

# Highlights of the Flame Acceleration in a Confined Nonuniform $\mathrm{H2}/\mathrm{O2}/\mathrm{N2}$ Mixture

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# Highlights of the Flame Acceleration in a Confined Nonuniform H<sub>2</sub>/O<sub>2</sub>/N<sub>2</sub> Mixture

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Gaseous explosion models generally assume the gas mixture to be uniform. However, in a real explosion, the vapor cloud may not be homogeneous, and repartitioning of the reactivity inside the cloud can be subject to wide spatial variations. In this work, experimental tests were run to study the flame propagation and acceleration in nonuniform mixtures. Experiments were performed in a long vertical confined tube with a square cross section, composed of four equal sections. A gate valve separated the tube into two parts, and the composition of the gases was different on each side of the valve. The opening of the valve permitted the mixing of gases by molecular diffusion. For nonuniform mixtures, a mode of propagation identical to that seen in uniform mixtures was observed; however, a third phase of propagation was found, in which the flame velocity increased strongly. This increase occurred with higher hydrogen concentration in an upwardpropagating flame. A concentration gradient can appreciably modify the trajectory and acceleration of a flame. Here, however, the incidence of pressure effects remained modest, since the combustion was confined and the final pressure depended mainly on the quantity of reactants available. © 2009 American Institute of Chemical Engineers Process Saf Prog 00: 000–000, 2009 Keywords: hydrogen; flame; concentration gradi-

ent; nonuniform reactive mixture

### INTRODUCTION

The story of industrialization has been punctuated by explosions, whose frequency and severity have

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increased with the expansion of industry. Several explosions, such as Flixborough (1974) in the US, La Mède (1992) in France, and more recently, Buncefield (2005) in England, are memorable. The extreme damages incurred by these accidents have considerably affected the neighborhoods and pushed the public authorities to reinforce the regulations governing industries.

100 Due to public demand, tools for explosion risk analysis and consequences have been progressively developed. These tools are based on observations, analysis of past accidents, or large scale experiments. Developed in the 70s and 80s, the Multi-Energy method [1] serves as a common tool of analysis. 106 Although related tools have become more and more predictive, important simplifications have been made, such as assuming a homogeneous combustible cloud 109 for the Multi-Energy method. However, in real situations, heterogeneities exist. For instance, turbulent jets, induced by massive leaks, are characterized by a much larger concentration near the breach than further downstream. Such leaks produce a significantly 114 nonuniform mixture. Simple simulations [2] show that assuming an equivalent homogenous mixture under-116 estimates damaging effects. Moreover, this method 117 was used in the Buncefield postaccident analysis. It 118 predicted 20-50 mbar of overpressure on buildings, 119 whereas the postaccident analysis revealed magni-120 tudes of overpressure around 700-1,000 mbar [3]. The propagation of an explosion in a space with a varied blockage ratio does not seem sufficient to explain 123 this discrepancy in the overpressures, and one expla-124 nation advocated by the Britannic expert committee is cloud heterogeneity.

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**Figure 1.** Global view of experimental set-up. [Color figure can be viewed in the online issue, which is available at **www.interscience.wiley.com**.]

Direct experimental works can assist in the confirmation of this assumption. Indeed, Girard et al. [4] noted a multiplication of maximum overpressure by a factor of three for a flame propagation in two mixtures of different equivalence ratios created by concentric soap bubbles, as well as an important increase of flame propagation speed. A modification of flame propagation speed has also been observed by Karim and Panlilio [5], who noted an increase of 70% for a flame propagating in a vertical open tube filled by a stratified methane/air mixture. The conclusions of Whitehouse et al. [6] are the same for a hydrogen/air flame. In a closed tube, they also noted an important increase in the maximum overpressure. For a freepropagation flame, Sochet et al. [7] showed that explosion effects are linked to the history of the flame through the cloud and that a concentration gradient may significantly modify the pressure evolution, maximum levels reached, and impulse. Furthermore, the effects of cloud heterogeneities are linked to modifications of the fundamental parameters of combustion, such as expansion ratio and laminar flame velocity. The variations generate a differential acceleration between rich and poor zones, with important modifications of the flame front surface. Hirano et al. [8] confirmed that the differential in expansion velocity induced by a heterogeneous mixture creates an increase of flame front area compared with the equivalent homogeneous mixture. This increase can be added to the effect of cloud heterogeneities, suggesting that the dynamics of flame can affect the kinematics of flame.

