

# Experimental investigation of the mechanisms of cellular instabilities developing on two-phase flames

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## 1 Context of the study

Spray combustion plays a major role in various domains such as motorization (spark ignition, diesel and turbojet engines) and in industrial applications (furnaces and boilers). The multiplicity of phenomena such as droplet formation, the vaporization process, turbulent mixing and heterogeneous combustion, which all interact in a dependent manner, makes spray combustion difficult to study. To overcome this complexity, simplified approaches are needed to control all these features. Hayashi and Kumagai [1] were the first to use an aerosol of mono-sized droplets uniformly distributed, generated with a rapid expansion apparatus, as a suitable idealized system for a fundamental study of two-phase combustion. This experimental configuration was later used by [2–4] to carry out studies on the comparison of flame speeds and it was found that under certain conditions a two-phase flame can burn faster than the equivalent gaseous mixture. It was further reported that due to the presence of droplets two-phase flames tend to become unstable, with cells growing on the flame surface. Recently Bradley *et al.* [5] showed that the flame speed promotion was due to these cellular instabilities which increase the fuel mass flux from the fresh gases toward the reactive zone.

A fast burning rate can be an advantage for combustion engines but is a serious disadvantage in a hazard context. Cellular instabilities have been demonstrated to play an essential role in flame speed enhancement. Thus, the ability to predict the onset of cellularity for two-phase mixtures seems to be essential from a scientific point of view. However, the mechanisms responsible for the development of cellular instabilities in heterogeneous mixtures are still poorly understood and only a few studies can be found. The present study proposes an experimental approach to isolate and evaluate various potential mechanisms to improve the comprehension of this domain.

## 2 Proposed approach

The propagation of two-phase flames can be divided into three main phenomena:

- a) The heat release of the burning flame evaporates the surrounding droplets: the droplets act as a thermal sink on the flame (proportional to the latent heat of vaporization of the liquid fuel);

- b) Due to evaporation, a gradient of gaseous fuel concentration along with a gradient of temperature is created around each droplet: this gradient of concentration locally changes the flame burning rate and modifies the mass and thermal diffusion in the reactive zone and the fresh gases;
- c) If the liquid fuel is not fully evaporated before the arrival of the flame, liquid droplets can be present in the reactive zone: in this case, droplets can act as a physical obstacle on the flame front.

The challenge is to study these phenomena in an isolated manner in order to extract the relevant mechanisms of the onset of cellular instabilities. The present approach proposes to consider the three steps described above as hypothetical mechanisms. Then the strategy consists in suppressing one potential mechanism while maintaining the other two. Herein the gradient of gaseous fuel concentration formed around the droplets is discarded and the fuel aerosol is replaced by an inert aerosol. Water was selected owing to its very low reactivity and its well-established thermodynamic data. Additionally, two different gaseous flammable environments were chosen in order to control and investigate the diffusive balance between mass and thermal diffusion which can be created by a gaseous fuel gradient: a  $C_3H_8/air$  mixture in order to explore the nonequidiffusive property, and a  $CH_4/air$  mixture for the equidiffusive property.

### 3 Experimental device and procedure

#### 3.1 The dual chamber

The experiments were performed in a pressure-release type dual chamber which consists of a spherical combustion chamber affording a volume of 1 L, centered in a high pressure chamber of 11 L. The combustion chamber is filled with the flammable mixture while the high pressure chamber is filled simultaneously with nitrogen at the same pressure. The combustion chamber is equipped with 6 evacuation valves which open during combustion and the gases are evacuated toward the high pressure chamber. The reader is referred to [6] for more details. This kind of combustion apparatus design allows safe operation and constant-pressure measurements which are well suited for flame instability studies, especially at high pressure [7]. A representative schema of the whole device is given on Fig. 1.

