

Effect of Preferential Diffusion, Activation Energy and Obstacles Separation Distance on the Acceleration of Premixed Flames

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1 Introduction

In later years, investigation about flame hydrogen enriched flames acceleration has greatly increased due to safety reasons. In particular, a hydrogen enriched fuel can present a deflagration to detonation transition (DDT). In a detonation the pressure raises sharply through the wave, sometimes over 20 times the initial pressure [1]. Although the effects of obstacles blockage ratio and separation have been generally considered to study the dynamics of flame acceleration, the effects of preferential diffusion and activation energy are also important. These effects can be studied by considering the Lewis (Le) and Zeldovich (Ze) numbers. The Le number relates the thermal diffusivity to the mass diffusivity while the Ze number is associated with the global activation energy and the level of temperature increase. In previous work, the authors have studied the propagation behavior of premixed natural gas (NG) / hydrogen (H₂) / Helium (He) air flames [2][3], propane (C₃H₈) / H₂ / He / air flames [4] a duct without obstacles with constant cross-section[2], a flame arrester housing with parallel plates [3] and an empty flame arrester housing [4]. Moreover, the influence of flame propagation velocity on the performance of parallel plates arrester element have also been addressed [5]. All these studies considered stoichiometric mixtures and took into account the effects of preferential diffusion and activation energy through the Le and Ze numbers, which for the mixtures of NG / H₂ / air varied between 0.60 to 1.36 and 4.24 to 8.45, respectively. On the other hand, the C₃H₈ / H₂ / He / Air mixtures presented Le and Ze numbers ranging from 0.99 to 1.36 and from 5.37 to 7.20, respectively.

As aforementioned, an easy way to achieve fast flames and DDT is through the presence of obstacles in ducts and it is common practice to use orifice plates as obstacles for flames propagating in ducts or tubes [6]. Normally, the obstacles considered in experiments are rigid, however, the effect of flexible obstacles has also been studied [7] showing that flame acceleration is more prominent with rigid obstacles. Other obstacles types has also been studied in the literature, as for example cylindrical obstacles [8] and continuous triangular cross-section obstacles [9]. The results available in the literature show that the flame acceleration can be intensified or damped depending on the separation distance between obstacles [10][11]. The orifice plates obstacles are characterized by their blockage ratio which is defined as $BR = 1 - A_{\text{free}}/A_{\text{total}}$. It has been shown that a $BR=0.70$ produces stronger flame acceleration than $BR=0.50$ and $BR=0.30$ [12]. Numerical simulations have shown that obstacles with higher BR increase the

turbulence intensity [13]. Also, in the case of hydrogen / air mixtures, the strongest flame acceleration was obtained for a equivalence ratio (ϕ) between 1.0 and 1.5 [12].

A theory for the mechanism of flame acceleration in a duct with obstacles has been proposed by Valiev et al. [14] for incompressible flow and has been extended by Bychkov et al. [15] for compressible flow. The mechanism establishes that a pocket of unreacted gases is formed between the obstacles, the mixture inside a pocket burns after the flame front has passed its axial position, generates a jet of product gases which leave the pocket and contributes to the flame acceleration by acting as a piston which moves in the direction of flame propagation. Numerical simulations of detonation propagation generally simplify the diffusion processes by applying the Sutherland's law [11]. However, the importance of such processes should not be neglected for flame acceleration and DDT studies. Moreover, the theoretical developments presented by Bychkov et al. [15] should be compared to new experimental data.

The objective of the present work is to experimentally study the effect of the preferential diffusion, activation energy and obstacles spacing on the acceleration of premixed flames inside a duct with orifice plates obstacles. Therefore, three stoichiometric mixtures are considered: NG / air, NG / H₂ / air and NG / He / Air. These mixtures span Le and Ze numbers between 1.09 and 1.36 and 6.09 and 8.45, respectively, at initial conditions of 40 kpa and 300 K.

