shaw cell permit to study the ultimate behaviour of the near-limit drifting flame balls, which were difficult to perform in the microgravity experiments, and fill a few gaps still existing in phenomenological understanding of FBDT and in experimental characterization of the quasi-2-dim ultra-lean hydrogen-air flames under minimal influence of natural convection.

Transition from flame ball-to-deflagration can be separated into three observable and measurable components - structural and morphological ones.

From microscopic structural viewpoint they are - 1) birth and decay of the transient self-extinguishing drifting flame balls, 2) birth, steady propagation of the self-sustained drifting flame balls and their quenching at side wall of test vessel, 3) birth and avalanche-like outward propagation of the self-branching drifting flame balls.

From macroscopic morphological viewpoint – competitive evolution of the system of the outwardly propagating drifting flame balls results in three basic shapes (morphotypes) of the flame trails – 1) ray-like, 2) dendritic, 3) quasi-continuous.

Cellular deflagration flames can be regarded as a highly coherent system of the self-breaking flame balls.

Further insight into the flame ball-to-deflagration transition phenomenon can be obtained by systematic identification and quantitative characterization of the topologically distinctive morphologies of the free propagating 3-dim ultra-lean slow flames under the Earth gravity conditions, which were qualitatively described a century ago.

## REFERENCES

1. Shapiro, Z. M., Moffette, R. T., Hydrogen Flammability Data and Application to PWR Loss-of-Coolant-Accident, WAPD-SC-545, Westinghouse Electric Corp., 1957.
2. Sherman, M.P., Hydrogen combustion loads in nuclear power plants and associated containment loads, Nuclear Engineering and Design, 25, 1984, pp.13-24.
3. Shepherd, J.E., Lee, J.H.S., On the transition from deflagration to detonation, in Major Research Topics in Combustion, Springer-Verlag, 1992, pp.439-488.
4. Lee, J.H.S., Berman, M., Hydrogen Combustion and its Application to Nuclear Reactor Safety, Advances in Heat Transfer, 29, 1997, pp.59-127.
5. Flame Acceleration and Deflagration-to-Detonation Transition in Nuclear Safety, NEA/CSNI/R(2000)7, 200.
6. Ciccarelli, G., Dorofeev, S., Flame Acceleration and Transition to Detonation in Ducts, Progress in Combustion Science, 34(4), 2008, pp.499-550.
7. Cheikhravat, H., Chaumeix, N., Bentaib, A., Paillard, C.E., Flammability limits of hydrogen-air mixtures, Nuclear technology, 178 (1), 2012, pp.5-16.
8. Bentaib, A., Meynet, N., Bleyer, A., Overview on hydrogen risk research and development activities: Methodology and open issues, Nucl. Eng. Technol., 47, 2015, pp.26–32.
9. Lee, J.H.S., The Detonation Phenomenon, Cambridge, Cambridge University Press, 2008, 388pp.
10. Sánchez, A.L., Williams, F.A., Recent advances in understanding of flammability characteristics of hydrogen, Progress in Energy Combustion Sciences, 41, 2014, pp.1–55.