An analysis carried out in a previous paper [9] highlighted the influence of the acoustic wave interaction on the flame front and the influences of thermal losses to and vapor condensation on the walls on the pressure signal and the mechanism of flame propagation. Taking these effects into account, in this study, we aimed to analyze the situation in the case of a nonuniform mixture.

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#### **EXPERIMENTAL SET-UP**

To carry out this study, a special setup was designed. We needed a very careful control of the initial conditions, including the repartition of the reactants. If any convection current were to appear, erratic pockets of mixture could be produced. Consequently, the diameter of the tube needed to be kept 230 231 small (a few centimeters) so that the formation of gra-232 dients could result from molecular diffusion. It was 233 further estimated that a length of tube on the order of 1 m would permit simulation of the range of reac-234 tivity gradients likely to be produced in practical sit-235 uations (0.1-1 m). 236

The tested setup was a 2-m-long tube, to simulate 237 the range of reactivity gradients that might appear in 238 239 industry. The sides consisted of 0.03-m-long square sections (Figure 1) to limit the possible convection **F**10 effects. The specifications of the setup were that it 241 should be transparent and resistant to a high-pressure explosion (around 150 bar). Gate valves separated 243 the tube into four equal sections. Each section of the 244 tube was composed of three PMMA walls and one 245 aluminum wall set in a metallic skeleton. Gas tight-246 ness was ensured by the application of silicone putty. 247 248 The skeleton provided support for all of the mechanical stresses.

The gate valves (Figure 2) were two steel shells in  $\frac{2}{12}$  which a thin aluminum sheet slides. The composition 251 of the gases was different on both sides of the valve. 252

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Figure 2. Gate valve. [Color figure can be viewed in the online issue, which is available at www. interscience.wiley.com.]



Figure 3. Opacimeters. [Color figure can be viewed in the online issue, which is available at www. interscience.wiley.com.]

The opening of the valve permitted the mixing of gases by molecular diffusion.

A special effort was made to develop the instrumentation necessary for conveniently interpret the tests; specifically, an opacimeter system consisting of a laser diode and a photovoltaic cell fixed on two aluminum supports magnetically mounted on the metallic skeleton was employed (Figure 3). Eight F3 opacimeters were distributed along the tube, with dual functions. They allowed detection of the flame by a modification of a luminous laser signal detected by the photovoltaic cell, consequently allowing us to obtain both the flame speed and trajectory. Moreover, this allowed deduction of the gas mixing ratio. Nanoparticles of ammonium chloride were added to only one part of the tube, and the particles diffused out with the gases after the opening of the gate valve. The "opacity" of the gas changed according to the



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Figure 4. Distribution of species in the tube after 10 min of diffusion. [Color figure can be viewed in the online issue, which is available at **www.interscience.** wiley.com.]

mixing and was detected by measuring the transmittance at each detector location. An experimental calibration law links the light attenuation ratio and the gas mixing ratio.

A second optical technique was developed to observe the flame area. It consisted of illuminating a thin slab of the tube with an argon ion laser via a rotating mirror. During its propagation, the flame dissociates the ammonium chloride particles without modifying the combustion. As green light is diffused by ammonium chloride, the flame area can be detected via high-speed video to the threshold 347 between high- and low-contrast regions. Pressure 348 was measured by a classical piezoresistive gauge. 349 One pressure gauge was located at the middle of 350 each section. Hence, pressure gauge 1 was at a dis-351 tance of 0.25 m, pressure gauge 2 at 0.75 m, pressure 352 gauge 3 at 1.25 m, and pressure gauge 4 at 1.75 m 353 from the bottom.