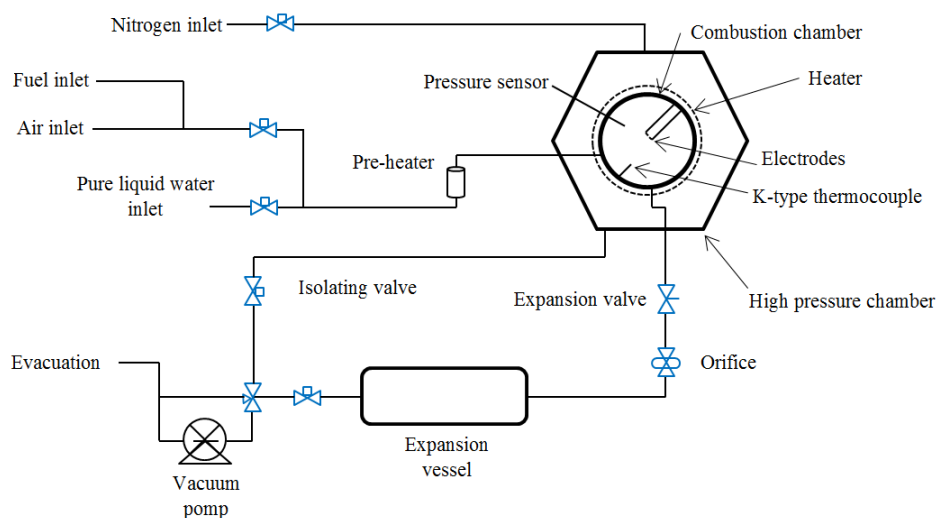


Figure 1: Schematic representation of the experimental apparatus.

### 3.2 Creation of the droplet aerosol

The gaseous mixture of water-air-fuel is first introduced in the combustion chamber after creating a vacuum. A switching valve ensures the injection of an accurate and controlled volume of pure liquid water. The water is then mixed with the gaseous mixture air-fuel and is subsequently vaporized by a pre-heater before its injection in the combustion chamber. An electric heating cable, rolled up around the combustion chamber, heats the fresh gases, thus maintaining the water in the gaseous state. The flammable mixtures of fuel-air were prepared upstream, thanks to two mass flow meters, at the desired equivalence ratio by blending individual gases.

The cloud of droplets is created from the gaseous water-air-fuel mixtures using the condensation technique of expansion cooling (based on the Wilson cloud chamber principle [8]) which is widely recognized to generate well-defined monodisperse droplets with a narrow size distribution. To achieve this purpose, the combustion chamber is connected through a pipe to a 0.5 liter expansion vessel. An expansion valve and an adjustable orifice are placed on this pipe and used to isolate the tank initially set to vacuum (see Fig. 1). The opening of the expansion valve induces a fast pressure drop which decreases the temperature of the mixture in the combustion chamber. The orifice enables the time of the pressure drop to be adjusted. When the partial pressure of the water becomes greater than its saturation pressure, the wet regime is reached, causing the formation of water droplets. Two tungsten electrodes centrally ignite the heterogeneous mixture and the flame spreads spherically.

A K-type thermocouple and a pressure sensor placed near-wall in the combustion chamber control the temperature and the pressure during the filling process of the gaseous mixture and measure their variations during the expansion of the aerosol formation.

### 3.3 Optical diagnostics and aerosol characterization

Two pairs of aligned transparent windows on the two chambers allow optical access. A conventional shadowgraphy system is employed in order to visualize the flame morphology. An Energetiq LDLS (Laser-Driven-Light-Source) is used as the light source and a high speed camera (Phantom v1210) continuously records the flame evolution. The droplet diameter measurements of the aerosol were performed by means of a Sympatec HELOS laser diffraction particle size analyzer. Statistical calculations are done from the droplet diameter data collected at each measurement and the Sauter Mean Diameter (*SMD*) is employed as a representative diameter of the aerosol. The number density  $N_d$ , defined as the number of droplets per unit of volume, was estimated from calculations based on the Beer-Lambert law using droplet diameter and laser attenuation measurements.

