ON THE USE OF SPRAY SYSTEMS: AN EXAMPLE OF R&D WORK IN HYDROGEN SAFETY FOR NUCLEAR APPLICATIONS

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ABSTRACT

The aim of the present work is to investigate the interaction between a water spray and a laminar hydrogen-air flame in the case of steam inerted mixture. A first work is devoted to study the thermodynamics involved in the phenomena via a lumped parameter code. The flow is two-phase and reactive, the gas is multi-component, the water spray is polydisperse and the droplet size has certainly an influence on the flame propagation. The energy released by the reaction between hydrogen and oxygen vaporizes suspended droplets. The next step of this study will be to consider a drift-flux model for the droplets and air under hypotheses that the velocity and thermal disequilibria are weak. The multi-component feature of the gas will be further taken into account by studying a gas mixture containing hydrogen, air and water vapor. A second study concerns an experimental investigation of the effect of droplets on the flame propagation using a spherical vessel. A Schlieren system is coupled to the spherical vessel in order to record the flame propagation on a digital high speed camera. Both studies will help improve our knowledge of safety relevant phenomena.

NOMENCLATURE

Symbols	Descriptions	Units
f_V	Volume fraction of liquid water	—
h_0^s	Sensible enthalpy	J.kg−1
Δh_0	Formation mass enthalpy at the reference temperature T = 0 K	J.kg−1
m	Mass	kg
n	Number of moles	mol
p	Pressure	Pa
T	Temperature	K
R_j	j - gas constant	J.kg−1.K−1
R	Perfect gas constant (= 8.313)	J.mol−1.K−1
S	Number of species in the gaseous phase	—
u	Velocity	m.s−1
v	Diffusion velocity	m.s−1
x_j	Molar fraction of the j species	—
y_j	Mass fraction of the j species	—
W	Molar mass	kg.mol−1
Greek		
ε	Specific internal energy	J.kg−1
ρ	Density	kg.m−3

Subscripts		
air	Refers to air	—
j	Refers to j-species	—
vap	Refers to vapor	—
liq	Refers to liquid water	—
gas	Refers to gas	—
init	Refers to initial state	—
fin	Refers to final state	—
tot	Refers to total quantity	—
H2	Refers to hydrogen	—
H2O	Refers to steam	—
N2	Refers to nitrogen	—
O2	Refers to oxygen	—
Superscripts		
init	Refers to initial	—
fin	Refers to final	—
comb	Refers to combustion	—
evap	Refers to vaporization	—
AICC	Refers to Adiabatic Isochoric Complete Combustion states	—

INTRODUCTION

A considerable effort in hydrogen safety R&D has occurred since the Three Mile Island accident in 1979 through experimental programs and computational tools development for a better understanding of hydrogen release, distribution and possible combustion regimes. As a result of this work, different types of mitigation systems such as recombiners, igniters and spray systems have been designed and installed in modern nuclear power plants. Mitigation systems related to hydrogen safety in nuclear power plants have been the subject of several experimental and theoretical/computational studies in the past. In French Pressurized Water Reactors, water spray systems have been designed in order to reduce overpressures in the containment and to remove from the atmosphere of this containment any scattered radioactive aerosols. Although the presence of a cloud of water droplets can generate flammable mixtures or would enhance flame propagation through turbulence in the case of steam inerted air-hydrogen mixtures, beneficial effects would be heat sinks and mixtures homogenization. The combustion of gaseous mixtures of hydrogen-air diluted or not has been the subject of several studies both in the case of laminar or turbulent regimes. However, 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 in the literature. In this work, two objectives are defined, the first one is to study the thermodynamics aspects via a lumped parameter (LP) analysis and the interaction of a laminar premixed air-hydrogen-steam flame with a water mist via a CFD analysis. Only the LP analysis is described in this paper. The numerical results are obtained with Cast3M, a LP and CFD code developed at the French Atomic Energy Commission (CEA). This code solves the differential partial equations of fluid and solid mechanics using finite element and finite volume methods. The second one is to investigate experimentally the interaction of the flame with water droplets in the case where the flame is initially laminar. The effect of the liquid water volume fraction and size distribution on the propagation of the flame will be investigated.

