UNSTEADY LUMPED-PARAMETER MODELING OF HYDROGEN COMBUSTION IN THE PRESENCE OF A WATER SPRAY

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ABSTRACT

In case of severe accidents in Pressurized Water Reactors, a great amount of hydrogen can be released, the resulting heterogeneous gaseous mixture (hydrogen-air-steam) can be flammable or inert and the pressure effects could alter the confinement of the reactor. Water spray systems have been designed in order to reduce overpressures in the containment, but the presence of water droplets could enhance flame propagation through turbulence or generate flammable mixtures since the steam present in the vessel could condense on the droplets and could not inert the mixture anymore. However beneficial effects would be heat sinks and homogenization of mixtures. On-going work is devoted to the modeling of the interaction between fine water droplets and a hydrogen-air flame. We present in this paper an unsteady Lumped Parameter model in detail with a special focus on hydrogen-air flame propagation in the presence of water droplets. The effects of the initial concentration of droplets, steam and hydrogen concentrations on flame propagation are discussed in the paper and a comparison between this model and our previous steady Lumped-Parameter model highlights the features of the unsteady approach. This physical model can serve as a validation tool for a CFD modeling. The results will be further validated against experimental data.

1. INTRODUCTION

The combustion of gaseous mixtures of hydrogen-air (diluted or not) has been the subject of many studies both in the case of laminar and turbulent regimes (see [1,2] and references therein). Yet in case where droplets of water are present no experimental data were found in the literature and the different mechanisms involved are not well described there [3]. Several studies have been devoted to the characterization of the flame propagation in the case of a homogeneous [4-7] or heterogeneous [8-9] hydrogen repartition in the confined vessel. The main mechanisms involved in a flame and a water spray interaction are identified: heat transfer between the flame front and the spray, momentum transfer (drag force, transport of droplets) and mass transfer (vaporization). The heat transfer mechanisms between a methane-air flame front and a water spray or mist have been fully described by Parra et al [5]. Besides the flame extinction mechanisms [6,3,7] and the mitigation effects of the spray [8-14] are characterized for methane- or propane-air flame. The fine analysis of the above topics is very limited in the literature and some discrepancies are observed between the experimental data and numerical results. The results of Proust [15] showed that the theories of Mitani [16] for a methane-air flame might be improved by integrating the following phenomena: the variation of the "thermal" parameters with the temperature, a more detailed chemical reaction mechanism, the incidence of the radiation by the burnt products and the influence of the water droplets size distribution. However the interaction between water droplets and a hydrogen-air flame inerted by steam is not well described. In the case of steam inerting air-hydrogen mixture, the presence of water droplets can generate flammable mixtures or enhance flame propagation through turbulence [12,17]. Yet beneficial effects would be heat sinks and homogenization of mixtures.

The present manuscript completes a previous steady Lumped-Parameter approach, that was only based on mass and energy transfers and aimed to obtain a good approximation of the thermodynamics aspects at final state [3]. That study had shown that liquid water generates heat sinks as expected but the final pressures can become higher than the Adiabatic Isochoric Complete Combustion (AICC) pressure (i.e. the maximum pressure without any droplets) when the initial hydrogen concentration attains a certain value. The steam due to the vaporization of droplets becomes important and makes the

global pressure increase. This paper is devoted to analyze the time evolution of the thermodynamic parameters and the influence of water droplets on the propagation of a hydrogen-air-water vapour flame. The effects of the spray characterization and the composition of the mixture are also studied.