2 Experimental setup

The experiments were carried out in a stainless-steel combustion duct with total length of 1050 mm. The duct was composed of three modules. Each module was 350 mm long with 96 mm x 96 mm internal square cross-section each. The modules were equipped with two 25 mm-thick glass windows (225 mm x 60 mm) which allow for the visualization of the flame propagation. Three piezoelectric absolute pressure sensors were used. Two mounted in the top ports (10 bar each) and one in the right-end flange (1 bar). A 20 L borosilicate spherical vessel contained inside a stainless-steel box was used as mixing chamber. The mixture preparation was performed by the partial pressures' method using a 2-bar absolute pressure transducer. The ignition source which provided 8 kV and 30 mA was a spark plug mounted in the centre of the left-end flange. The schematic representation of the experimental setup is present in Figure 1.

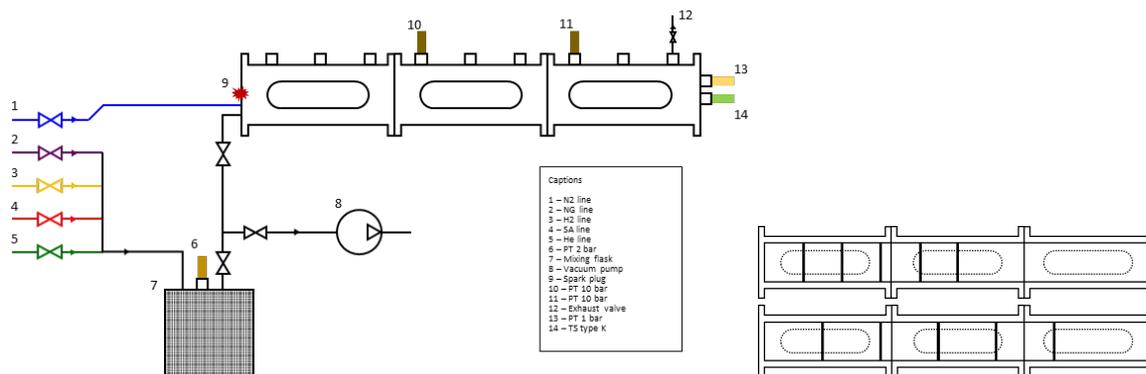


Figure 1. Schematic workbench setup.

To achieve flame acceleration an array of 5 obstacles was used. The obstacles were 3 mm thick square aluminium plates with square orifices. The outer hydraulic diameter (D_h) of the obstacle is 96 mm and the inner hydraulic diameter (d_h) of the orifice is 48 mm. Therefore, these obstacles present a $BR=0.75$. The distance between the obstacles was varied between $1.0D_h$ and $1.5D_h$ in order to observe the effect of the obstacles spacing. The flame propagation was recorded by using a high-speed video camera Phantom v411. The schematic representation of the obstacles position is depicted in Figure 1. The gases used in these experiments were natural gas (NG), hydrogen, helium and synthetic air. The NG volumetric composition was 90.8% CH₄, 6.0% C₂H₆, 1.2% C₃H₈, 0.5% CO₂, and 1.55% N₂, the H₂

had a purity of 99.99%, the helium (He) had a purity of 99.995%, and the synthetic air has a composition of 20% O₂ and 80% N₂. All the experiments were conducted at room temperature (around 300 K) and 40 kpa of initial pressure. The flame propagation velocities were obtained by using the software Tracker [16].

