Laminar flame speed determination for $H_2/N_2/O_2$/Steam mixtures using the spherical bomb method

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1 Introduction

During severe accident in Pressurized Water Reactor (PWR), the interaction between the fuel rode and steam leads to the build-up of an explosive atmosphere inside the containment building [1]. This atmosphere is mainly composed of hydrogen, oxygen, nitrogen and water. In case of an ignition by an energy source (electrical discharge spark, hot surface, etc.), a flame occurs and is capable to threaten the containment building. To assess this capability, a criterion of flame acceleration permits to distinguish fast flames from slow flames [1]. The criterion of flame acceleration is based on properties of the mixture and geometry [2]. The present study focuses then on the evaluation of fundamental combustion parameters: laminar flame speed, Markstein length, activation energy and Zeldovich number. The experimental work is conducted over conditions relevant to severe accident in PWR: equivalence ratio ($0.8 \leq \varphi \leq 4$), initial temperature (296, 333, 363, 413 K), dilution (N_2/O_2 = 3.76, 5.67, 9), and steam (%mol.). The simulation of laminar flame speeds is conducted using Mével detailed mechanism [3].

2 Experimental details and numerical calculations

2-1 Experimental set-up

The experiments leading to the laminar flame speeds were conducted in two different stainless steel spherical bombs. The first one [4] was used for ambient temperature studies (296K), and the second one [5] for high temperatures studies (333, 363, 413K). These two vessels are similar, they differs only by their volume: 93L and 56L. Both vessels are made of two concentric spheres in between which circulates a heat transfer fluid in order to regulate the inside temperature. Two independent gas manifolds are mounted, one to introduce the reactants and another one to pump the combustion products. For the high temperatures studies, a septum injector nut is directly connected to the vessel for the liquid water injection. For both devices, two opposite quartz windows for optical observations are mounted. The measuring of initial temperature is conducted with K type thermocouples placed inside the vessel. The spark- ignition of

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the mixture is realized by two thin tungsten electrodes located along a diameter of the sphere which are linked to a high voltage and adapted probes to monitor the input power. High frequency pressures transducers (Kistler 601A/6001 ; ± 0.1% full scale ; 0-250 bar abs) are mounted flush with the inner wall to measure the pressure during the combustion. To observe the flame propagation and measure the laminar burning velocity, a Z-type Schlieren arrangement and high-speed camera are employed. Regarding the huge disparity of the laminar flame speeds obtained during the experiments, two different cameras were used. One for the high laminar flame speed (>300cm/s) (Phantom V2520; up to 39 000image/s) and another one for the low laminar flame speed (<300cm/s) (Phantom V1610; up to 33 000 image/s). The spark triggers the camera recording, the measurement of the pressure and the input power via a TTL generator connected to the electrodes.

2-2 Experimental procedure

Experiments were conducted at initial pressure of 100 kPa and initial temperatures of 296, 333, 363, 413 K. For each run, a common systematic procedure was followed. First, the spherical vessel bomb and the gas introduction lines were pumped down to less than 30 Pa by two primary pumps. Then, hydrogen gas, air or nitrogen gas and oxygen gas were successively introduced into the vessel to produce the desired mixture based on the Dalton's law of partial pressures. All these gas components were supplied by Air Liquide (purity>99.9999%). The air is composed of 20.9% O_2 + 79.1% N_2. For experiments with steam, the liquid water (distilled water) was the first component to be introduced with a syringe. Each time, the water partial pressure was checked to be less than the equilibrium vapor pressure in order to make sure that all the liquid phase evaporated. The partial pressures and the initial total pressure were measured using different capacitance manometers: MKS 690A Baratron and MKS 631. The final accuracy on the equivalence ratio is 0.8%. Once the initial total pressure was reached, the bomb was isolated by closing the gas introduction line, and the mixture was resting during 5 minutes before the ignition in order to dissipate the turbulence induced by the filling process. The initial temperature was checked prior to the ignition. After the experiment, the vessel was flushed with compressed air in order to remove all the combustion products, and finally pumped for next experiment. The flame speed for each equivalence ratio was measured two times in order to make sure of the experimental result repeatability.

Propagation flame images obtained with the high speed camera were then processed with a home-made code based on Matlab® in order to determine the evolution of the spherical flame radius in function of the time. This determination permits then to extract parameters (stretch κ, unstretched spatial flame speed V_S^0) leading to the laminar flame speed S_L^0 [5].

2-3 Numerical calculations

Laminar flame speeds S_L^0 are simulated using COSILAB code [6] for freely propagating flames, based on Mével mechanism [3] in order to verify their validity in the conditions of the present work. They are then used to obtain other fundamental combustion parameters as the laminar flame thickness δ [7], the Effective Lewis number Le_{eff} [8] and the activation energy Ea [9].