2. PRESENTATION OF THE UNSTEADY LUMPED-PARAMETER MODEL

We consider a spherical propagation of the flame and the space is divided into two regions (Fig. 1): a burnt zone (subscript $_b$) and a fresh one (subscript $_f$).
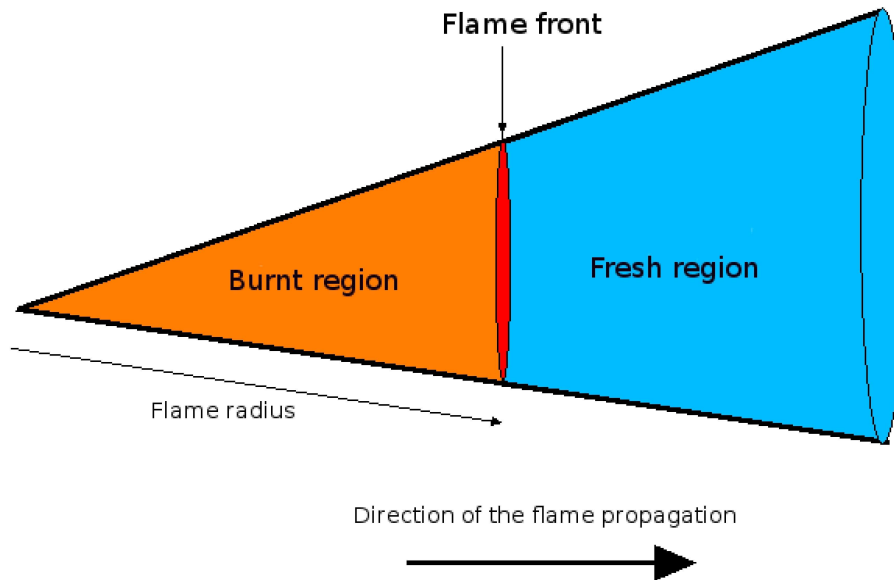

Figure 1. Schematic of the propagation of the flame front

The hypotheses that we make in the current work are:

a) There is no evaporation in the fresh zone because the time scale for the flame to cross the containment is much smaller than the time required scale to evaporate the droplets. We model droplets with a diameter smaller than 10^{-4} m (100 μm) and it is reasonable to consider that a drop completely evaporates as it crosses the flame ($r_{l,f}(t) = r_l(t=0)$ and $r_{l,b}(t) = 0$, where r_l is the mean droplet radius). Moreover, the time evolution of the droplet radius is difficult to establish in a Lumped-Parameter approach, because there is only one value of each variable in each cell at each time instant. In the burnt region, it is not possible to distinguish easily the sizes of the droplets far from the front from these near the front. This difficulty does not exist anymore in a CFD modeling.

b) The combustion reaction occurs at the temperature of the fresh gas $T_{g,f}$.

c) Conservation of mass and energy (impermeable and adiabatic walls).

d) The gases are considered as ideal gases.

e) The volume is constant.

f) The combustion is complete.

g) The whole heat Q liberated by the combustion vaporizes the liquid water and raises the temperature of the resulting gas mixture.

h) Air is considered as a binary mixture (N_2-O_2).

i) Hydrogen combustion is a single-step reaction.

j) The gaseous mixture and the liquid water are initially at 298.15 K and under atmospheric pressure.

The modeling of the propagation of a hydrogen-air flame under water spray conditions is described as followed:

1) The expansion of the flame front is defined by $\dfrac{dR_b}{dt} = \sigma S_L - \left(\dfrac{R_b}{3p} \dfrac{dp}{dt} \right)$ [18]

   where p is the pressure and σ the flame expansion factor defined by the density ratio of unburned gas $\rho_{g,f}$ to burned gas $\rho_{g,b}$ at two sides of the flame front: $\sigma = \dfrac{\rho_{g,f}}{\rho_{g,b}}$. S_L is the stretched fundamental flame velocity. It is constant in our study and fixed at 0.5 m.s^{-1} from experimental data for a mixture containing 15 mol% of hydrogen (see [19] and references therein). The pressure p is supposed uniform in both zones because there is no shock and the acoustic velocity is higher than 300 m.s^{-1} for hydrogen so that the pressure equalizes almost immediately in both regions.

2) The stretched spatial flame velocity u is equal to $u = \dfrac{dR_b}{dt}$.

3) We obtain the volume of the burnt zone $V_b(t) = \dfrac{4}{3} \pi R_b^3(t)$ and the volume of the fresh zone $V_f(t) = V_{tot} - V_b(t)$.

4) We calculate the number of the droplets in the fresh region $N_f(t) = N_0 \dfrac{V_f(t)}{V_{tot}}$.

5) We compute the fresh hydrogen mass $m_{H_2,f}(t) = V_{g,f} \rho_{H_2}(t)$ where $\rho_{H_2} = \dfrac{X_{H_2} p}{R/W_{H_2} T_{g,f}}$.

6) We define the reaction progress variable $\xi(t)$ by: $\dfrac{m_{H_2,f}(t=0) - m_{H_2,f}(t)}{m_{H_2,f}(t=0)}$.