354 We considered a nonuniform mixture with an av-355 erage nitrogen ratio of 62.5%. This was accomplished 356 by setting in contact the upper part (1 m) of the 357 tube, filled with mixture M3 (30% vol.  $H_2 + 15\% O_2$ + 55%  $N_2$ ), with the lower part (1 m) of the tube filled with mixture M1 (20%  $H_2 + 10\% O_2 + 70\%$ N<sub>2</sub>). The ignition took place 10 min after the opening of the valve. The distribution of the species at the ignition time, calculated by Fick's (one-dimensional) law of diffusion, is presented in Figure 4. The gradi- $\mathbf{F}$ ent zone extends 0.70 m, from 0.65 to 1.35 m.

#### PRESSURE ANALYSIS

368 The pressure signals obtained after ignition of the nonuniform mixture (Figure 4) are reported in Figure 5. There were identical responses in each section. A 3**F5**0 high-frequency signal of low amplitude (typically 100 mbar at 100 Hz), corresponding to the local 372 peaks of  $F_i$  overpressure, is superimposed on an en-374 velope of large amplitude at low frequency (typically 3 bar at 10 Hz, between 0 and F). The fitted pressure signals obtained in each section are superimposed, meaning that the pressure was homogeneous in the tube overall.

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**Figure 5.** Pressure signals in each section of the tube—nonuniform mixture. [Color figure can be viewed in the online issue, which is available at **www.interscience.wiley.com**.]



**Figure 6.** Increase in size of the pressure signal between 0 and 0.3 s of the pressure signals—nonuniform mixture. [Color figure can be viewed in the online issue, which is available at **www.interscience.wiley. com**.]

An increase in size of the pressure signal between **F6** 0 and 0.3 s is presented in Figure 6. One notes a first phase, of 0.062 s duration, where the pressure increases almost linearly upto an inflection point 11, marking a break in the slope. Beyond this point, the increase in pressure is linear upto a second inflection point, I2, at the time tb (0.103 s). The I2 point marks a second break in slope. Beyond tb, the pressure increases linearly to a maximum at P2, reached at tc (0.1456 s). Then, the pressure decreases exponentially until time tr (0.0586 s), upto a final pressure value systematically smaller than the atmospheric pressure, typically about 0.7 bar.

A traditional model of combustion by sections was used. In a closed chamber, the inflammation of a gas pocket leads to the development of a flame front in both directions in the chamber. The combustion of each section by the flame front causes a compression of fresh and burned gas due to the expansion of the combustion products. In the burned gas, the progressive compression causes a highly increased tempera-



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**Figure 7.** Pressure evolution versus time—nonuniform mixture. [Color figure can be viewed in the online issue, which is available at **www.interscience.wiley.com**.]

ture. In the fresh gas, the increasing pressure leads to a temperature increase as well, and due to the burned gas expansion, the gases are moving.

To calculate the thermodynamics parameters during the combustion, the following assumptions were considered:

- -the gas follows the ideal gas law
- -the combustion occurs at constant volume
- —the gaseous mixture is divided into *N* sections of the same mass
- -the specific heat at constant pressure of the ideal gas is constant
- —the pressure is the same at each point of the chamber at a given time.

480 Heat losses appear due to thermal and volume contractions of the gas and are conducive to a reduction in pressure effects. The combustion by sections 483 model was adapted to our study by including thermal 484losses by conduction and condensation and the effect of mixture nonuniformities. First, each section of 486 burned gas transfers heat to the wall by turbulent thermal conduction in a nonstationary regime. Sec-488ond, each section of burned gas also transfers heat by a nonstationary flow of water vapor to the cold wall, where it condenses integrally. 491

By introducing the experimental flame trajectory into the model of combustion by sections, the pressure and the flame surface can be determined. The pressure signal calculated with this model is very well correlated with the experimental signal (Figure 7).

