Isotope Effect on the Characteristics of the Flame-Ball-to-Deflagration Transition in Ultra-Lean Hydrogen- and Deuterium-Air Mixtures in Horizontal Hele-Shaw Cell

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1 Introduction

Two specific features of hydrogen-air premixed gaseous combustion under the Earth gravity conditions are known from seminal works of Coward [1,2] – 1) incompleteness of combustion and 2) difference between concentration limits for upward (around 4 vol.% H2) and downward (around 9 vol.% H2) flame propagation and. 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 [3] of and advances [4] 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.

Third specific feature of the ultra-lean hydrogen-air combustion was discovered by Ronney [5] in a set of the microgravity experiments in drop towers, in aircrafts and during Space Shuttle missions. During gradual increase of initial hydrogen concentration (from 3 to 10 vol.% H_2) drastic changes in overall ultra-lean flame structure and properties occur.

In [18] it was coined to name this intrinsic for ultra-lean hydrogen-air combustion phenomenon – as a Flame-Ball-to-Deflagration-Transition (FBDT). Description of the DFBT phenomenon, studied in microgravity, were, mainly, qualitative due to limitations posed by short duration of experiments in drop towers/airplanes/space missions and by invasive character of experiments at Space Shuttle. A lot of the important fundamental and practical questions were unanswered –What is a driving mechanism and main stages of transition from the flame balls to cellular flames? At what critical conditions and how specifically the baric cellular deflagration flames are transforming into the nearly iso-baric drifting flame balls? What are the different (distinct from the macroscopic shape, behavior and dynamics perspectives) sub-types of the flame balls in ultra-lean hydrogen-air mixtures with different stoichiometries? Within what concentration limits the different flame balls exist?

After Ronney's groundbreaking works in microgravity a lot of experimental and theoretical studies of the flame-ball-like phenomena have been initiated.

Maruta et. al. undertook the comprehensive studies under the microgravity [6] and the Earth gravity conditions [6,8] of the multiple and substantially different near-limit premixed and non-premixed stretched [9] combustion regimes in heated tubes and in microchannels with permanent mixtures flow bearing in mind both fundamental knowledge and industrial applications of microscale combustion [10].

Experimental R&D program of Shoshin and de Goey with co-workers [11,12] was focused on behavior and characteristics of the flame balls, attached to ceramic porous burner with by a permanent gas mixture flow supply, under the Earth gravity conditions.

Multiple experimental studies of Kuznetsov et.al [13,14] were performed in vertical Hele-Shaw cell and were, in first turn, aimed to study the flame acceleration and DDT effects. Motivation was to understand potential impact of FA and DDT on safety of the fuel cells.

Earlier experiment of Ronney in the horizontal and vertical Hele-Shaw cells [15] and the experiments of his co-authors and followers [16, 17] were performed using collateral ignition of flames. Sparks were located at the edge of the experimental setup. Primary interest of these experimental sets was to obtain a solid research basis for hydrocarbon-based technologies (engines, microscale heating, etc.) improvements – to understand better the buoyancy and thermal expansion effects on propagation rates and shapes of the premixed gas flames [15], to study role of viscosity, differential diffusion, heat losses [16], to study oscillatory effects [17].

Despite a substantial progress in experimental studies of the separate physico-chemical and gasdynamic effects during flame-ball-like combustion in recent 20 years, a clear integral picture of the Flame-Ball-to-Deflagration-Transition for the free propagation ultra-lean flames, directly observed in 3-dim microgravity conditions at Space Shuttle, is still absent.

Importance of a coherent and, most important, predictive framework for explanation and quantitative modeling of the FBDT phenomenon was recently raised due to pragmatic need [18], induced indirectly by Fukushima accident, - to reduce the uncertainties, still existing in estimation of the lower concentration limits for hydrogen-air flame acceleration under severe accident conditions at nuclear power plants.