1 LUMPED PARAMETER (LP) ANALYSIS: HYDROGEN COMBUSTION UNDER LIQUID WATER CONDITIONS

The main mechanisms involved between a flame and a water spray 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 [1]. Besides the flame extinction mechanisms [2,1,3] and the mitigation effects of the spray [4,5,6,7,8,9,10] 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 [11] showed that the theories of Mitani [12] for a methane-air flame might be improved by integrating: 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 known. In the case of steam inerted air-hydrogen mixture, the presence of water droplets can generate flammable mixtures or enhance flame propagation through turbulence [8,13]. Yet beneficial effects would be heat sinks and homogenization of mixtures.

1.1 Lumped Parameter code

A Lumped Parameter model has been developed in the Cast3M code in order to study the thermodynamic aspects involved in the interaction between water droplets and a hydrogen-air flame. We are only interested in the final state in this work. That is why the chemical kinetics is not accounted for and the code does not calculate any reaction rates. The LP model code allows us to know the final states simply and quickly. The results will be used to validate the equilibrium states of our future CFD analysis. We consider an air-hydrogen-steam mixture and an amount of liquid water in a confined space. At this stage, the gases and droplets speeds and the sizes of droplets are not taken into account. The energy released by the reaction air-hydrogen vaporizes the liquid water.

The analysis is performed using an incomplete combustion hypothesis by considering the ratio of burned hydrogen number of moles to available hydrogen number of moles as progress variable. In this paper, however, we only present the case of complete combustion. We can also study a semi-confined space by adding the heat transfers with the outside space.

The hypotheses we assumed in this work are summarized in figure 1: 1) conservation of mass and energy (impermeable and adiabatic walls), 2) the gases are considered as ideal gases, 3) the volume is constant, 4) the combustion is complete, 5) the whole energy liberated by the combustion vaporizes the liquid water, 6) air is considered as a binary mixture (N2-O2), 7) hydrogen combustion is a single-step reaction and 8) the gaseous mixture and the liquid water have initially the same temperature T_0. We compute the final species number of moles using the initial data and the hypotheses.

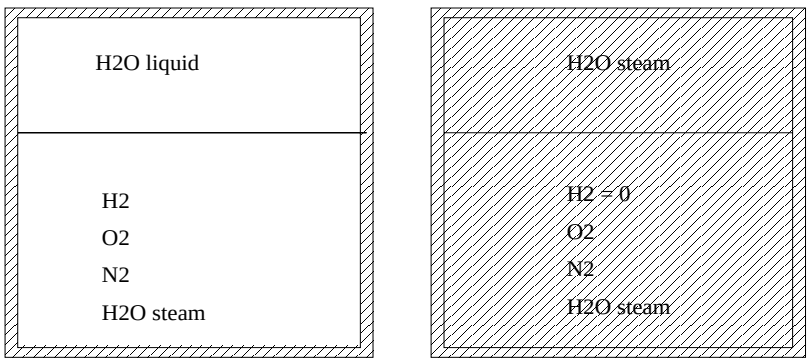

Figure 1. The species before and after combustion

As we assume that the combustion reaction is complete and single-step and the mixture is lean or

stoichiometric, $n_{H_2}^{fin} = 0$, $n_{H_2O^{comb}}^{fin} = n_{H_2}^{ini} + n_{H_2O_{vap}}^{ini}$, $n_{O_2}^{fin} = n_{O_2}^{ini} - (\frac{1}{2} n_{H_2}^{ini})$ and $n_{N_2}^{fin} = n_{N_2}^{ini}$. Besides, the whole liquid water is supposed to evaporate so that $n_{H_2O^{evap}}^{fin} = n_{H_2O_{liq}}^{ini}$. The initial number of moles of liquid water is defined by: $n_{H_2O_{liq}}^{ini} = \frac{\rho_{H_2O} \cdot f_V \cdot V_{tot}}{W_{H_2O}}$ where f_V is the initial volume fraction of liquid water. The energy released by the air-hydrogen reaction is computed usng the CHEMKIN coefficients [14]. The initial values for temperature T_0, pressure p_0 and steam molar fraction $x_{H_2}^{init}$ are equal to 298 K, 1 atm (1.013 10^5 Pa) and zero, respectively.