3 Methodology

Three fuel mixtures were used in this study and are presented in Table 1. The mixtures were selected based on their Le and Ze numbers. The Le number of individual fuels were determined by the Eq. (1), proposed by Qiao et al. [17]. The effective Lewis number (Le_{eff}) was determined by using the volumetric concentration of each fuel and its individual Le number as shown in Eq. (1), which was proposed by Bouvet et al. [18]. The Ze number was calculated by using the Eq. (2) with the activation energy obtained as given by Law [19]. The flame propagation velocities were obtained by using the using Cantera Software [20] and the San Diego Mechanism [21]. It has been shown in previous work that the Ze number did not change significantly when other chemical kinetic mechanism were used [3]. In the equations below α is the thermal diffusivity of the mixture, D_{i/O_2} is the binary mass diffusion of the fuel with respect to the oxygen, x_i is the molar fraction, E_a is the activation energy, R is the universal gas constant, T_b and T_u are the adiabatic flame temperature and the unburned gas temperature, respectively. In the detonations literature the reduced activation energy is commonly used ($\varepsilon = E_a/RT_b$), however, by observing Eq. (2) it can be noticed that ε is the first term of the Ze number.

$$Le_i = \frac{\alpha}{D_{i/O_2}} \quad Le_{eff} = \sum_{i=1}^n x_i Le_i \quad (1)$$

$$Ze = \frac{E_a}{RT_b} \left(\frac{T_b - T_u}{T_b} \right) \quad \frac{E_a}{R} = -2 \left[\frac{\partial(\ln \rho_u S_L)}{\partial(1/T_b)} \right]_p \quad (2)$$

Table 1. Mixtures considered for the experiments.

Code	Mixture	$\phi \pm \omega\phi$	Le	Ze (40 kPa)
Mix A	100% NG + Air	1 ± 0.029	1.09	8.20
Mix B	[50% NG + 50% H ₂] + Air	1 ± 0.020	0.60	6.09
Mix C	[100% NG] + 10% He + Air	1 ± 0.032	1.36	8.45

The procedure to determine the uncertainty of the mixture's equivalence ratio involves the determination of the uncertainties related to each partial pressure measured during the mixture preparation step. This procedure has been presented in the authors' previous work [22]. The uncertainty of the equivalence ratio is presented in Table 1. The theoretical approach proposed by Bychkov et al. [15] allows for the determination of the flame tip propagation velocity [Eq. (3)] and the flame tip position [Eq. (4)]. In the Eqs. (3) and (4) the reference Mach number is $Ma = S_L/c_0$, the initial expansion ratio is $\Theta = \rho_u/\rho_b$. The other parameters used in the equations can be found in Eqs. (5) to (7). The properties of the mixtures were determined by using the Cantera Software [20] and the San Diego Mechanism [21].

$$\frac{dZ_f}{dt} = Ma \frac{\chi \sigma_0^2}{S_L} Z_f^2 + \sigma_1 Z_f + \theta_1 S_L \quad (3)$$

$$Z_f = \frac{2\theta_1 S_L [\exp(\sigma_2 t) - 1]}{(\sigma_2 - \sigma_1) \exp(\sigma_2 t) + (\sigma_2 + \sigma_1)} \quad (4)$$

$$\chi = (1 - \alpha)^{-1} + \gamma - 1 \quad \theta_1 = \theta - Ma(\gamma - 1)(\theta - 1)^2 \quad (5)$$

$$\sigma_1 = \sigma_0 \left[1 - Ma \left(\frac{\theta}{1 - \alpha} + 2(\gamma - 1)(\theta - 1) \right) \right] \quad (6)$$

$$\sigma_0 = \frac{\theta - 1 S_L}{1 - \alpha R} \quad \sigma_2 = \sqrt{\sigma_1^2 + 4Ma\chi\theta_1\sigma_0^2} \quad (7)$$

4 Results and discussion

After the ignition, the flame propagates with laminar finger shape and transitions to a mushroom shape as the flame accelerates and goes through the obstacles. As the flame travels the duct, it becomes turbulent, due to the influence of reflected shock waves (both from the duct and the obstacle ahead of the flame) and the unburned gas pocket downstream the obstacle, creating recirculation zones. As the gas pockets burns the burned gases expands into the center of the duct. The flame propagation position obtained from the experiments for the stoichiometric mixture of NG and air is shown in Figures 2. It is possible to observe that the flame accelerates as it goes through the obstacles. The recirculation zones can be observed in Figure 2. In the first stage of flame propagation, the gas inside the recirculation zone takes longer time to burn, due to the laminar surface of the flame. At the second part, the flame surface is turbulent, burning the gas inside the zone faster. This increase in burning rate can in part explain the higher flame tip velocity in the last obstacles. However, the burning of the mixture in the recirculation zones creates a jet of burned gases which leaves the space between the obstacles and acts as a piston, thus, contributing to the flame acceleration.

