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Fundamental combustion properties of oxygen enriched hydrogen/air mixtures relevant to safety analysis: Experimental and simulation study

R. Mével^{b,*}, J. Sabard^a, J. Lei^a, N. Chaumeix^a

^a Institut de Combustion, Aérodynamique, Réactivité et Environnement, CNRS, Orléans, France

^b Graduate Aeronautical Laboratories, California Institute of Technology, Pasadena, USA

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ABSTRACT

In order to face the coming shortage of fossil energies, a number of alternative methods of energy production are being considered. One promising approach consists in using hydrogen in replacement of the conventional fossil fuels or as an additive to these fuels. In addition to conventional hydro-electric and fission-based nuclear plants, electric energy could be obtained in the future using nuclear fusion as investigated within the framework of the ITER project, International Thermonuclear Experimental Reactor. However, the operation of ITER may rise safety problems including the formation of a flammable dust/hydrogen/air atmosphere. A first step towards the accurate assessment of accidental explosion in ITER consists in better characterizing the risk of explosion in gaseous hydrogen-containing mixtures. In the present study, laminar burning speeds, ignition delay-times behind reflected shock wave, and detonation cell sizes were measured over wide ranges of composition and equivalence ratios. The performances of five detailed reaction models were evaluated with respect to the present data.

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Introduction

For the last decades, our societies have been facing an energy problem due to the depletion of the natural resources of coal, natural gas and oil. Therefore, there is a strong need to find new sources of abundant and affordable energy. Hydrogen is nowadays considered as the fuel of the future in replacement of the conventional fossil fuels or as an additive to these fuels in order to reduce pollutant emissions. Currently, most of the hydrogen is produced from fossil fuels for cost efficiency [1]. However, the production of hydrogen could be achieved

through the hydrolysis of water using electric energy. Hydrogen would thus constitute the energy vector of the future and enable the storage and use for transportation of electric energy. The production of electricity could be achieved using renewable sources, such as solar and wind energy [2], or using nuclear power plants, currently operating on nuclear fission and, in the future, on nuclear fusion. The production of hydrogen through water hydrolysis induces the formation of a stoichiometric hydrogen–oxygen mixture which would present a very significant hazard in case of accidental release in surrounding air. The production of hydrogen during accidental events in fission nuclear power

* Corresponding author.

E-mail address: mevel@caltech.edu (R. Mével).

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plants also constitutes an important hazard and the recent accident at the Fukushima nuclear complex has demonstrated the continuous relevance of studying hydrogen–air mixtures combustion. The most promising source of energy for the next century is based on nuclear fusion. This possibility led the international community to design the international scientific program ITER, for International Thermonuclear Experimental Reactor. However, ITER operation may rise safety problems. Indeed, because of the interaction of the strong plasma with the wall surface of the vacuum vessel, large amounts of particles, tungsten, beryllium and graphite, can be generated. In case of water or air ingress into the vacuum vessel, the high temperature conditions along with the interaction of steam with particle would produce hydrogen which could possibly lead to the formation of a flammable two-phase atmosphere made of combustible solid particles and hydrogen [3,4].