7) We compute the gas temperature by taking into account the exchange with the liquid water in the fresh zone: $\dfrac{d\left((1-\alpha_l)c_V T_{g,f}\right)}{dt} = -\alpha_l H(T_{g,f} - T_l)$, where α_l is the volume fraction of droplets, c_V the heat capacity of fresh gases, $T_{g,f}$ the temperature in fresh gases, T_l the temperature of droplets (constant since there is no evaporation) and H the exchange coefficient in fresh gases ($H = \lambda \text{Nu}/(2 r_l^0)$), where λ is the thermal conductivity of fresh gases, Nu is the Nusselt number equal to 2 in a Lumped-Parameter approach and r_l^0 the

constant droplet radius in the fresh gases). We also consider the exchange between the fresh zone and the burnt one by calculating an isentropic equation of state (adiabatic flame) and neglecting radiation and conduction phenomena: $(1-\gamma(T_{g,f}))\dfrac{d\rho_{g,f}}{\rho_{g,f}}+\dfrac{dT_{g,f}}{T_{g,f}}=0$, where $\gamma(T_{g,f})$ is the heat capacity ratio and $\rho_{g,f}$ is the density in the fresh zone.

8) We calculate the number of moles for each specie (hydrogen, oxygen, steam and nitrogen) in the whole containment (both zones):

$n_{H_2}(t)=(1-\xi(t))n_{H_2}(t=0)$,

$n_{O_2}(t)=n_{O_2}(t=0)-\dfrac{1}{2}\xi(t)n_{H_2}(t=0)$,

$n_{H_2O}(t)=n_{H_2O}(t=0)+\xi(t)n_{H_2}(t=0)+\dfrac{m_l(t=0)-m_l(t)}{W_{H_2O}}$, where

$m_l(t)=N_f(t)\dfrac{4}{3}\pi r_l^3(t=0)\rho_l$ and W_{H_2O} is the water molar mass, $n_{N_2}(t)=n_{N_2}(t=0)$.

9) We compute the heat Q released by the combustion reaction:

$Q(t)=\dfrac{m_{H_{2,f}}(t=0)-m_{H_{2,f}}(t)}{W_{H_2}}\Delta_r H_{T_0}^o$ where $\Delta_r H_{T_0}^o$ = -241.8 kJ.mol^{-1} is the formation enthalpy associated with the hydrogen combustion reaction.

10) We solve the following equation and stop the calculation when the combustion is complete i.e. all hydrogen is consumed ($\xi=1$): $Q-m_{evap}L_{vap}+\int_{T_0}^{T_{g,b}}\sum_j n_j c_{p,j}(\tau)d\tau=0$ where m_{evap} is the vaporized liquid mass, L_{vap} is the latent heat, $c_{p,j}$ is the heat capacity of the j-specie (J.mol^{-1}.K^{-1}) and n_j is the number of moles of the j-specie, in order to determine the temperature in the burnt region.

11) We deduce the pressure p by the ideal gas relation: $p(t)=\rho_{g,b}(t)\dfrac{R}{W_{g,b}}T_{g,b}(t)=\rho_{g,f}(t)\dfrac{R}{W_{g,f}}T_{g,f}(t)$, where the density in burnt gases $\rho_{g,b}(t)$ is

given by $\rho_{g,b}(t)=\dfrac{\rho_{g,f}(t)W_{g,b}T_{g,f}(t)}{W_{g,f}T_{g,b}(t)}$. $W_{g,f}$ and $W_{g,b}$ refer to the molar masses of the fresh and the burnt gases respectively.

3. RESULTS AND DISCUSSIONS

3.1 Time evolutions of pressure and temperature during the flame propagation

The initial conditions corresponding to the following results are summarized in the Table 1. In Fig. 2, the time evolution of the hydrogen mass is shown. Fig. 3 gives the time evolution of the temperature in fresh gases. We observe an increase of 60% (about 180 K). Fig. 4 and 5 compare the time evolutions of the temperature in burnt gases and the pressure respectively with the final values of the temperature and the pressure found with the previous steady Lumped-Parameter model [3]. We first observe that the maxima of temperature and pressure are attained only at the end of the combustion and not during the hydrogen consumption. Besides, we note that the final values of temperature and pressure found

with the unsteady Lumped-Parameter model, described above, coincide with those found with the steady Lumped-Parameter version.

