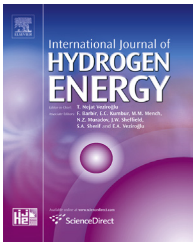
INTERNATIONAL JOURNAL OF HYDROGEN ENERGY 38 (2013) 7510–7518

Available online at www.sciencedirect.com

SciVerse ScienceDirect

journal homepage: www.elsevier.com/locate/he



Effects of non-equidiffusion on unsteady propagation of hydrogen-enriched methane/air premixed flames

V. Di Sarli [a,*], A. Di Benedetto [b]

[a] Istituto di Ricerche sulla Combustione, Consiglio Nazionale delle Ricerche, Via Diocleziano 328, 80124 Napoli, Italy
[b] Dipartimento di Ingegneria Chimica, dei Materiali e della Produzione Industriale, Università degli Studi di Napoli Federico II, Piazzale Tecchio 80, 80125 Napoli, Italy

ARTICLE INFO

Article history:
Received 24 January 2013
Received in revised form
7 March 2013
Accepted 10 March 2013
Available online 1 May 2013

Keywords:
Hydrogen-enrichment
Non-equidiffusive effects
Premixed combustion
Unsteady propagation
Flame-vortex interaction
Large eddy simulation

ABSTRACT

A Large Eddy Simulation (LES) model was developed to simulate the unsteady propagation of hydrogen-enriched methane/air premixed flames around toroidal vortices. Although the LES model does not take into account the non-equidiffusive effects associated with the hydrogen presence (preferential diffusion and non-unity Lewis number), it gives good predictions of experimental data previously obtained for lean mixtures with hydrogen mole fraction in the fuel (hydrogen plus methane) varying from 0 to 0.5. In particular, for each fuel composition, size and velocity of the toroidal vortex generated ahead of the propagating flame front are well reproduced along with the evolution of the flame shape and structure resulting from the interaction with the vortex. The negligible role played by the non-equidiffusive effects has been attributed to the fact that, at the conditions investigated, the characteristic time of hydrogen diffusion is one order of magnitude higher than the characteristic time of flame roll-up around the vortex.

1. Introduction

Enriching natural gas with hydrogen is a valuable method to improve stability of lean premixed combustion in both stationary [1] and mobile [2] systems.

Hydrogen and methane (the primary component of natural gas) are different fuels not only in terms of reactivity (the maximum adiabatic laminar burning velocity is around 320 cm/s for hydrogen and 40 cm/s for methane) [3], but also in terms of transport properties (the molecular diffusion coefficient in air is ~0.6 cm^2/s for hydrogen and ~0.2 cm^2/s for methane and oxygen; the Lewis number, i.e., the ratio of thermal diffusivity to mass diffusivity, is ~0.3 for hydrogen and ~1 for methane) [4].

Substitution of hydrogen to methane increases the laminar burning velocity [5–7], owing to the increase in both supply of active radicals (such as OH, H and O) and flame temperature. Furthermore, it has been found that non-equidiffusion (i.e., preferential diffusion and non-unity Lewis number) effects come into play, especially in lean conditions, making the flame behavior particularly sensitive to the differences between the molecular diffusion coefficients of the fuel and the oxidant, and the heat diffusivity of the mixture [4,6,8–11]. Such effects manifest themselves in local increase in hydrogen concentration (up to more stoichiometric values) and temperature (up to over-adiabatic values) that enhances the burning rate and the flame resistance to extinction [1,4,8–10]. Non-equidiffusion is also responsible for

---

[*] *Corresponding author*. Tel.: +39 0817622673; fax: +39 0817622915.
E-mail addresses: valeria.disarli@irc.cnr.it (V. Di Sarli), almerinda.dibenedetto@unina.it (A. Di Benedetto).

0360-3199/$ – see front matter Copyright © 2013, Hydrogen Energy Publications, LLC. Published by Elsevier Ltd. All rights reserved.
http://dx.doi.org/10.1016/j.ijhydene.2013.03.126

the development of diffusional-thermal cells over the flame surface (diffusional-thermal instability) [11] that increase the surface area [4,6]. For pure hydrogen, it has been found that this increase in small-scale wrinkling can promote the tendency to self-turbulization of outwardly propagating spherical flames (even in the absence of any initial turbulence) [12].

The effective use of hydrogen-enriched natural gas imposes serious consideration to safety issues, particularly those associated with accidental gas explosions.

When an explosion occurs, the premixed flame propagating away from an ignition source undergoes a transition from initially laminar to fully turbulent combustion regime. This transition is triggered by the flow field (i.e., the vortex structures) generated, ahead of the moving flame front (i.e., in the unburned gas), by the unsteady interaction of the flame itself with the obstacles encountered along the path (walls, flow cross section variations, vessels, pipes, tanks, instrumentation, etc.) [13,14]. The obstacle-induced flame acceleration increases the rate of pressure rise [13,15].