3 Results

3-1 Laminar flame speed

Experiments were conducted with $H_2/N_2/O_2$/Steam mixtures at 100kPa for an equivalence ratio, φ, ranging from 0.8 to 4. The lean limit of 0.8 was chosen in order to avoid flame wrinkling. Four initial temperatures (296, 333, 363, 413 K) were investigated. In order to test the effect of dilution, the N_2/O_2 ratio was fixed at three values (3.76, 5.67, 9). At N_2/O_2 ratio of 3.76, the effect of steam (0, 20, 30% mol.) was studied. Figure 1 shows the laminar flame speed evolution of $H_2/N_2/O_2$ mixtures with the three different N_2/O_2 ratios (3.76, 5.67, 9) for several initial temperatures. As it can be seen in each case, a higher initial temperature always induces an increase of the laminar flame speed. The position of the peak value is slightly shifted to the rich mixtures when the temperature is increasing. Keeping the temperature constant, it can also be noticed that the laminar flame speed is decreased with the increase of the dilution ratio. In these conditions, it appears that increase the dilution ratio is more efficient to slow down the combustion than the decrease on the temperature. Indeed, the laminar flame speed S_L^0 decreases by 38% when the temperature is changed from 413 K to 296 K, while it declines by 65% when the dilution ratio is changed from 3.76 to 9. Figure 2 shows the Markstein length versus the equivalence ratio for the same conditions. In every case the Markstein length increases with the increase of the equivalence ratio but doesn't seem sensitive to the modification of the initial temperature. It appears that it slightly decreases with the increase of the N_2/O_2 ratio, suggesting an increasing of the the flame front instability while the dilution is increasing.

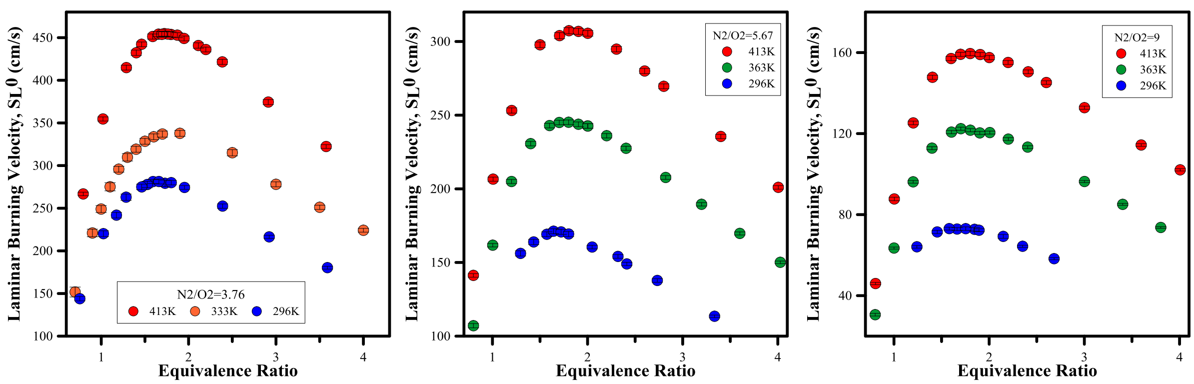

Figure 1. Laminar flame speed versus the equivalence ratio under different initial temperatures (293, 333, 363, 413K) for three N_2/O_2 ratio (3.76, 5.67, 9)

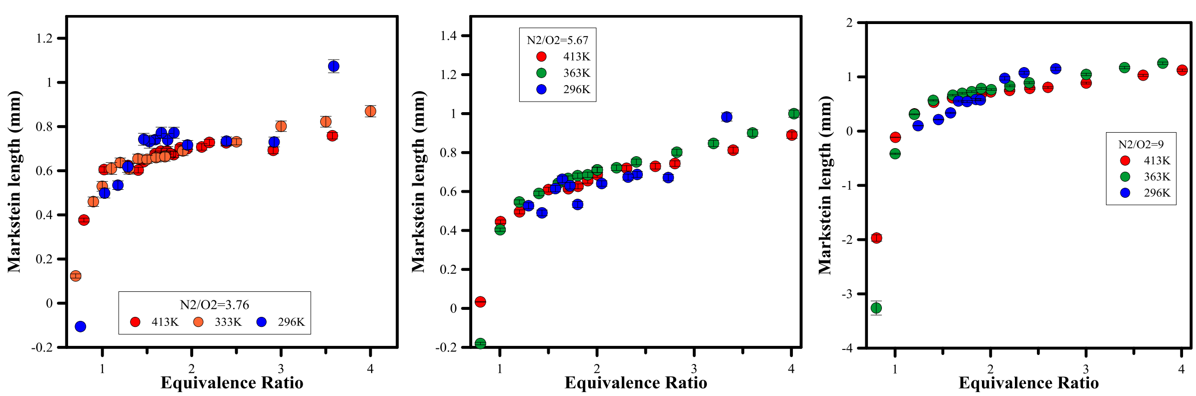

Figure 2. Markstein length versus the equivalence ratio under different initial temperatures (293, 333, 363, 413K) for three N_2/O_2 ratio (3.76, 5.67, 9)

Most of the literature results were determined for H_2/Air mixture (N_2/O_2=3.76) at ambient temperature and are represented in Figure 3. As it can be seen, the obtained results in this study are in good agreement with the experimental results reported by Lamoureux et al. [7] and by Tse et al. [10]. It should be noticed that Hu et al. [11] also conducted studies at 373 K and 443 K.