It is also important to know the initial quantity of water vapor present in the gaseous mixture before the aerosol formation. This parameter quantifies both the gaseous and the liquid quantities of water at the moment of ignition for the aerosol combustion. As the water is injected with the flammable mixture in the combustion chamber, the initial quantity of water vapor is defined as a dilution rate  $f$ , such that:

$$f = \frac{n_{water}}{n_{total}} \times 100 \quad \text{with} \quad n_{water} = \frac{M_{water} \cdot V_{water}}{\rho_{water}} \quad \text{and} \quad n_{total} = \frac{P_u \cdot V_{chamber}}{R \cdot T_u} \quad (1)$$

where  $n_{water}$  and  $n_{total}$  are respectively the molar quantities of water vapor and of total gaseous mixture prepared in the chamber,  $V_{water}$  is the volume of injected liquid water,  $\rho_{water}$  and  $M_{water}$  are respectively the density and the molar mass of liquid water,  $P_u$  and  $T_u$  the initial conditions of pressure and temperature of the gaseous mixture and  $V_{chamber}$  the volume of the chamber.

## 4 Results

### 4.1 Experimental considerations and specifications

For equidiffusive flames, the thermal conduction from the flame toward the fresh gases is equal to the mass diffusion of the fresh gases to the flame front which stabilize the flame. However, for nonequidiffusive flames, an imbalance between the mass and the thermal diffusivity can either stabilize or destabilize the flame. A proper way to quantify the intensity of this diffusive balance is to determine the Lewis number. The definition from Addabbo *et al.* [9] with the effective Lewis number  $Le_{eff}$ , was adopted as follows:

$$Le_{eff} = 1 + \frac{(Le_E - 1) + (Le_D - 1) \cdot A}{1 + A} \quad \text{with} \quad A = 1 + Ze \cdot (\Phi - 1) \quad (2)$$

where  $Le_E$  and  $Le_D$  are respectively the Lewis number of the reactant that is in excess and deficient,  $Ze$  the Zeldovich number corresponding to  $Ze = E_a \cdot (T_{ad} - T_u) / R \cdot T_{ad}^2$ , with  $E_a$  the activation energy,  $T_{ad}$  the adiabatic flame temperature,  $T_u$  the temperature of the fresh gases and  $R$  the ideal gas constant. Here,  $\Phi$  is defined as the ratio of the mass of excess-to-deficient reactants in the fresh mixture relative to their stoichiometric ratio (equal to  $\phi$  for fuel-rich mixtures and  $1/\phi$  for lean-fuel mixtures).

Figure 2 (left) shows the calculation of the effective Lewis number  $Le_{eff}$  for the two mixtures  $C_3H_8/air$  and  $CH_4/air$  against the equivalence ratio. The methane-air mixture is stable over the equivalence ratio range. The predominant thermal diffusivity for a lean propane-air mixture tends to stabilize the flame whereas for the rich case, the strong mass diffusion can result in the propane-air flame becoming unstable. Fixed equivalence ratios were chosen to perform the experiments according to these specifications. Two equivalence ratios of 0.7 and 1.3 were selected for the propane-air mixture in order to investigate the two sides of the diffusive balance (respectively  $Le_{eff} > 1$  and  $Le_{eff} \leq 1$ ) and a conventional stoichiometric equivalence ratio was taken for the methane-air mixture.