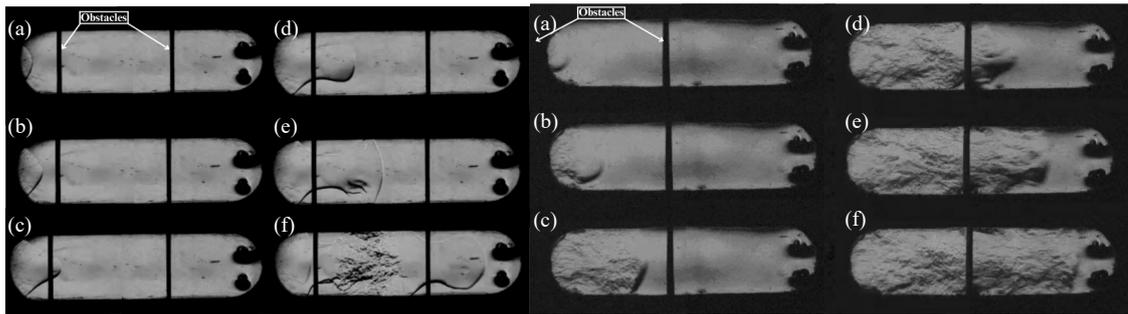


Figure 2. Flame tip position at windows 1 (left) and 2 (right) for mixture A (100% NG / Air). Flame propagation from left to right and the photos follow a-f. Videos were taken at 18000 fps.

The evolution of the dimensionless flame propagation velocity is observed in Figure 3. The higher flame propagation speed was achieved at the last obstacle in the second module. The observed flame propagation behavior agrees with the numerical and experimental studies available in the literature [23]. The zones where no data was collected are not observable due to the location of the windows in the flame propagation duct. Figure 3 shows that the fast flame propagation regime (> 500 m/s) was reached in the second window for the three mixtures and for the obstacles' separation of $1.0D_h$. However, the mixture with the highest Le number had a maximum velocity very close to the 500 m/s. On the other hand, for the obstacles' separation of $1.5D_h$ the flame velocity did not oscillate with the same intensity at the second window, nevertheless, the fast flame regime was reached for all the mixtures. Figure 5 also shows the velocities obtained by applying the theory by Bychkov et al. [15]. It is clear the theory underestimates the velocity on the second window. The influence of the Le and Ze numbers on the flame propagation velocity can be analyzed by observing Figure 3. The mixtures with lower Le and Ze numbers tend to present higher propagation velocities along the duct, this is more obvious for windows 2 due to the scale of the graph. The lower Le number means that the mass diffusion dominates over the heat diffusion. Therefore, there is more fresh reactants being diffused into the reaction zone, thus increasing the burning rate. Note that, the mixtures with Lewis numbers below a critical value ($Le <$

Le_{cr}), were $Le_{cr} < 1$, tend to accelerate when they are positively stretched [2]. On the other hand, when $Le > Le_{cr}$, the positively stretched flames are decelerated.