The computation is divided into several steps:

1. calculation of amount of energy released due to complete combustion
$$f_{gas}(T_{fin}) = -(h_0^s(T_0) - R^{init} \cdot T_0 + \Delta h_0^{init}) + (h_0^s(T_{fin}) - R^{fin} \cdot T_{fin} + \Delta h_0^{fin})$$
$$= \Delta(\frac{m_{air}}{m_{tot}} \varepsilon_{air} + \frac{m_{H2}}{m_{tot}} \varepsilon_{H2} + \frac{m_{vap}}{m_{tot}} \varepsilon_{vap})$$
where $R^{init} = \sum_j R_j y_j^{init}$, $R^{fin} = \sum_j R_j y_j^{fin}$ (the steam provided by the vaporization of liquid water is not taken into account here), R_j is the ideal gas constant for the j species, m the mass, ε the internal energy, h_0^s the mixture sensible enthalpy (J.kg−1) and Δh_0 the formation enthalpy ;

2. calculation of amount of energy necessary to heat and evaporate liquid water and heat steam
$$f_{liq}(T_{fin}) = \frac{m_{liq}}{m_{tot}} \cdot \left([\varepsilon_{liq}(100, p_0) - \varepsilon_{liq}(T_0, p_0)] + [\varepsilon_{vap}(100) - \varepsilon_{liq}(100, p_0)] + [\varepsilon_{vap}(T_{fin}) - \varepsilon_{vap}(100)] \right)$$

3. under the hypothesis of final thermal equilibrium, we solve the polynomial equation $f_{gas}(T_{fin}) = f_{liq}(T_{fin})$ with the final temperature T_{fin} as unknown by a Bairstow algorithm;

4. calculation of the final density $\rho_{fin} = \sum_{j=1}^{S} n_j W_j / V_{tot}$, with n_j the number of moles for the j specie, V_{tot} the total volume and S the number of species in the gaseous phase;

5. calculation of the final pressure p_{fin} from the ideal gas relation: $p_{fin} = \rho_{fin} \frac{R}{W} T_{fin}$.

1.2 Results of the LP model

First of all, we shall limit some of our parameters in order to have physically meaningful results i.e. a) we take a value for the initial hydrogen molar fraction $x_{H_2}^{init}$ inside the flammability limits and b) we take a value for f_V less or equal to 10^{-3} (a value of f_V higher or equal to 10^{-2} would correspond to more than 10 kg of liquid water for 1 kg of ambient gases).

In figure 2, the evolutions of final temperature T_{fin} as a function of initial molar fraction $x_{H_2}^{init}$ (taken between 8% and 25%) are shown for different values of f_V . Let us make several observations: 1) in the absence of the liquid water (f_V = 0), the final temperature corresponds to the Adiabatic Isochoric Complete Combustion (AICC) temperature, as it should; 2) for a given $x_{H_2}^{init}$ the temperature decreases with increasing f_V i.e. more energy is spent into heating and evaporating of bigger amount of liquid water; 3) at fixed temperature, $x_{H_2}^{init}$ increases with f_V , as expected, because the energy released by the combustion gets higher with $x_{H_2}^{init}$ and is able to vaporize a greater and greater amount of liquid water and 4) the temperature increases linearly with $x_{H_2}^{init}$ and the decrease of temperature between the case where there is no liquid water at the beginning and the case where f_V = 10^{-4} is about 340 K.