To overcome the limitations of the decisive experiments under the microgravity [5] and the Earth gravity conditions [15] and to answer on the above mentioned fundamental questions, raised in [18], it was proposed in [19] to use a horizontal, cylindrically symmetric, closed Hele-Shaw cell with central ignition. Experimental studies [20] of the FBDT in quiescent hydrogen-air gas mixtures in proposed surrogate of "zero gravity" conditions with axisymmetric ignition revealed a set of the innovative characteristics of the outward propagating quasi-2-dim ultra-lean flames in hydrogen-air premixed gas mixtures.

Goal of this report – to compare the essential phenomenological features of the FBDT in hydrogen- and deuterium-air premixed gas mixtures in horizontal Hele-Shaw cell.

2 Experimental data

Details of the experimental setup, measurement procedures and video capturing of the ultra-lean flames in horizontal Hele-Shaw cell are described in [20, 21]. Both hydrogen-air and deuterium-air premixed mixtures were studied in the following experimental conditions: diameter of cylindrical horizontal Hele-Shaw cell – 15 cm, thickness of channel – 5 mm, variation of hydrogen concentration [H_2] in range 4-12 vol. % H_2, variation of deuterium concentration in range 4-16 vol. % D_2 at normal pressure and temperature.

3 Key Characteristics of the 2-dim Flame-Ball-to-Deflagration-Transition

The key characteristics of the quasi-2-dim Flame-Ball-to-Deflagration-Transition, revealed in in [20, 21], consists of two families.

3.1 Qualitative characteristics

3.1.1 Macroscopic morphotypes of the 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.1a), 2) "dendritic" (see Fig.1b), 3) "quasi-continuous" (see Fig.1c).

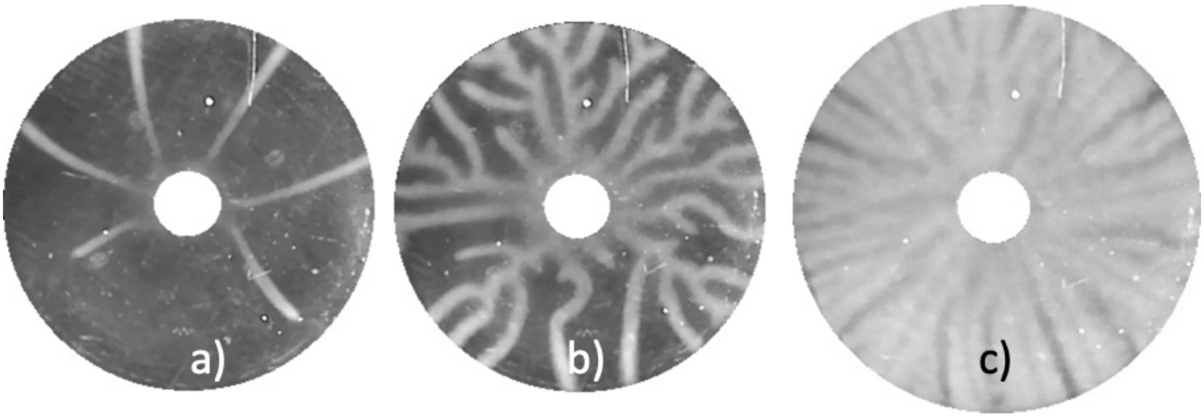

Figure 1. Three characteristic morphotypes of the ultra-lean hydrogen-air flames in a narrow flat horizontal channel, induced by central axisymmetric ignition: a) “ray-like” (6.3 vol.% H2), b) “dendritic” (7.2 vol.% H2), c) "quasi-continuous" (9.0 vol.% H2).

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 scale of the Hele-Shaw dimensions. Each of the revealed ultra-lean flame morphotype has its own concentration range of existence (see Table 1 below).

3.1.2 Microscopic elementary building blocks - drifting flame balls

In contrast to the well-studied 3-dim low-speed, laminar locally plane flames (deflagrations) with continuous reaction front, the quasi-2-dim ultra-lean flames in horizontal Hele-Shaw cell propagate as a system of the multiple discrete, locally spherical flame fronts, which can be attributed [20] as drifting flame balls, predicted in [22] (see Fig.2), in contrast to the stationary flame balls [23].