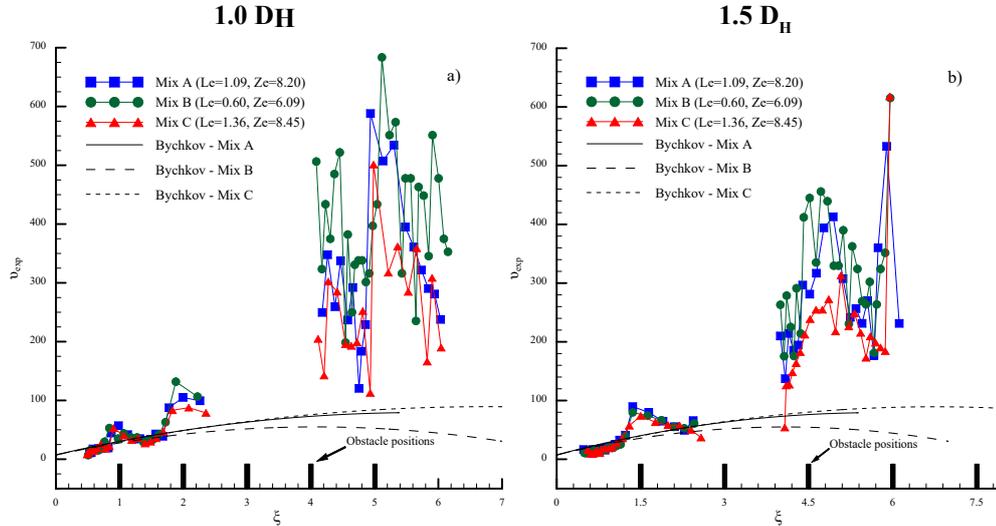


Figure 3. Flame tip velocity versus relative position.

The Figure 4 shows the experimental results compared to the analytical formulation developed by Bychkov et al. [15]. The theoretical models for the incompressible flow and the compressible flow assumptions are shown in these figures. The theoretical and experimental results show good agreement for the early stages of flame acceleration observed in the first window. As the flame accelerates, the theoretical model and the experimental results start to show significant differences (second window).

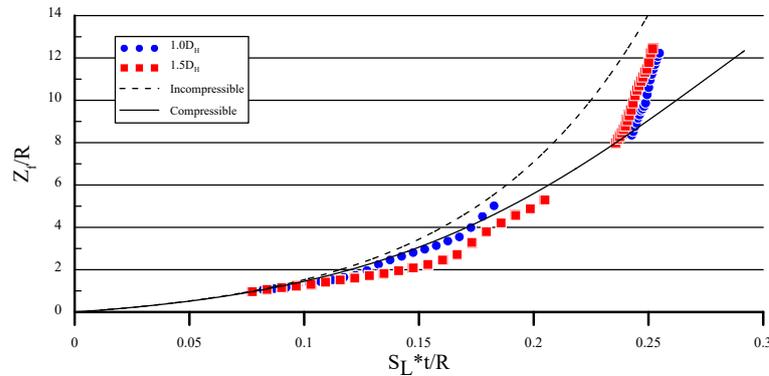


Figure 4. Dimensionless flame tip position versus dimensionless time for 100% NG / air mixture. The lines are the formulation developed Bychkov et al. [15] for compressible and incompressible models.

5 Conclusions

Experiments were conducted with three different mixtures, considering Lewis (Le) numbers ranging from 0.60 to 1.36 and Zeldovich (Ze) numbers ranging from 4.24 to 8.45. It was observed that the mixtures with lower Le numbers achieve higher flame propagation velocities. The mixtures with higher Ze numbers present higher activation energy, therefore showing lower flame propagation velocity. Note that hydrogen addition has the effect of lowering both, the Lewis and the Zeldovich numbers. On the other hand, helium addition increases both dimensionless numbers. The orifice plates separation distances of $1.0D_H$ and $1.5D_H$ both showed the development of fast flames for the three mixtures considered. However, due to the experimental configuration, the maximum pressure was higher for the $1.5D_H$ separation distance. The flame achieves higher velocities at the beginning of windows 2 for the

1.0Dh separation distance. The theoretical model showed a reasonable agreement for the flame tip position at the early stages of flame propagation (windows 1). However, at the more advanced stages (windows 2) the theoretical model showed low accuracy. The theoretical results were also compared with the flame propagation velocity, confirming the inaccuracies for the later stages of flame propagation.

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