Several studies can be found in the literature concerning hydrogen based mixtures combustion and parameters such as: laminar flame speeds [5–28] auto-ignition delay times [29–54] and detonation parameters [52,55–85]. The previous studies on the determination of the laminar burning speed of hydrogen–air mixtures include more than 300 data points and have been performed with different experimental facilities including spherical expanding flame in closed vessel and counter flow twin-flame. The ranges of compositions and conditions of these studies are: $\Phi = 0.125\text{--}5.6$; $P_1 = 35\text{--}500$ kPa; $T_1 = 298\text{--}523$ K. In addition, data are available at up to 2 MPa for He diluted mixtures [8]. The different studies on ignition delay-time include more than 1000 data points and cover the following ranges: $\Phi = 0.0625\text{--}2$; $P = 21\text{--}8815$ kPa; $T = 774\text{--}2672$ K; $X_{\text{diluent}} = 0\text{--}0.9985$. Concerning the studies on the sensitivity to detonation, the references previously mentioned include more than 700 experimental data points. The conditions investigated in these studies encompass the following ranges: $\Phi = 0.19\text{--}5.5$; $P_1 = 1\text{--}1236$ kPa; $T_1 = 123\text{--}800$ K; $X_{\text{diluent}} = 0\text{--}0.90$. In addition, it should be noted that the effects of a large variety of diluent have been investigated including air, N_2 , H_2O , CO_2 , Ar, He, CF_3H , CF_4 and their various mixtures. Despite the large amount of combustion related data available on hydrogen–oxygen mixtures, no data could be found in the literature concerning the particular mixtures of interest for ITER conditions. In order to be able to make good assessment of the combustion parameters important in safety analysis and to provide these fundamental data to Computational Fluid Dynamics, CFD, codes, it is important to acquire a detailed experimental database covering a number of physical and chemical properties. The analysis of the propensity of a given mixture to explosion relies on both (i) the combustion and transport properties and (ii) the scale and geometry of the containment [86]. For the combustion properties, in addition to flammability limits and the minimum ignition energy, different properties need to be known in order to perform such analysis, namely:

- The laminar burning speed, and subsequent associated Markstein length, especially its variation with composition and thermodynamic conditions. It is important as it is useful to validate chemical kinetic mechanisms and it is also used as a normalizing parameter in the

determination of the turbulent flame speed in CFD codes [87]. The validation of a chemical kinetic mechanism is important as it is the only mean to derive the global activation energy and the Zeldovich number that is used in explosion studies [88].

- The auto-ignition delay time versus the composition, temperature and pressure. It enables the assessment of the possibility of detonation initiation [61,89]. A short delay indicates that a coupling between a reaction zone and a leading shock wave is possible while a delay on the order of a hundred of microseconds indicates the unlikely-hood of such a coupling.
- Detonation parameters such as the Chapman-Jouguet detonation velocity and more importantly the cell size. The latter is a very good estimation of the sensitivity to detonation [61,89]. A cell size on the order of a millimeter indicates a very high sensitivity to detonation while a larger cell size of hundreds of millimeters indicates the very low sensitivity to detonation. Not only the direct initiation of a detonation can be assessed by the knowledge of this parameter but also the tendency to transit from slow flames to detonation [86,89].

The present study aims at obtaining over wide ranges of conditions and compositions the laminar burning speed, the ignition delay-time, and the detonation cell size for stoichiometric $H_2\text{--}O_2$ mixtures diluted with air.

Materials and methods

Flame speed experiments

The constant volume vessel is a stainless steel sphere (i.d. 476 mm) equipped with two opposites windows (97 mm diameter, 30 mm thick). The inner surface is black polished. Two metallic electrodes located along a diameter of the sphere are linked to a high voltage source. Ignition was produced by an electric spark located at the center of the sphere. The voltage and current of the discharge were measured with a high voltage probe and a current probe as seen in Fig. 1. The spherical bomb is equipped with a Kistler pressure sensor (601A equipped with a flame arrestor model 6505) in order to measure the evolution of the pressure as the flame propagates.

The visualization of the flame is obtained via a Schlieren system. It consists of two concave spherical mirrors (focal length 1 m), the light source was a white continuous lamp and it was made as a point source via one bi-convex lens (focal length 20 mm). A numerical high speed camera, PHOTRON APX, with an acquisition frequency ranging between 2,000 and 120,000 images per second, is used to record the Schlieren images of the growing flames. These images allow the measurement of the radius of the flame as a function of time (see Fig. 2) using an automatic homemade MATLAB program or a manual process with the 5.2 VISILOG Software as described in Dubois et al. [90].

To measure flame speeds, the spherical configuration has the advantage that the flame is well characterized through the experimental assessment of the stretch rate that undergoes

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