Table 1. Default initial conditions

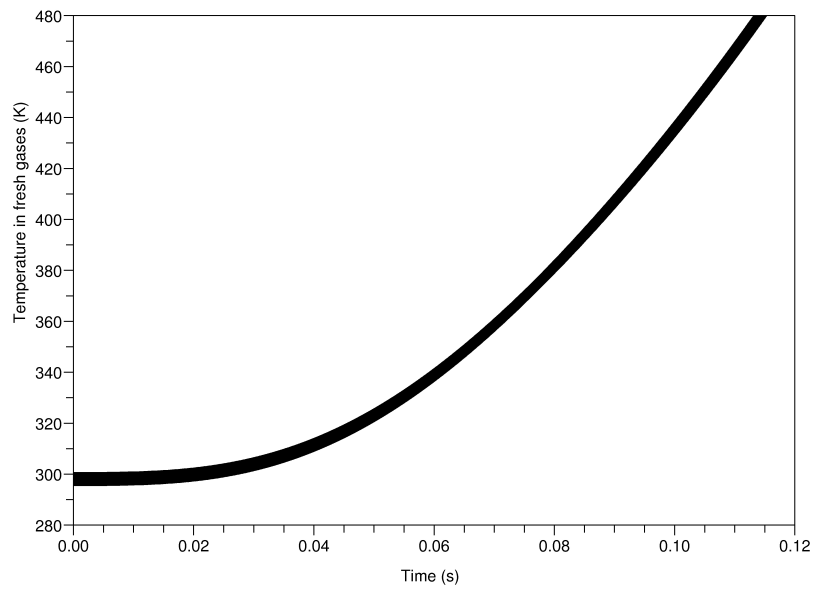| Names	Values	Units
Pressure	101325	Pa
Gas temperature	298.15	K
Liquid temperature	298.15	K
Liquid volume fraction	$5.\ 10^{-5}$	-
Mean droplet radius	5.10^{-5}	m
Hydrogen mole fraction	0.15	-
Steam mole fraction	0	-
Fundamental stretched velocity	0.50	$m.s^{-1}$
Total radius of the containment	0.25	m
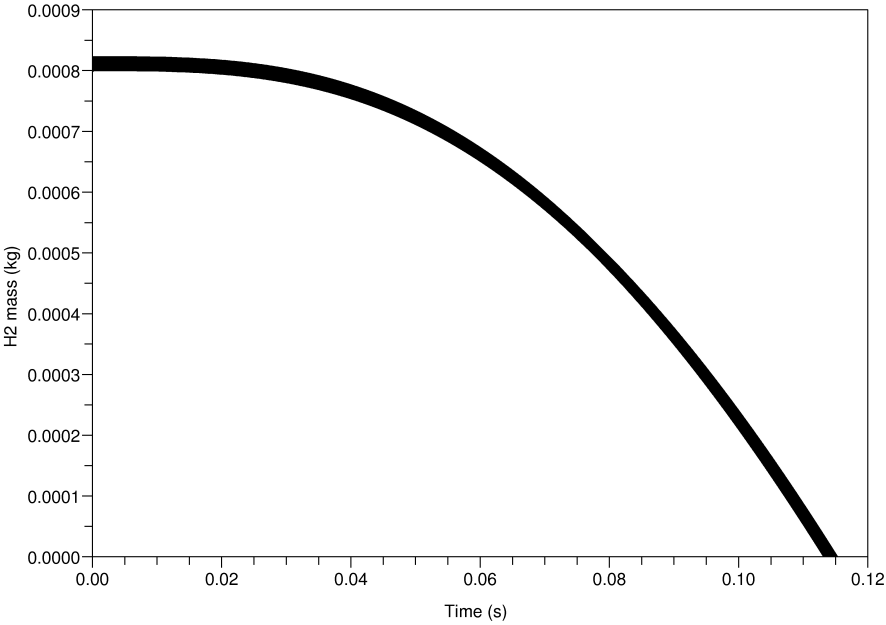
Figure 2.Time evolution of the hydrogen mass.

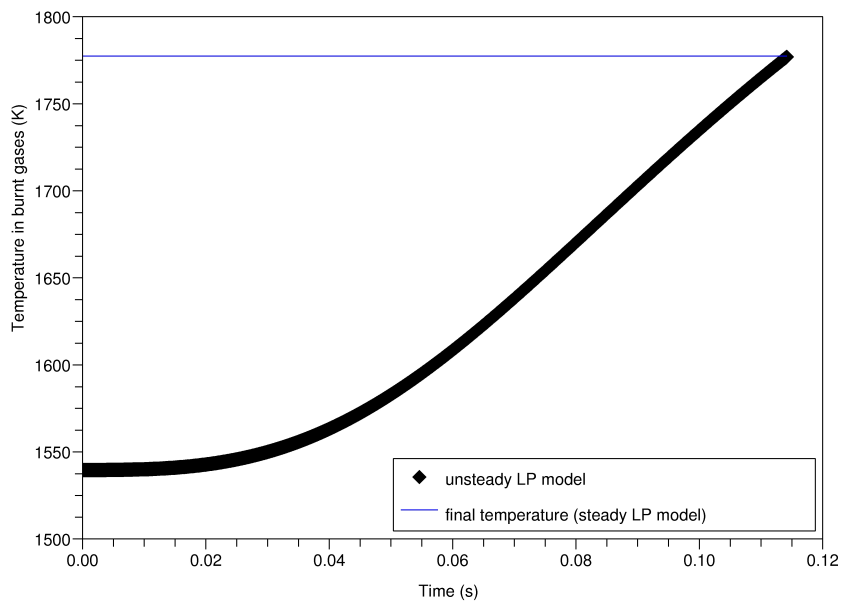Figure 3.Time evolution of the temperature of the fresh gases.