3.1.3 Basic types of drifting flame balls

Three basic types of the drifting flame balls - as the archetypical elements (basic constituents) – were recorded [20]: self-quenching, self-sustaining and self-branching ones. Mutual interaction between different drifting flame balls is, mainly, governed by initial hydrogen-air mixture stoichiometry and stochastic fluctuations in space and time.

3.1.4 Critical morphological phenomena

During flame evolution (its ontogenetic growth) three critical phenomena, dependent upon mixture stoichiometry, have been observed: 1) formation of pre-flame kernel, 2) primary bifurcation (self-fragmentation) of the kernel into a first generation of the drifting flame balls, 2) higher order (secondary, tertiary, etc.) bifurcations of the reaction fronts of the drifting flame balls themselves.

3.2 Quantitative characteristics

3.2.1 Concentration limits

Concentration limits for distinct types of macroscopic morphotypes and their constituents are listed in Table 1.

Table 1. Basic types of the drifting flame balls in horizontal Hele-Shaw cell (5 mm slot width).

	Discrete reaction front type	Abbreviation	Characteristic for ultra-lean flame morphotype	Behaviour	Concentration limit, vol.% H2	
					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

3.2.2 Integral thermodynamic and topological characteristics of the drifting flame balls

All the observed in [20] 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. Thermodynamic indicator - fraction of the hydrogen burnt (combustion incompleteness) 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 indicator - fractal dimension of the interface between the combustion products (water vapor "trails") and the unburnt initial reagents characterizes overall (macroscopic) morphology (shape, constituents, sizes, behavior) of the 2-dim ultra-lean flames in horizontal Hele-Shaw cell.

4 Results of Comparative Analysis

All abovementioned qualitative features of the FBDT in hydrogen-air gas mixtures were reproduced in the deuterium-air mixtures. All quantitative characteristics – concentration limits and integral indicators (thermodynamic and topological) have appropriate regular bias on stoichiometry scale (see Fig.2).

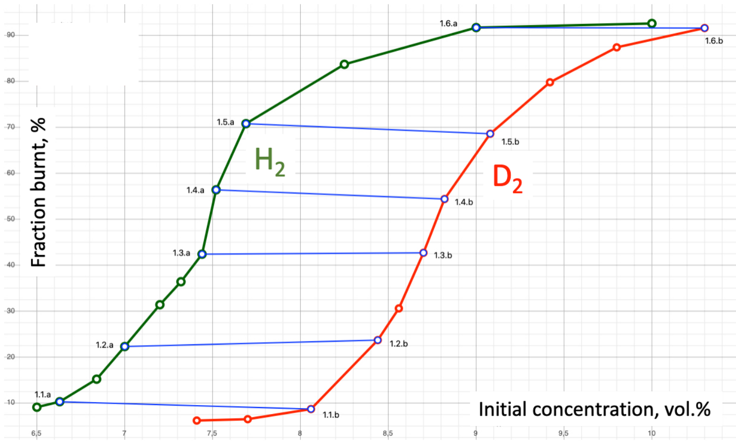

Figure 2. Dependence of the fraction burnt of hydrogen (green) and deuterium (red) versus their initial concentrations in mixtures with air in horizontal Hele-Shaw cell (5 mm slot thickness)

5 Conclusions

1. It was experimentally revealed in horizontal Hele-Shaw cell that the Flame-Ball-to-Deflagration-Transition (FBDT) characteristics in deuterium-air gas mixtures are simbatic to the hydrogen-air characteristics with appropriate regular bias on stoichiometry scale.
2. The following FBDT characteristics were recorded and measured. Qualitative characteristics - 1) macroscopic morphotypes of the ultra-lean flames, 2) microscopic elementary building constituents - drifting flame balls, 3) critical morphological phenomena, governing the mechanism of the FBDT. Quantitative ones: 1) concentration ranges for morphotypes and constituents, 2) dependencies of the fractal dimension of the product-reagents interface and combustion incompleteness upon initial stoichiometry.
3. Observed shift in the critical values of initial hydrogen concentrations is like the shift of the concentration limits for downward propagation of the hydrogen and deuterium deflagration flames, revealed by Koroll and Kumar.

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