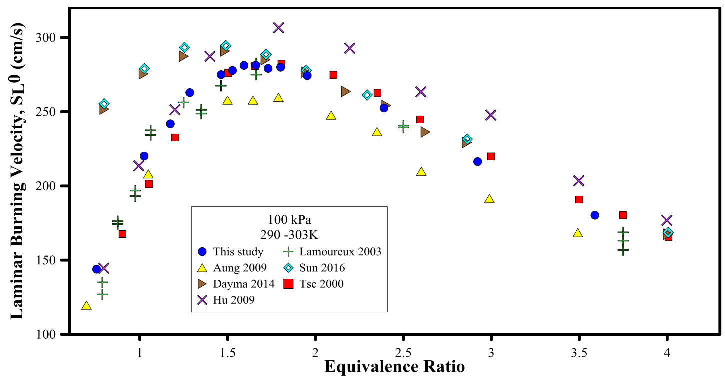

Figure 3. Laminar flame speed comparison for H_2/Air mixtures at 100 kPa and ambient temperature

The influence of steam dilution at 413K on laminar flame velocity and Markstein length are respectively shown on Figure 4 and Figure 5. Figure 4-a presents results for a wide range of equivalence ratio while Figure 4-b presents results for a fixed equivalence ratio (φ=1). It appears that steam dilution is a good suppressor of the combustion. For instance, at φ=1, S_L^0 is reduced by almost 95% when the steam fraction is set to 50%mol. On the Figure 4-a, it can be seen that the position of the peak value is slightly shifted to the lean mixtures when the steam dilution increases. On Figure 5, the Markstein length increase more rapidly when the steam fraction is higher.

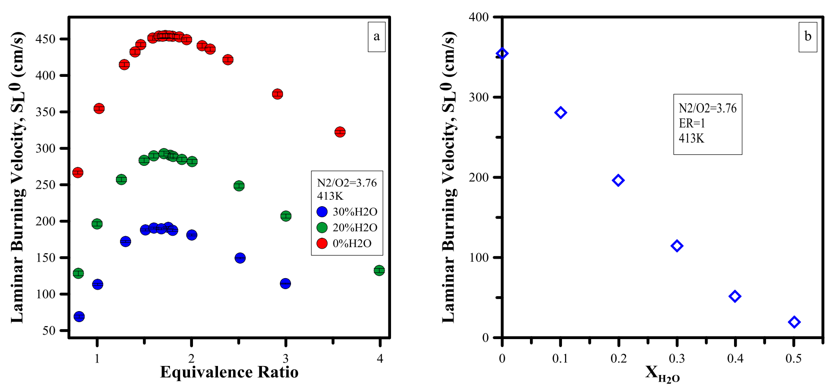

Figure 4. Laminar flame speed (a) versus the equivalence ratio (413K - N_2/O_2 =3.76) with different amount of steam (0, 20, 30%mol.) (b) versus the steam molar fraction (413K - N_2/O_2 =3.76 - φ=1)

3-2 Fundamental combustion parameters

Numerical simulations of the laminar flame speed were performed using COSILAB code [6] for freely propagating flames, based on Mével mechanism [3]. Figure 6 shows the fundamental combustion parameters versus the equivalence ratio (activation energy, Zeldovich number, Lewis number and flame thickness) for $H_2/N_2/O_2$ mixtures at 413 K with and without steam. The simulations show that activation