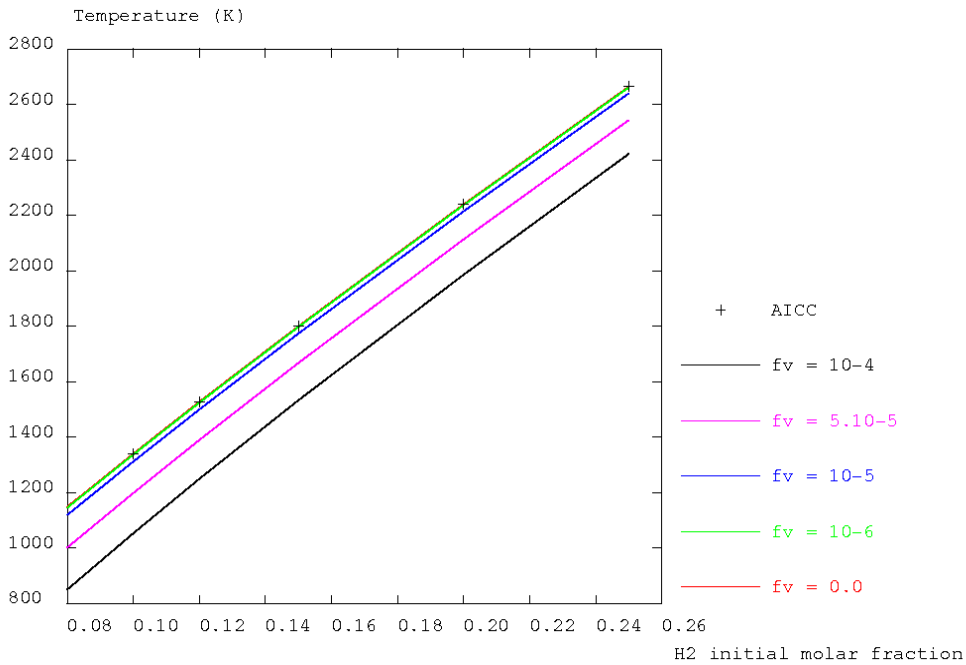

Figure 2. Evolutions of final temperatures as a function of the molar fraction $x_{H_2}^{init}$ for different initial volume fractions of liquid water f_V

The evolution of the final pressure (figure 3) is nevertheless unexpected. When $x_{H_2}^{init}$ increases from 8% to 18%, the increase of pressure is similar to the temperature one but is not linear. We observe that all pressure curves join in a zone that corresponds to $x_{H_2}^{init}$ between 19% and 21%. Beyond this critical zone the pressures are higher than the AICC pressure. So we assume that the steam pressure due to vaporization becomes important as $x_{H_2}^{init}$ increases. In the critical zone, the corresponding final pressure is nearly equal to the AICC pressure. We have for all f_V :

$$p_{AICC} \approx p_{f_V}$$

i.e. $p_{H2}^{AICC} + p_{O2}^{AICC} + p_{N2}^{AICC} + p_{H2O^{comb}}^{AICC} \approx p_{H2}^{f_v} + p_{O2}^{f_v} + p_{N2}^{f_v} + p_{H2O^{comb}}^{f_v} + p_{H2O^{evap}}^{f_v}$, then

$p_{tot}(x_{H2}^{AICC} + x_{O2}^{AICC} + x_{N2}^{AICC} + x_{H2O^{comb}}^{AICC}) \approx p_{tot}(x_{H2}^{f_v} + x_{O2}^{f_v} + x_{N2}^{f_v} + x_{H2O^{comb}}^{f_v} + x_{H2O^{evap}}^{f_v})$ hence

$A = (x_{H2}^{AICC} - x_{H2}^{f_v}) + (x_{O2}^{AICC} - x_{O2}^{f_v}) + (x_{N2}^{AICC} - x_{N2}^{f_v}) + (x_{H2O^{comb}}^{AICC} - x_{H2O^{comb}}^{f_v}) \approx x_{H2O^{evap}}^{f_v}$.