Figure 4. Time evolution of the temperature of the burnt gases and comparison of the final temperature with the final one found with the steady Lumped-Parameter model.
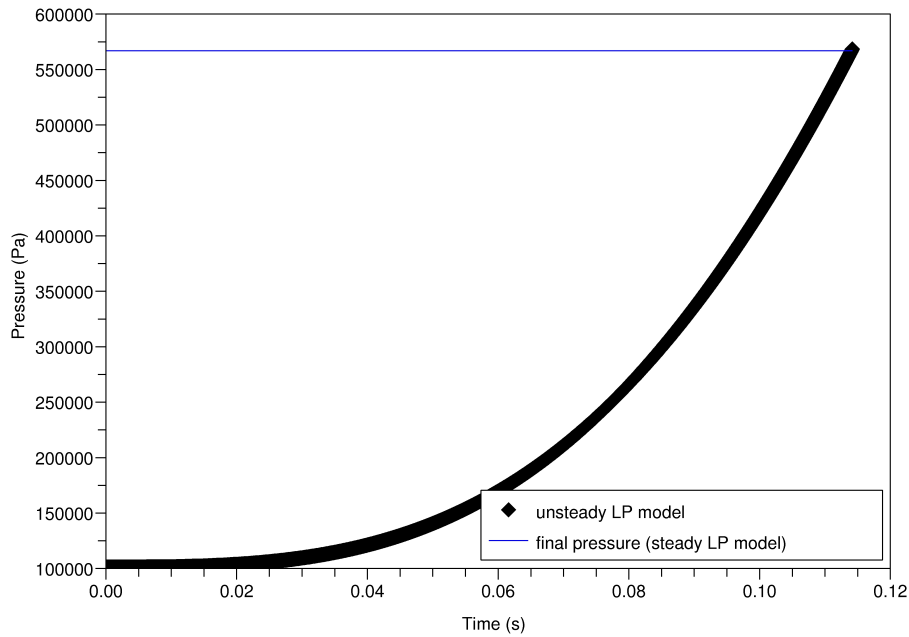
Figure 5. Time evolution of the pressure and comparison of the final pressure with the final one found with the steady Lumped-Parameter model.

3.2 Effects of some parameters on the stretched spatial flame velocity

The flame propagation can be influenced by various factors under water spray conditions: the droplet radius, the droplet volume fraction and the gaseous composition of the mixture. The hypotheses of the Lumped-Parameter approach allow us to study the impact of the droplet concentration and the gaseous composition of the mixture on the flame propagation but not the impact of the droplet size. We have performed parametric studies changing only one parameter for each study. The main mechanism of the flame velocity increase/decrease is through variations in the flame expansion factor σ and the pressure p (see the first step of our modeling [18]).

Fig. 6 to 9 show the effects of the initial hydrogen mole fraction, steam mole fraction and droplet volume fraction α_l on the stretched spatial flame velocity u. In Fig. 6, the influence of the hydrogen concentration on the time evolution of u is studied for a poor mixture (10 and 20 mol% hydrogen). The richer the gaseous mixture is initially, the faster the stretched spatial flame velocity decreases under water spray conditions: the global percentage decrease is 23% at 10%mol of hydrogen and 35% at 20%mol. The Fig. 7 shows that the presence of water vapor in the containment before combustion makes the stretched spatial flame velocity u decrease (about 11% at the beginning of the combustion with 20%mol of steam and the global reduction of the flame velocity is about 40%). In Fig. 8, the flame velocity declines when the droplet concentration is increased for a given initial droplet radius (between 5 and 7 % with an initial droplet volume fraction α_l from 10^{-5} to 6.10^{-5} and the global decrease is about 38%). In Fig. 9, the influence of the initial droplet volume fraction on the flame expansion is studied. The larger the droplet volume fraction α_l is initially, the faster the flame expansion factor σ is reduced (when α_l varies from 10^{-5} to 6.10^{-5}, σ diminishes between 9 and 11% and the global decrease is between 34 and 39%).