11. Coward, H.F., Brinsley F., The Dilution-limits of Inflammability of Gaseous Mixtures, Part I. The Determination of Dilution-limits. Part II. The Lower Limits for Hydrogen, Methane, and Carbon Monoxide in Air, Journal of the Chemical Society, Transactions, 105, 1914, pp.1859-1885.
12. Coward, H.F., Brinsley F., The Dilution-limits of Inflammability of Gaseous Mixtures, Part III. The Lower Limits of some Mixed Inflammable Gases with Air. Part IV. The Upper Limits of some Gases, Singly and Mixed, Journal of the Chemical Society, Transactions, 115, 1919, pp.27-36.
13. Lee, J.H.S., The Propagation of the Turbulent Flames and Detonations in Tubes, in P. M. Rentzepis and C. Capellos (eds.), Advances in Chemical Reaction Dynamics, 1986, pp.345-378.
14. Dorofeev, S.B., Turbulent Combustion and DDT Events as an Upper Bound for Hydrogen Mitigation Techniques, AECL-11762, NEA/CSNIR/R(96)8, 1996.
15. Dorofeev, S.B., et al., Evaluation of limits for effective flame acceleration in hydrogen mixtures, Journal of Loss Prevention in the Process Industries, 14 (6), 2001, pp. 583-589.
16. Cicarelli, G., et al., Fast-flame limit for hydrogen/methane-air mixtures, Proc. Combustion Institute, 37 (3), 2019, ppp.3661-3668.
17. Christiansen, E.W., Law, C.K., Sung, C-J., Steady and pulsating propagation and extinction of rich hydrogen/air flames at elevated pressures, Combustion and Flame, 124 (1-2), 2001, pp.35-49.
18. Addabbo, R., Bechtold, J. K., Matalon, M., Wrinkling of spherically expanding flames, Proc. Combust. Inst., 29, 2002, pp.1527–1535.
19. Mitani, T., Williams, F.A., Studies of Cellular Flames in Hydrogen-Oxygen-Nitrogen Mixtures, Combustion and Flame, 39, 1980, pp.169-190.
20. Ronney, P.D., A study of the propagation, dynamics, and extinguishment of cellular flames using microgravity, 27th Aerospace Sciences Meeting, AIAA, Reno AIAA-89-0157, 1989.
21. Ronney, P. D., Near-limit flame structures at low Lewis number, Combust. Flame, 82, 1990, pp.1-14.
22. Ronney, P. D., Whaling, K. N., Abbud-Madrid, A., Gatto, J. L., Pisowicz, V. L., Stationary premixed flames in spherical and cylindrical geometries, AIAA J. 32, 1994, pp.569-577.
23. Ronney, P. D., Understanding Combustion Processes through Microgravity Research, Proc. Combust. Inst. 27, 1998, pp.2485-2506.
24. von Bohm, G., Clusius, K., Die Structur aufsteigender H2-O2-Flammen, Zeitschrift fur Naturforschung A, 3(7), 1948, 386-391.
25. Dorofeev, S., et al., Effect of Scale and Mixture Properties on Behaviour of Turbulent Flames in Obstructed Areas, IAE-6127/3 or FZKA-6268, 1999.
26. Thompson, H.W., Ztshr. Physikal. Chem, B18, 1932, pp.219-240.11
27. Terpsta, M.A., Flammability Limits of Hydrogen-Diluent Mixtures in Air, PhD thesis, 2012.
28. Schefer, R.W., Flammability Limits of Hydrogen/Air Mixtures, SGS4-20, 2010.
29. Jo, Y.D., Kim, J.J., A method of measurement for the flammability limits of hydrogen, in: Proc. XX IMEKO World Congress “Metrology for Green Growth”, Busan, Korea, 2012.
30. N’Guessan, K., Chaumeix, N., Pavageau, J., Cuvillier, T., Expanding the boundaries of the explosion risk assessment for H2/O2/N2 mixtures in cindiitions relevant to radioactive materials transportation, Por. 19th Int. Symp., Packaging and Transportation of Radioactive Materials, PATRAM, 2019.
31. Pfahl, U.J., Shepherd J.E., Flammability Limits, Ignition Energy and Flame Speeds in H2-CH2-NH3-N2O-O2-N2 Mixtures, Report FM97-4, 1997, Explosion Dynamics Lab, Pasadena, California.
32. La Fleur, A., Ternary and Quaternary Explosion Regions and Le Chatelier’s Formula, Revc. Travaux. Chim. Pays Bas, 56, 1937, pp.442-473.
33. Zlochower, I.A., Green, G., The limiting oxygen concentration and flammability limits of gases and gas mixtures, J. Loss Prevention in Process Industries, 22(4), 2009, pp.499-505.
34. Schroeder, V., Holzapfel, K., Explosion Characteristics of Hydrogen-Air and Hydrogen-Oxygen Mixtures at Elevated Pressures, ICHS, 2005.
35. Britton L.G., Two hundred years of flammable limits, Process Safety Progress, 2002.