The table 1 compares the molar fractions of the involved species for the AICC and the f_V = 10^{-4} case, for two initial molar fractions for hydrogen of 18% and 22 %. This table shows that for $x_{H_2}^{init}$ = 18%, the difference between the vaporized molar fraction $x_{H2O^{evap}}^{f_v=10-4}$ and the term A is negative and equal to -2.3 10^{-6}. For $x_{H_2}^{init}$ = 22%, this difference is now positive and equal to 6.10^{-7}. This proves that the steam pressure due to vaporization becomes important when $x_{H_2}^{init}$ is larger than some critical value.

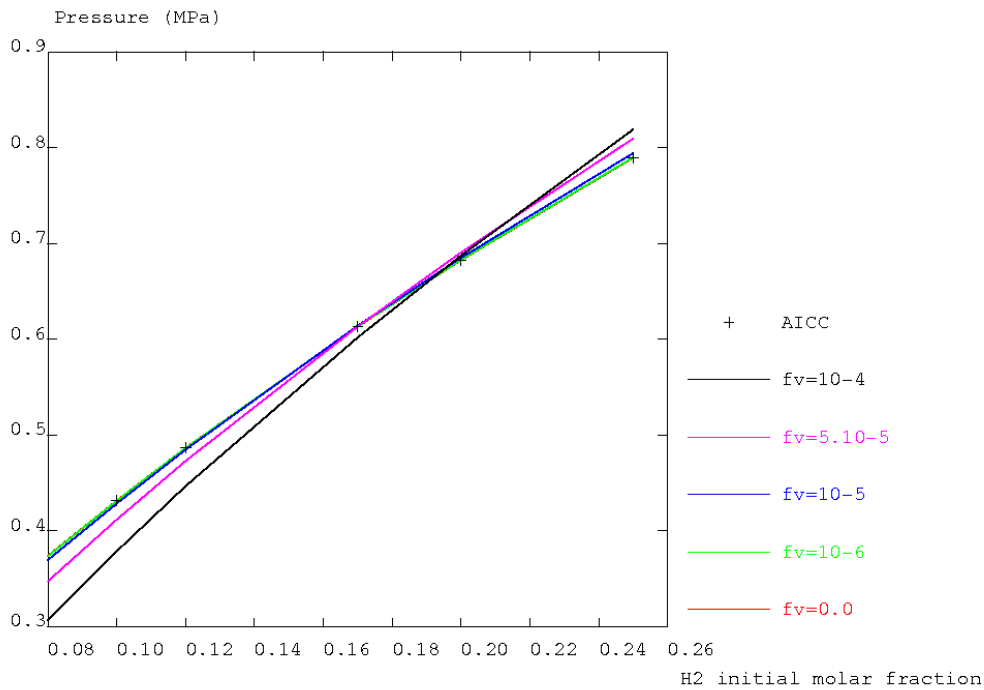

Figure 3. Evolutions of final pressures as a function of the molar fraction $x_{H_2}^{init}$ for different initial volume fractions of liquid water f_V

Table 1. Comparison of the molar fractions of species after combustion for $x_{H_2}^{init}$ =18% and $x_{H_2}^{init}$ = 22%

	($x_{H_2}^{init}$ =18%)	AICC	f_V = 10^{-4}	($x_{H_2}^{init}$ =22%)	AICC	f_V = 10^{-4}
$x_{N_2}^{fin}$		0.712	0.620		0.692	0.601
$x_{O_2}^{fin}$		0.090	0.079		0.061	0.052
$x_{H_2O^{comb}}^{fin}$		0.198	0.172		0.247	0.215
$x_{H_2}^{fin}$		0.0	0.129		0.0	0.132

The final density (figure 4) as a function of $x_{H_2}^{init}$ decreases: 1) as f_V increases for a fixed value of $x_{H_2}^{init}$, the final density ρ_{fin} increases and 2) for fixed f_V and increasing $x_{H_2}^{init}$, we expect that ρ_{fin} decreases.