We note that all these results were obtained under hypothesis of a constant fundamental stretched flame velocity S_L, fixed from experimental data [19]. The flame velocity would actually strongly

depend on the level of turbulence generated by flame-droplet interaction, which in turns depends on droplet diameter. This is the subject of current CFD studies.
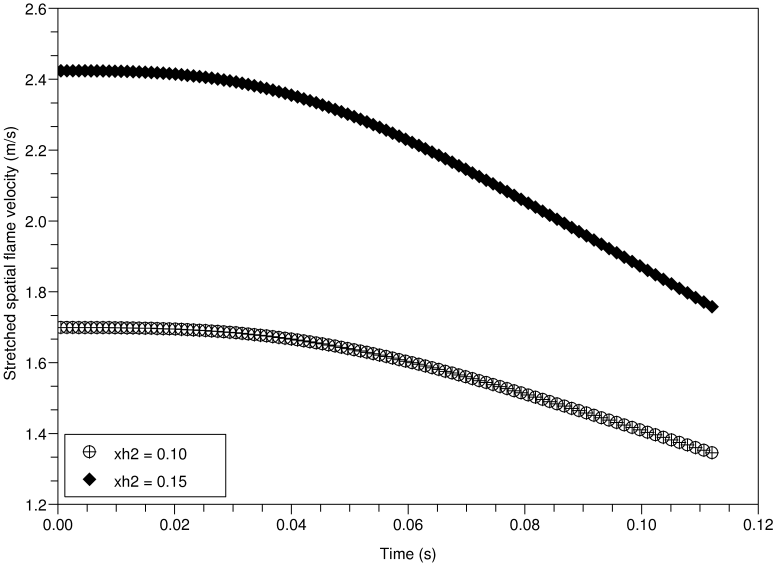

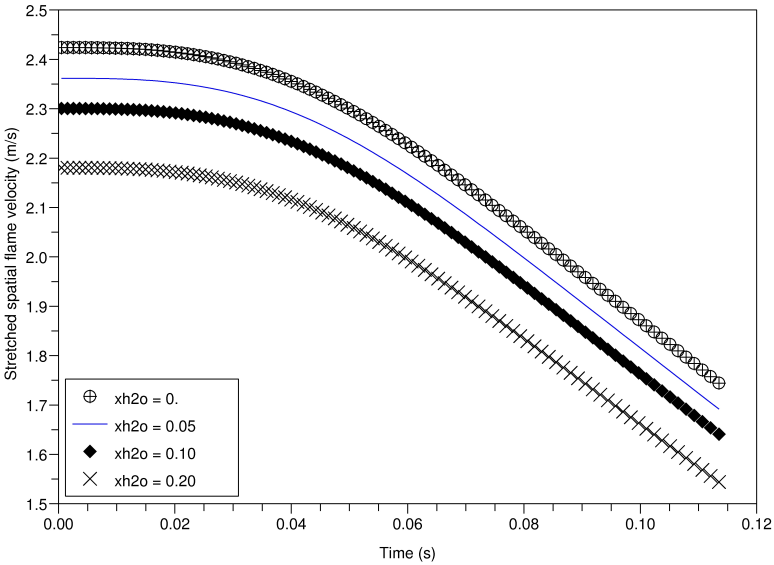Figure 6.Time evolution of the stretched spatial flame velocity u under various initial hydrogen mole fractions.