### FLAME PROPAGATION ANALYSIS

#### **Flame Trajectory**

The flame propagation was deduced by exploiting 501 high-speed video. A detailed analysis is possible 502 by tracing the trajectory of the flame with time 503 (Figure 8). **F8**<sup>4</sup>

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As for the pressure signal, an oscillation in the trajectory that is similar to the pressure oscillations is superimposed on the average evolution. These oscillations disappear after point B, corresponding to the second inflection point I2. Three distinct modes appear during the flame propagation: between O and A, A and B, and B and C. The sections OA to BC are characterized by a mean flame velocities of 13.5 m s<sup>-1</sup> in section OA, 21.8 m s<sup>-1</sup> in section AB, and 10.5 m s<sup>-1</sup> in section BC. Notably, between A and B, we observed a nonlinear increase in the propagation velocity.

It is also interesting that the mean flame velocity in section OA (13.5 m/s), where the concentration of the mixture corresponds to M1, is similar to the flame velocity in section OA (15.5 m s<sup>-1</sup>) for the homogeneous mixture M1. However, the mean velocity in section BC  $(10.5 \text{ m s}^{-1})$  is very different from the velocity at the end of propagation in the case of the homogeneous tests of the corresponding mixture M3 (47 m s<sup>-</sup>

The break in the flame trajectory coincides with the beginning of the gradient zone at 1 m in the direction of flame propagation.

### **Flame Structure**

One can also extract the evolution of the flame surface with respect to the section of the tube according to the position of the flame (Figure 9).

The combustion by sections code made it possible to estimate the effects of compression on the size and the position of the gradient zone at the time of its interaction with the flame. The gradient zone extends 70 cm, from 0.65 to 1.35 m (Figure 10). We calculated, using the combustion by sections code during the flame propagation, that the gradient zone is pushed about 30 cm toward the higher part of the tube, extending from 0.95 to 1.65 m.

## COMPARISON OF COMBUSTION BETWEEN NONUNIFORM AND UNIFORM MIXTURES

### **Pressure Signals**

In the case of a nonuniform mixture (Figure 6), there is a difference in the high-frequency fraction of the signal compared with homogeneous mixtures.



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Figure 9. Ratio of the flame surface on the tube section-nonuniform mixture. [Color figure can be viewed in the online issue, which is available at **www.** interscience.wiley.com.]



Figure 10. Interaction of gradient concentration with the flame-nonuniform mixture. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

With the nonuniform mixtures, the first and the third zones are identical to those of the homogeneous mixture. The first zone extends over 50 ms with frequency characteristics of 100 and 350 Hz and an amplitude of 50 mbar, whereas the third zone, extending from 90 to 147 ms, presents an average frequency of 150 Hz and a low amplitude of 30 mbar. In contrast, in the second zone, ranging between 50 and 90 ms and presenting a frequency of 100 Hz, there is a disappearance of the harmonics and a strong amplifica-625 tion of the amplitude of the oscillations (200 mbar) 627 according to the fundamental mode.

A comparison of the envelope pressure signals for the heterogeneous mixture and the homogeneous mixtures M1, M2 (25% vol. H2 + 13% vol. O2 + 63%

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Figure 11. Pressure signals of uniform mixtures and nonuniform mixture. [Color figure can be viewed in the online issue, which is available at www. interscience.wiley.com.]