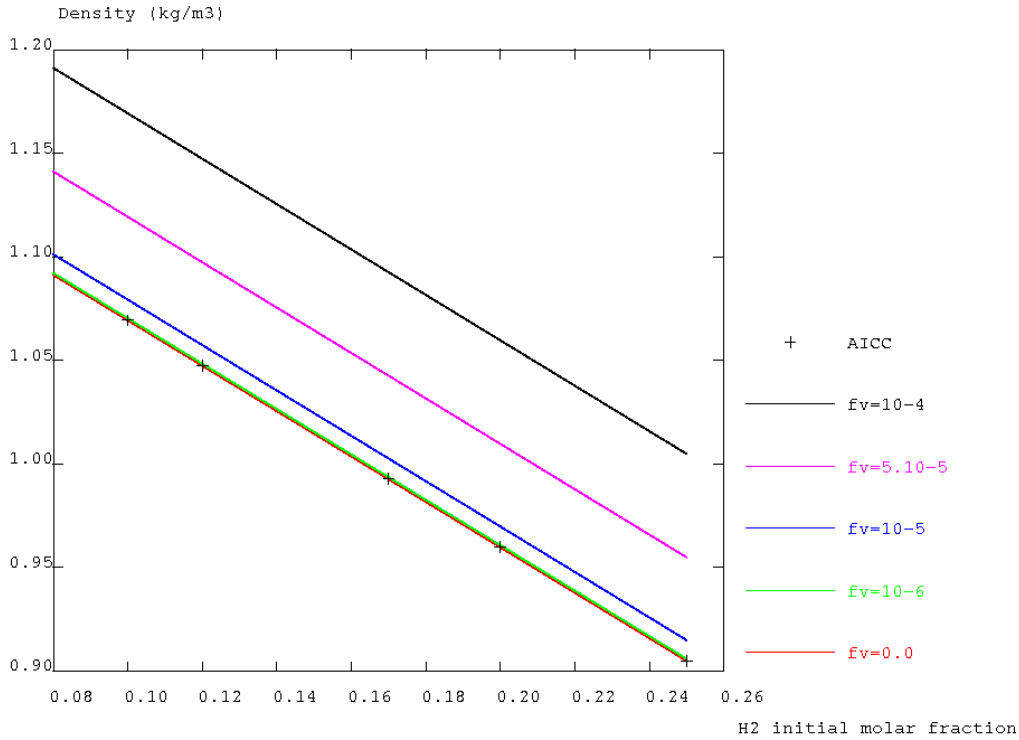

Figure 4. Evolution of the final density as a function of x_{init}^{H2} for different initial volume fractions of liquid water f_V

This first study improves our knowledge about the thermodynamic effects involved during the interaction of a water spray and a hydrogen-air flame. A CFD study is the next step of this work and is in progress. We can assume that a mist can be defined as a cloud composed of suspended droplets and gases. We can do two hypotheses in order to model the physical problem: the velocity difference between the two phases is weak – we do not study the injection of droplets but we are interested in suspended droplets and the temperature of droplets is close to the saturation temperature.
These hypotheses allow us to consider a 4-equation model, based on the Navier-Stokes equations (four conservative equations for the mass of mixture, the mass of liquid phase, the momentum of mixture and the energy of mixture), instead of the so called two-fluid model in order to model the droplets-gases flow. The system is closed by relations involving the relative speed between the liquid and gaseous phases and the droplets' temperature. We also take into account a species conservation equation for the multi-component gaseous phase. Experimental data to validate both Lumped Parameter and CFD models is however necessary. This is the object of the following study.

2 EXPERIMENTAL STUDY: EXPANDING SPHERICAL FLAMES IN WATER MIST

A new experimental set-up has been designed in the laboratory (ICARE-CNRS) [15,16] and is described in this paper. The spray system and the first results on the characteristics of the spray will be also presented.