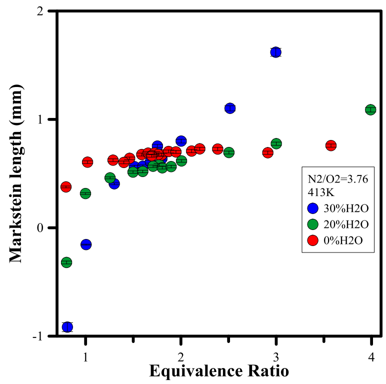

Figure 5. Markstein length versus the equivalence ratio (413K - N_2/O_2 =3.76) with different amount of steam

energy, Ea, increases with the presence of steam. It increases also at N_2/O_2 ratio of 9 but stays equivalent at N_2/O_2 ratio of 3.76 and 5.67. On this parameter the effect of steam seems more significant than the one of N_2/O_2 ratio. For example, the minimum Ea increases by almost 31 kJ.mol^{-1} when the steam fraction changes from 0 to 20% mol. while it is increasing by 10.5 kJ.mol^{-1} when the N_2/O_2 ratio changes from 3.76 to 9. As the Zeldovich number is a dimensionless measure of the activation energy [2], the presence of steam and the increase of the N_2/O_2 ratio have the same effect on it as on the activation energy. The increase of diluent molar fraction (steam and nitrogen) leads to a decrease of the mixture Lewis numbers (Figure 6-c) and an increase of the flame thickness (Figure 6-d).

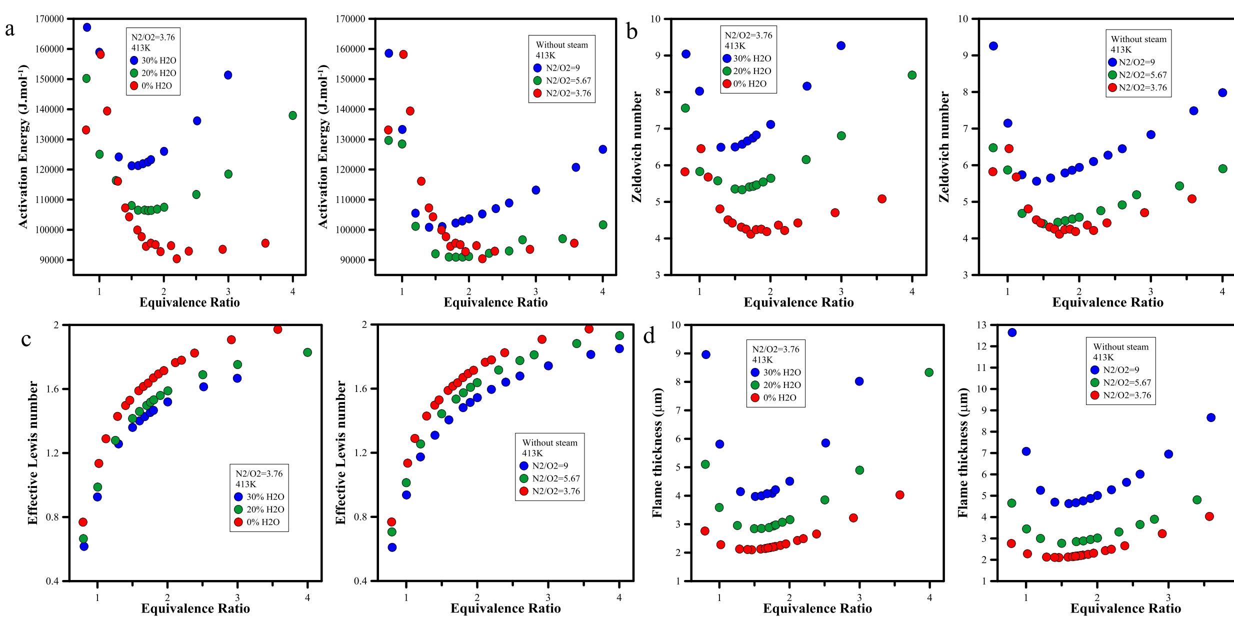

Figure 6. Fundamental combustion parameters of $H_2/N_2/O_2$ mixtures at 413K with and without steam (a) Activation Energy (b) Zeldovich number (c) Effective Lewis number (d) Flame thickness

4 Conclusion

Laminar flame speeds of $H_2/N_2/O_2$ mixtures have been studied in spherical bomb under a wide range of equivalence ratio φ ($0.8 \leq \varphi \leq 4$), initial temperature (296, 333, 363 and 413K) and N_2/O_2 ratio (3.76, 5.67, 9) at 100 kPa. The effect of steam addition in the mixtures was also investigated. Higher temperatures and ratio N_2/O_2 promote the faster flame propagation while steam addition has a suppressing effect. A very good agreement was found with data of Lamoureux et al. [7] and Tse et al. [10] for H_2/air mixtures at ambient temperature.

For the same mixtures and conditions, numerical calculations on laminar flame speeds S_L^0 were conducted in order to obtain the fundamental combustion parameters. The results presented here concerned the initial temperature 413K with and without steam. It has been shown the decrease of the mixture Lewis number and the increase of activation energy and flame thickness when dilution is raised (excepted between the ratios N_2/O_2=9 and N_2/O_2=5.67).

In this study, the measurements of the laminar flame speeds at 363K with a N_2/O_2 ratio of 3.76, and 333K with N_2/O_2 ratio of 5.67 and 9 were not conducted. The study will be completed in a near future.

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