36. Zeldovich, Ya.B., Frank-Kamenetskii, D.A., On theory of uniform propagation of flame, Proc. Academy of Sicences USSR, 19, 1938, 693-698.
37. Zeldovich, Ya.B., Frank-Kamenetskii, D.A., Theory of thermal propagation of flame, Journal of Physical Chemistry, 12(1),1938, pp.100-105 (in Russian)
38. Zeldovich, Ya.B., et al., Mathematical theory of combustion and explosion, Science Publ., Moscow, 1980 (in Russian)
39. Clavin, P., G. Searby, Combustion Waves and Fronts in Flows: Flames, Shocks, Detonations, Ablations Fronts and Explosion of Stars, Cambridge University Press, 2016.
40. Zeldovich, Ya.B., Theory of combustion and detonation of gases, 1944, Moscow, Academy of Science USSR (in Russian).
41. Buckmaster, J., et al., Combustion theory and modelling, Proc. Combustion Institute, 30(1), 2005, pp.1-19.
42. Kirillov, I.A., On fundamental concentration limits for basic regimes of combustion in premixed hydrogen-air gas mixtures, oral report on research seminar “Physico-chemical kinetics in gas dynamics”, Institute of Mechanics, Moscow State University, 7 April, 2016. http://www.imec.msu.ru/content/education/seminars/chemphys/2016-04-07.pdf (in Russian).
43. Kirillov, I.A., Physics-based Approach for Reduction Uncertainties in Concentration Limits of “Slow-to-Fast” Flame Transition in Hydrogen-Air Gas Mixtures, Hydrogen Management in Severe Accidents, Technical Meeting EVT1701911, 25-28 September 2018, Vienna, TECDOC, IAEA (in press).
44. Law, C.K., Egolfopoulos, F.N., A Unified Chain-Thermal Theory of Fundamental Flammability Limits, 24th Symp. on Combustion, 1992, 137-144.
45. Ju, Y., Maruta, K., Niioka, T., Combustion limits, Applied Mechanics Review, ASME, 2001.
46. Wu, M. S., Ronney, P. D., Colantonio, R. O., and Van Zandt, D. M., Combust. Flame 116, 1999, pp.387–397.
47. Yuan, J., Tse, S., Law, K., Dynamics of Flame-Ball Formation from Localized Ignition: Effects of Elevated Pressure and Temperature, Proc. Comb. Symp., 29, 2002, pp.2501-2507.
48. Fernandez-Galisteo, D., Numerical and Asymptotic Analyses of Lean Hydrogen-air Deflagrations, PhD thesis, Universidad Carlos III De Madrid, 2009.
49. Fernández-Tarrazo, E., Sánchez, A.L., Liñán, A., Williams, F.A., Flammability conditions for ultra-lean hydrogen premixed combustion based on flame-ball analyses, Int. J. Hydrogen Energy, 37, 2012, pp.1813–1825.
50. Buckmaster, J., Smooke, M., Giovangigli, V., Analytical and Numerical Modeling of Flame-Balls in Hydrogen-Air Mixtures, Combustion and Flame, 94, 1993, pp.113-124.
51. Ronney, P.D., Premixed-gas flames, in: Microgravity Combustion: Fires in Free Fall (H. Ross, Ed.), Academic Press, London, U.K., 2001, pp. 35-82.
52. Denisenko, V.P., Kirillov, I.A., Melikhov, A.S., Critical morphological phenomena during free 2-dim propagation of the reaction fronts in ultra-lean hydrogen-air mixtures under conditions, imitating zero gravity, oral report, IX research seminar “Modeling of nuclear fuel cycle”, 3-7 February 2020, Snezhinsk (in Russian).
53. Denisenko, V.P., Kirillov, I.A., Melikhov, A.S., Plaksin, V.Yu., Critical morphological phenomena during ultra-lean hydrogen-air combustion in closed horizontal Hele-Shaw cell, ICHS2021 (in preparation).