2.1 Experimental set-up

The bomb is a stainless steel sphere (i.d. 500 mm) equipped with 4 opposites quartz windows (100 mm optical diameter, 40 mm thick); it has a black polished surface in order to suppress multiple diffusion (figure 5). Two tungsten electrodes (diameter 2 mm), located along a diameter of the sphere, are linked to a high voltage source (about 10 kV). The gap between the electrodes is adjustable and is usually fixed around 1 mm. Ignition was produced at the centre of the sphere. The voltage and intensity discharge were measured with a high voltage probe and a current probe (figure 6).

Figure 5. Schematic of the spherical bomb.

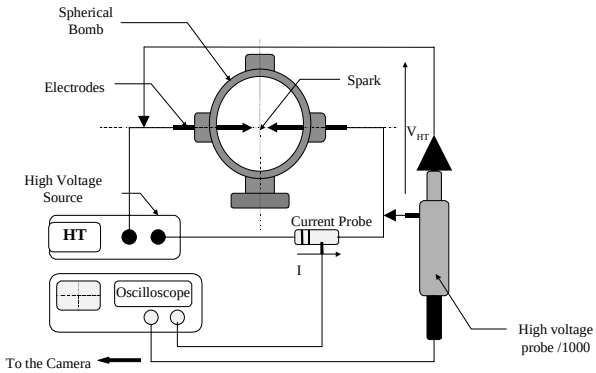

Figure 6. Schematic of the ignition device and the voltage and current probes.

As far as hydrogen is concerned, the visualization of the flame is obtained via the classical Schlieren apparatus. It consists of 2 concave spherical mirrors (100 mm diameter and 1 m focal length), the source light is a continuous argon ion laser, the laser beam is focused via 2 lenses (diam. 75 mm and 20 mm and 150 mm and 22 mm focal lengths respectively). A numerical high speed camera (FASTCAM APX) with an acquisition frequency up to 120 000 images per second was used to register the schlieren images of the growing flame. The images are processed (Visilog 5.2 image processing) in order to derive the radius of the flame in function of time.

2.2 Experimental set-up

The spherical bomb is equipped with different nozzles in order to inject a spray of water inside the bomb prior to ignition or during the flame propagation. To do so, two different nozzles were used and characterized. The first one is a mono-fluid nozzle (SS-LNND-0.60 from Spraying Systems), the water was fed to the nozzle at different initial pressures (between 7 and 50 bars). The size distribution according to the water pressure was measured using a real-time measurements sizer based on the laser light diffraction. As it is shown in figure 7, the size distribution is shifted towards lower value as the

pressure of water is increased. The Sauter diameter decreases from 60 µm at 5 bars to 33 µm at 50 bars.