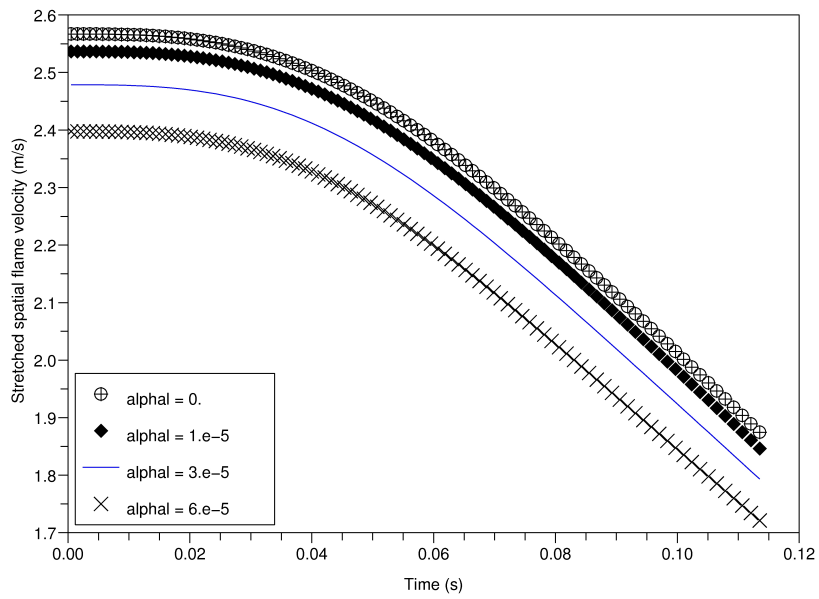Figure 7.Time evolution of the stretched spatial flame velocity u under various initial steam mole fractions.

Figure 8.Time evolution of the stretched spatial flame velocity *u* under various initial liquid water volume fractions α_l.
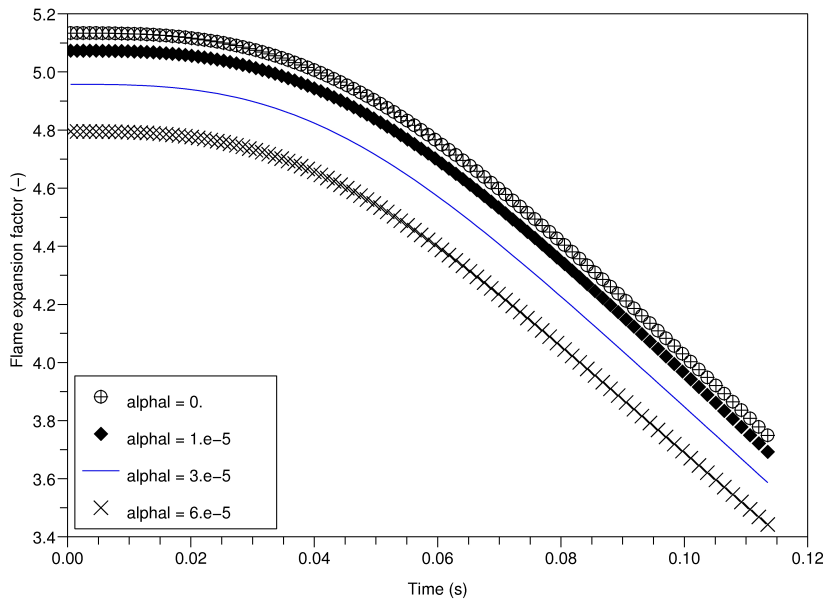

Figure 9.Time evolution of the flame expansion factor σ (density ratio of fresh gas to burnt gas) under various initial liquid water volume fractions α_l.

4. CONCLUDING REMARKS

The unsteady Lumped-Parameter study is an additional step to the full CFD modeling. The previous steady Lumped-Parameter approach had demonstrated that liquid water generates heat sinks as expected but the final pressures can become quite important when the initial hydrogen concentration attains a certain value. This present analysis was devoted to obtain results on the dynamics of this phenomenon and the effects of the initial conditions on the flame propagation (stretched spatial flame velocity, flame expansion factor and combustion time).

We note that:

- The maxima of temperature and pressure are attained at the end of the combustion and not during the process. The steady Lumped-Parameter model is adequate for estimations of the maxima of both parameters in the study of hydrogen combustion under water spray conditions.

- Increasing the concentration of droplets has beneficial effects on the flame expansion factor since it is reduced when the initial droplet volume fraction is increased.

- The flame propagation is influenced by the concentration of the droplets in the mixture and not at all by the droplet sizes under the hypotheses that we made. The spatial stretched flame velocity decreases when the initial volume fraction of droplets α_l increases.

- The beneficial effects of water droplets on the flame velocity are amplified when the gaseous mixture is richer in hydrogen and/or when the initial amount of steam is higher.

A CFD work is currently in progress in order to complete our study on the effects of water spray systems in case of accidental production of hydrogen in Pressurized Water Reactor. It will improve our knowledge in the influence of the droplet sizes and velocities on the propagation of a hydrogen-air flame inerted by steam.