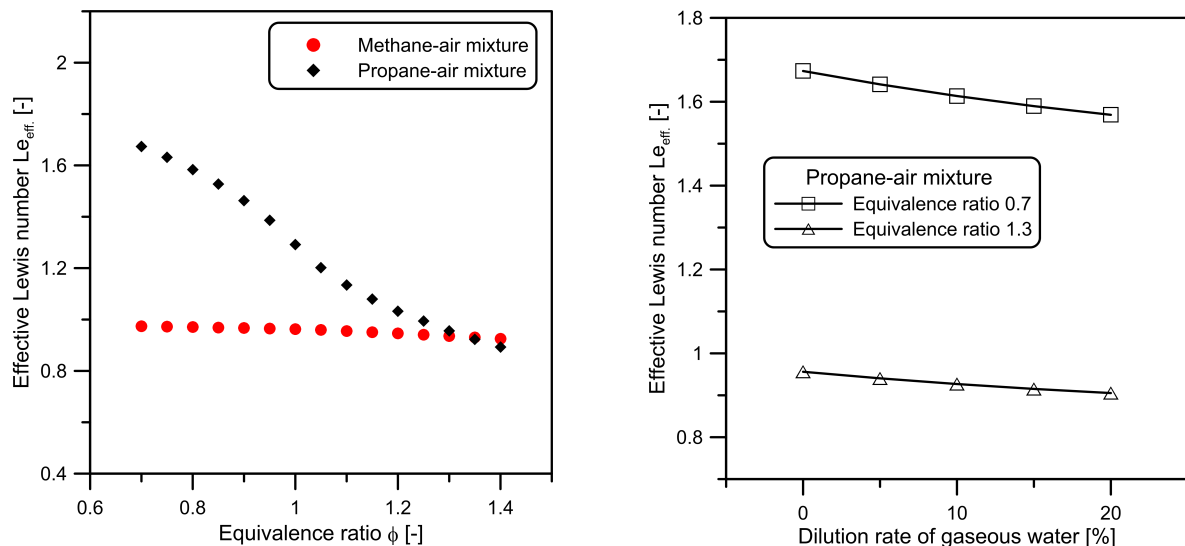


Figure 2: Left: Calculated effective Lewis number against the equivalence ratio for methane ( $CH_4$ ) and propane ( $C_3H_8$ ) in air. Right: Calculated effective Lewis number for a propane-air mixture against the dilution rate of gaseous water at  $\phi = 0.7$  and  $\phi = 1.3$ .

The creation of water mist causes the reactive mixture to burn in an environment with water present both in a gaseous and a liquid state. However the water vapor causes dilution due to the reduction in reactants concentration. Thus it is important to ensure that the water vapor resulting from the aerosol formation does not modify the thermal and mass diffusion controlled by the two air-fuel mixtures. The effective Lewis number was therefore computed for the equidiffusive mixture against a realistic (below the upper flammability limit) water dilution rate range (see figure 2 (right)). The curves demonstrate that the effective Lewis number changes very slightly with the water dilution rate. Consequently, the water aerosol can be considered as a proper inert aerosol, acting only as a thermal agent. Moreover it is to be noted that the chemical effect of the water vapor is considerably lower than the thermal effect and was assumed to be negligible.

## 4.2 Two-phase flame observations

The initial water dilution rate was varied from 3 % to 10 %, two initial pressures of 3 *bar* and 4 *bar* were explored and several pressure drops together with several times of pressure drop were performed. A droplet diameter range of 6-11  $\mu\text{m}$  and number densities between 0.5 and  $2.10^{10}$  *droplets/m*<sup>3</sup> were covered. The initial temperature was kept constant at 358 *K*. The same data-set of initial conditions was kept for the three different mixtures in order to allow relevant comparisons.

Three main behaviors were observed during the experiments: 1) stable and smooth flame; 2) deformed and cellular flame and 3) flame extinction. The latter was encountered for the three flammable mixtures because of the high latent heat of water vaporization. Nevertheless, cellular flames were observed only for some propane-air mixtures at  $\phi = 1.3$  while all the methane-air flames and all the lean cases of propane-air were stable and non-cellular (see figure 3).

This result shows that the diffusive balance of the flammable gaseous mixture is thus predominant for the onset of cellular instabilities and that the presence of droplets triggers the development of cells only for the nonequidiffusive gaseous mixture where the Lewis number is  $Le \leq 1$ . Further investigations are necessary but currently these results suggest that the gradient of fuel concentration forming around the droplets could be that of significant mechanisms of cell development by affording a suitable diffusivity disparity with a Lewis number less than unity.



Figure 3: Examples of shadowgraph frames. Left: methane-air-water flame aerosol at near-stoichiometry. Middle: propane-air-water flame aerosol at  $\phi = 0.7$ . Right: propane-air-water flame aerosol at  $\phi = 1.3$ .

Diagrams were constructed to exhibit the experimental observations with the initial conditions of pressure at ignition and water dilution rate and with the measured number density  $N_d$  and  $SMD$  for each

mixture. It was found that the nature of the water aerosol in terms of droplet diameter and number density seems to have no effect on the onset of cellular instabilities. Moreover, to go further in our investigation, characteristic parameters will be computed. Thus a vaporization Damköhler number  $Da$  (expressing the ratio between the vaporization time of the droplets to the chemical time of combustion) will be calculated to determine if the droplets were present or not within the reactive zone and could act as an obstruction. These computations will provide additional important information on the present results.

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