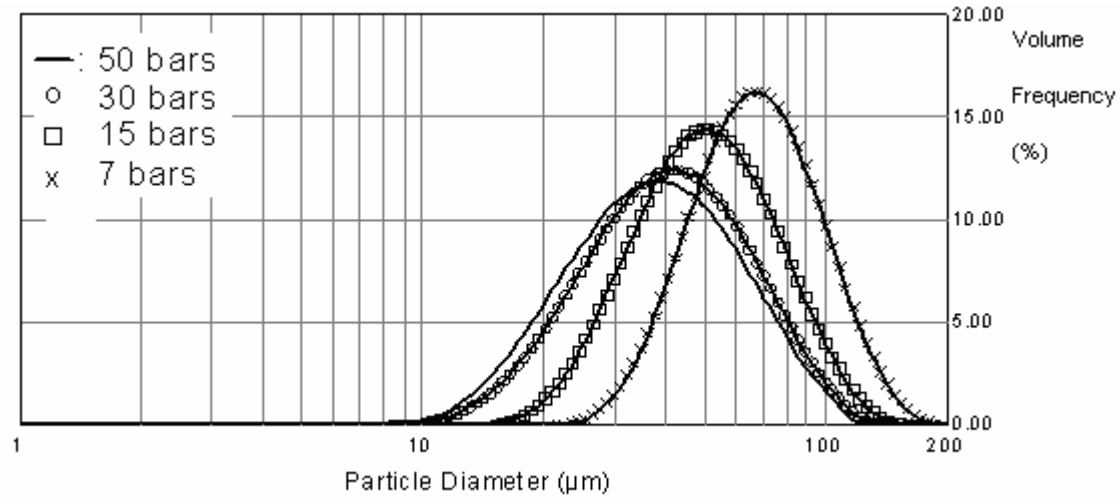

Figure 7. Spray size distribution according to the water pressure using a mono-fluid nozzle.

The second type of nozzle that was characterized is a bi-fluid from Sprayins Systems (LNND-SU1A 1650). In this case water was entrained using compressed air at different pressures. The sprays obtained in this configuration were characterized using the same sizer. The main results are summarized in figure 8.

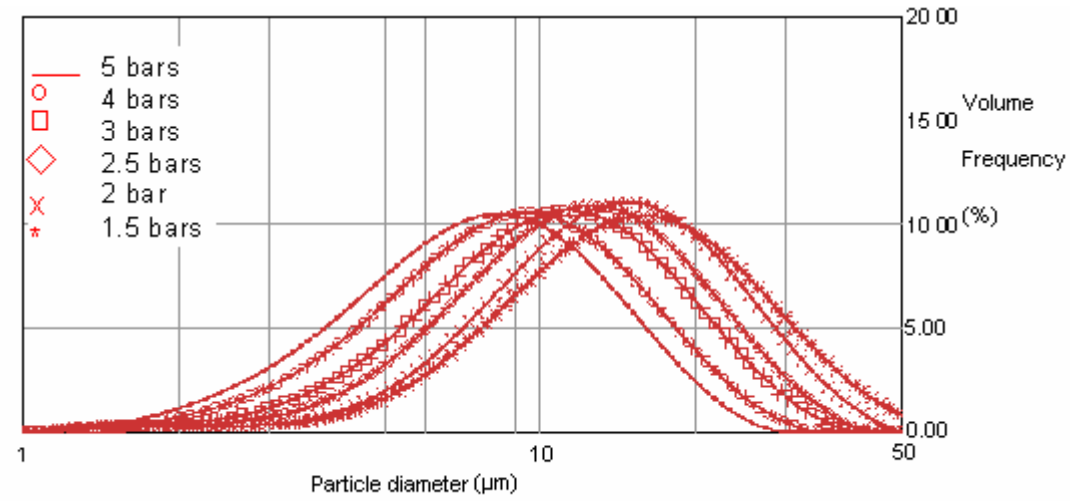

Figure 8. Spray size distribution according to the water pressure using a bi-fluid nozzle.

As it can be seen from figure 8, in the case of a bi-fluid nozzle, the effect of the pressure is limited on the size distribution. The Mean Sauter diameter decreases from 11.5 µm down to 6 µm as the pressure is raised from 1.5 up to 5 bars.

3 CONCLUSIONS

The LP analysis has shown that liquid water generates heat and pressure sinks as expected. It has also highlighted that the steam due to vaporization becomes important as the amount of initial hydrogen attains a certain value (between 19% and 21% with our data) so that the final pressures are higher than the AICC pressure. This first study is a preliminary work for the full CFD modeling taking into account the reaction rate and a polydisperse spray. It has also highlighted that the steam due to vaporization becomes important as the amount of initial hydrogen attains a certain value (between 0.19 and 0.21 with our data) so that the final pressures are higher than the AICC pressure. This first study is a preliminary work for the full CFD taking into account the reaction rate and a polydisperse spray. The first part of the experimental study was devoted to characterize the spray in terms of size distribution before coupling the combustion with the spray.

4 ACKNOWLEDGMENTS

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