vol. N2), and M3 is presented in Figure 11. Even F11 though each mixture contains the same quantity of reactants, the shape of the pressure signals differs, as well as the combustion times and the maximum pressures of the explosions. In particular, although the time of combustion is longer (256 ms, mixture M1; 114 ms, mixture M2; 31 ms, mixture M3; 147 ms, nonuniform mixture), the maximum pressure of explosion is higher for the nonuniform mixture (3.1 bar, nonuniform mixture; 2.3 bar, mixture M1; 2.8 bar, mixture M2; 4.2 bar, mixture M3). This difference is more important with the mixture M3, which is present only in one small fraction of the tube. Heterogeneous mixtures compared with the homogeneous mixture M3, the explosion dynamics are significantly different, which could be explained by considering that during the ignition, the M3 mixture is present only in a small fraction of the tube (20 cm at top). The same can be said for the mixture M2. The pressure signals differ in form, combustion times, and maximum pressure. On first consideration, one might expect that the combustion times are different, since the M3 mixture is more reactive. However, it could also be the case that being present in the tube at the end of the propagation the M3 mixture would rather lead to suppression, similar to the case of the uniform mixture M3.

Nonuniform mixtures compared with the experiments with a uniform mixture M1, there is a good correlation of the signals upto 80 ms (i.e., until the 1.3 m position), which is explained by the presence of mixture M1 in the lower part of the tube. There is also a reasonable agreement between experimental flame surfaces and the model.

### Flame Surface

When ignition occurs in the part of the tube where the composition is the M1 mixture, the excited frequencies are identical to those identified for the experiments undertaken with the homogeneous M1



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Figure 12. Flame surface/tube section for uniform mixtures and nonuniform mixture. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]



Figure 13. Flame trajectory—nonuniform mixture. [Color figure can be viewed in the online issue, which is available at **www.interscience.wiley.com**.]

mixture. The observation that the surface of the flame Af appears to be more dependent on the values of these frequencies and less on the equivalence ratio of the mixture explains why the evolutions in Af/At (At, tube area) appear similar for the heterogeneous mixture and the homogeneous mixture M1 (Figure 12). A significant difference in the evolution of flame surfaces implies a difference in the combustion rate.

However, the fundamental flame velocity and the burning rate are not constant during the propagation 747 of the flame in the case of nonuniform mixtures 748 (Figure 13), which is opposite to what is seen with F13 the uniform mixtures. As the flame meets mixture M3 (in particular at the end of the propagation), 752 for which the laminar flame velocity  $S_{S1}$  is about 2.5 m s<sup>-1</sup> and the ratio Af/At is about 4, it is calcu-753 lated that the flame velocity should be  $\sim 10 \text{ m s}^{-1}$ , which is in good agreement with the experiments  $(10.53 \text{ m s}^{-1} \text{ in section BC}).$ 

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One can also interpret the jump in velocity between A and B in the trajectory of the flame in terms of the acceleration effect of the flame, since the 760**AQ1** velocity of expansion ( $S_1 a$ ) increases from 4 m s<sup>-1</sup> to  $17 \text{ m s}^{-1}$  over a length of 0.2 m. Acceleration is calculated by:  $(17 - 4)^2/0.2 = 850 \text{ m s}^{-2}$ . By using the generalized Taylor model and a wavelength of disturbance equal to the diameter of the tube, one calculates that the flame must accelerate 20 m s<sup>-1</sup> in this zone, which is in conformity with the observation.

### CONCLUSIONS

In case of nonuniform mixtures, we observed a mode of flame propagation identical to that seen with homogenous mixtures; however, a third phase of propagation was also observed, where the flame velocity increases strongly. This increase could be due to the presence of a concentration gradient, which can very appreciably modify the trajectory and acceleration of a flame, according to the methods employed in this study that could be highlighted. The modest impact on pressure effects observed within the framework of this study likely result from the combustion being confined, which causes the final pressure to depend mainly on the quantity of reactants available and not on the mode of combustion. In other circumstances, in a partially confined enclosure or in a free atmosphere, the piston effect of the flame on the pressure effects is significant; thus, the impact of the concentration gradient could be much stronger.

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