REFERENCES

1. Kitagawa, T., Nakahara, T., Maruyama, K., Kado, K., Hayakawa, A., Kobayashi, S. Turbulent burning velocity of hydrogen-air premixed propagating flames at elevated pressures. *International Journal of Hydrogen Energy*, **33**, No. 20, 2008, pp. 5842-5849.
2. Tang, C., Huang, Z., Jin, C., He, J., Wang, J., Wang, X., Miao, H. Explosion characteristics of hydrogen-nitrogen-air mixtures at elevated pressures and temperatures. *International Journal of Hydrogen Energy*, **34**, No. 1, 2009, pp. 554-561.
3. Joseph-Auguste, C., Cheikhravat, H., Djebaïli-Chaumeix, N. and Deri, E., On the use of spray systems: an example of R&D work in hydrogen safety for nuclear applications. *International Journal of Hydrogen Energy*, 2009, doi 10.1016/jhydene.2009.01.018 (corrected proofs).
4. Kudriakov, S., Dabbene, F., Studer, E., Beccantini, A., Magnaud, J.-P., Paillère, H., Bentaïb, A., Bleyer, A., Malet, J., Porcheron, E., Caroli, C. The TONUS CFD code for hydrogen risk analysis: Physical models, numerical schemes and validation matrix. *Nuclear Engineering and Design*; **238**, No. 3, 2008, pp. 551-565.
5. Parra, T., Castro, F., Méndez, C., Villafruela, J.M., Rodrìguez, M.A. Extinction of premixed methane-air flames by water mist. *Fire Safety Journal,* **39**, 2004, pp. 581–600.
6. Prasad, K., Li, C., Kailasanath, K. Simulation of water mist suppression of small scale methanol

liquid pool fires. *Fire Safety Journal*, **33,** 1999, pp. 185–212.

7. Downie, B., Polymeropoulos, C., Gogos, G. Interaction of a water Mist with a Buoyant Methane Diffusion Flame. *Fire Safety Journal,* **24,** 1995, pp. 359–381.

8. Modak, A.U., Abbud-Madrid, A., Delplanque, J.-P., Kee, R.J. The Effect of monodispersed water mist on the suppression of laminar premixed hydrogen-, methane-, and propane-air flames. *Combustion and Flame*, **144**, 2006, pp. 103–111.

9. Prasad, K., Patnaik, G., Kailasanath, K. A numerical study of water-mist suppression of large scale compartment fires. *Fire Safety Journal*, **37**, 2002, pp. 569–589.

10. Jones, S.J., Averill, A.F., Ingram, J.M., Holborn, P.G., Battersby, P., Nolan, P.F., Kempsell, I.D., Wakem, M.J. Mitigation of hydrogen-air explosions using fine water mist sprays. Symposium series No. 151. IChemE, 2006.

11. Yang, W., Kee, R.J.. The Effect of Monodispersed WaterMists on the Structure, Burning velocity, and Extinction behavior of Freely Propagating, Stoichiometric, Premixed, methane-Air Flames. *Combustion and Flame*, **130**, 2002, pp. 322–335.

12. Thomas, G.O. On the conditions required for explosion mitigation by water sprays. *Trans I Chem E: Part B - Process Safety and Environmental Protection*, **78**, 2000, pp. 339–354.

13. van Wingerden, K., Wilkins, B. The influence of water sprays on gas explosions. Part II: mitigation. *Journal of Loss Prevention in the Process Industries*, **8**, No. 2, 1995, pp. 61–70.

14. Lentati, A.M., Chelliah, H.K. Dynamics of Water Droplets in a Counterflow Field and their Effect on Flame Extinction. *Combustion and Flame*, **115**, 1998, pp. 158–179.

15. Proust, C. Flame-water droplets interactions. 1996.

16. Mitani, T. A flame inhibition theory by inert dust and spray. *Combustion and Flame,* **43**, 1981, pp. 243-253.

17. van Wingerden, K., Wilkins, B. The influence of water sprays on gas explosions. Part I: water spray-generated turbulence. *Journal of Loss Prevention in the Process Industries*; **8**, No. 2, 1995, pp. 53–59.

18. Eschenbach, R.C. and Agnew, J.T. *Combustion and Flame,* **2**, 1958, pp. 273–285.

19. Huang, Z., Zhang, Y., Zeng, K., Liu, B., Wang, Q., Jiang, D. Measurements of laminar burning velocities for natural gas-hydrogen-air mixtures. *Combustion and Flame*, **146**, No. 1-2, 2006, pp. 302-311.