In previous papers [16,17], we ran explosion tests for stoichiometric hydrogen-methane/air mixtures in a closed cylindrical vessel. Results allowed us to quantify the link between the increase in laminar burning velocity and the increase in maximum pressure and maximum rate of pressure rise for hydrogen mole fraction in the fuel, x_{H_2}, varying from 0 (pure methane) to 1 (pure hydrogen).

In a more recent work [18], we focused on the dynamic flame-flow interaction during explosions of lean and stoichiometric hydrogen-enriched methane/air mixtures with x_{H_2} varying in the range of 0–0.5. We used Time-Resolved Particle Image Velocimetry (TRPIV) to investigate the unsteady flame propagation around toroidal vortices generated at the wake of a circular orifice. We have found that, for both stoichiometries, as x_{H_2} increases, a transition occurs from a combustion regime in which the vortex only wrinkles the flame front ($x_{H_2} < 0.2$) to a more vigorous regime in which the interaction almost results in the separation of small flame pockets from the main front ($x_{H_2} > 0.2$).

The aim of the work presented in this paper was at quantifying the role played by non-equidiffusion in determining the flow field ahead of unsteadily propagating hydrogen-enriched premixed flame fronts and, thus, the evolution of the front shape and structure resulting from the flame-flow interaction. To this end, a Large Eddy Simulation (LES) model was developed that does not take into account the non-equidiffusive effects associated with the hydrogen presence. Results of LES computations were compared to experimental data previously obtained for lean mixtures [18].

2. Simulated experiments

In Ref. [18], experiments of dynamic flame-vortex interaction were carried out for lean (equivalence ratio equal to 0.8) and stoichiometric hydrogen-methane/air mixtures with hydrogen mole fraction in the fuel, x_{H_2}, varying from 0 (pure methane) to 0.5. To this end, the twin-section combustion bomb sketched in Fig. 1 was used. It consisted of a small cylindrical pre-chamber (height = 35 mm and diameter = 70 mm) linked to a main chamber (150 mm × 150 mm × 150 mm) via a circular orifice (height = 25 mm; diameter = 30 mm; 90° corners at both the inlet and exit faces). Ignition was provided at the center of the bottom end of the pre-chamber starting from quiescent premixed charges. After ignition, the propagating flame front pushed unburned gas ahead of it through the orifice. This gas movement resulted in a toroidal vortex being shed into the main chamber. As the flame progressed through the charge, it interacted with the vortex induced at the orifice wake. In order to characterize the flame-vortex interaction, Time-Resolved Particle Image Velocimetry (TRPIV) was employed. The use of micron sized droplets of olive oil as the seeding material enabled not only recording of the velocity field (ahead of the flame front), but also visualization of the moving flame front (through the consumption of the oil particles by the front itself).

In this work, large eddy simulations were performed for lean mixtures (with different x_{H_2} values), given that the non-equidiffusive effects manifest themselves mainly when hydrogen is the limiting reactant [19].

3. The Large Eddy Simulation (LES) model

The LES model of unsteady premixed flame propagation used in this work has been described and validated previously for

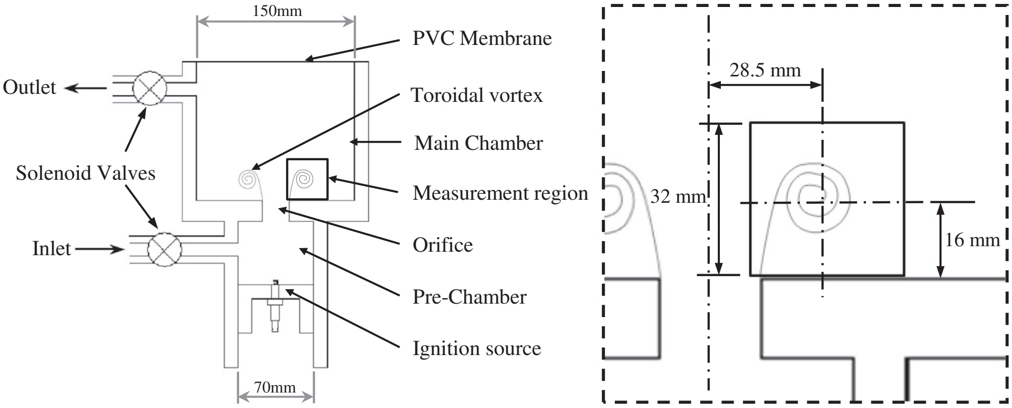


Fig. 1 – Schematic representation of the combustion bomb used in the experiments [18] simulated in this work (not to scale). Details (size and location) of the measurement region are also given.