The acoustic-parametric instability for gaseous mixtures with Lewis number smaller than one

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1 Introduction

The interaction between pressure waves and the surface of the flame is a feed-back process in which the wave intensity and the heat released by the flame influence each other. Markstein [1] concluded that coupling between both phenomena was made possible by the variation of the total flame surface caused by the pressure changes. The oscillating velocity field created by the waves produces oscillations of the amplitude of the cellular structures of the flames. This variation of the surface alters, in turn, the total amount of fuel consumed and the heat released by the flame which is proportional to the flame area.

Two different instabilities due to flame - pressure waves interaction have been identified [2]. In the *acoustic* instability (see [2]) the cellular structures of the flame front oscillate with the frequency of the acoustic oscillating field. Two effects tend to muffle it. For large wavenumbers, the instability is damped by diffusive processes. For small wavenumbers, it is absorbed by the effect of gravity. The *acoustic* instability corresponds, for zero amplitude of the excitation velocity, to the *Darrieus-Landau* planar instability. For increased values of the oscillating velocity [2], the *acoustic* instability has the notable property of being able to stabilize the *Darrieus-Landau* instability.

The *acoustic* instability may develop, for enhanced oscillating velocity, into the *parametric* instability. Under the latter, the growth rate is generally higher than in the *acoustic* case. The cellular structures of the flame oscillate with a frequency half of the *acoustic*, a fact that was recognized by Markstein as the typical property of the Kapitsa *parametrically* dumped pendulum, who consequently named so the instability.

From the point of view of the severity of an explosion, gaseous mixtures can be classified into two kinds [3]: a) If the two instabilities co-exist for some ranges of the amplitude of the acoustic perturbation, a planar flame front is never stable and the *acoustic* instability transform spontaneously into the *parametric* one; b) If they do not co-exist for any range of acoustic velocities, the *acoustic* instability tends to suppress the Darrieus-Landau instability, the *parametric* instability regime is never reached and planar flame fronts are stable as long as the oscillating velocity field exists.

These two different propagation regimes have been confirmed by the observations of Searby [2] and Aldredge and Killingsworth [4] who performed experiments with downwards propagating flames inside a cylindrical and an annular burner respectively. It was found that the flame propagation was divided into four separate stages: Promptly after ignition, the flame surface quickly became wrinkled due to the *Darrieus-Landau* instability. Then, as the flame propagates further, sound waves started to be generated. Due to the fundamental *acoustic* instability, these acoustic waves caused an attenuation

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of flame wrinkles. The *Darrieus-Landau* instability was suppressed and the flame became planar. Depending on whether the *parametric* and the *acoustic* instability coexist or not, the secondary *parametric* instability may develop producing significant flame acceleration and the appearance of large organized pulsating cellular structures. If they do, the final stage of development is characterized by those coherent cellular structures transforming into incoherent flame surfaces fluctuations.
Therefore, gaseous mixtures prone to the *parametric* instability may suffer, in closed chambers, a very significant acceleration of the flame propagation velocity. Especially for lean mixtures, this increase of the combustion rate will be very considerable.

2 Analysis

The mathematical treatment of the *acoustic* and *parametric* instabilities is based on the work of Pelce and Clavin [5], who obtained an equation for a perturbed flame front in a gravitational field under the assumptions of high activation energy and large scale wrinkling. Based on those results, Searby and Rochwerger [3] managed to derive equations for the growth rate of the *acoustic* and *parametric* instability and were able to calculate the stability limits for both cases numerically. In such a formulation we may consider an infinitely thin flame propagating in vertical direction. The flame front is represented by the function $F(\mathbf{x},t)=0$ in a reference moving with the flame front. Small perturbations could be represented in the form $F(x,t)=F(t)exp(ikx)$. With the temporal part $F(t)=Y\ exp(\sigma t)$ the planar flame front will be stable with respect to perturbations for all growth rates fulfilling $Re(\sigma)<0$ and unstable otherwise.
Considering the linear stability problem, the second order differential equation (1), [6] describes the evolution of perturbations of a flame surface of small amplitude considering periodic monochromatic velocity fluctuations normal to the flame front,

$$AF_{tt} + U_L\ k\ BF_t + k\ g_a\ C_1 F - k\ \omega U_a cos(\omega t)\ C_1 F + U_L^2\ k^2\ C_2 F = 0, \tag{1}$$

for a gas of arbitrary characteristics. In this equation, A, B, C_1, C_2, are dimensionless coefficients which take the form,

$$A = 1 + (\theta-1)/(\theta+1)kL\left(Ma - \theta J/(\theta-1)\right), \qquad B = 2\theta\left(1+\theta kL(Ma-J)\right)/(\theta+1), \tag{2}$$

$$C_1 = (\theta-1)\left(1 - kL\left(Ma - J\theta/(\theta-1)\right)\right)/(\theta+1), \tag{3}$$

$$C_2 = \theta(\theta-1)\left(-1 + kL\left((3\theta-1)Ma - 2J\theta + 2Prh_b(\theta-1) - I(2Pr-1)\right)/(\theta-1)\right)/(\theta+1). \tag{4}$$

where U_l is the laminar burning velocity, g_a the acceleration of gravity, U_a the velocity of the oscillating acoustic velocity field, T_u, T_b, ρ_u, ρ_b the temperatures and densities of the unburned and burned gases. Those coefficients depend on the following one-step Arrhenius reaction parameters of the flame, $\theta=(T-T_u)/(T_b-T_u)$, $\gamma=(\rho_u-\rho_b)/\rho_u$, $h(\theta)=\rho(\theta)\chi(\theta)/(\rho_u\chi_u)$ where χ is the thermal diffusion. Additionally, the Markstein number in its Pelce and Clavin [5] definition, is given by

$$Ma = J/\gamma - 0.5Ze(Le-1)\int_0^1 \left(h(\vartheta)ln(\vartheta)\right)/\left(1+\vartheta\gamma/(1-\gamma)\right)d\vartheta. \tag{5}$$

with the integrals

$$H = \int_0^1 (h_b - h(\vartheta))\,d\vartheta,\ J = \gamma/(1-\gamma)\int_0^1 h(\vartheta)/\left(1+\vartheta\gamma/(1-\gamma)\right)d\vartheta,\ I = (\theta-1)\int_0^1 h(\vartheta)\,d\vartheta. \tag{6}$$

Equation (1) may be transformed for a more convenient treatment. The change of variables $\alpha=A$, $\beta=U_LkB$, $\psi=kg_aC_1+U_L^2k^2C_2$ and $\delta=k\omega U_aC_1$ allow writing the equation (1) as

$$\alpha F_{tt} + \beta F_t + \left(\psi - \delta cos(\omega t)\right)F = 0. \tag{7}$$

whose solution, as stated by Searby and Rochwerger [3], is of the kind

$$F = Y(z)e^{-\kappa z}e^{iky}, \tag{8}$$

where the new variables are defined as $z=1/2\omega t$, $\kappa=\beta/(\omega\alpha)$, $a=4\alpha\psi-\beta^2\omega^2$ and $q=2\delta\omega^2\alpha$. Substituting (8) in (7), a simpler differential equation for the variable Y appears,

$$Y'' + \left(a - 2q\cos\left(2z\right)\right)Y = 0, \tag{9}$$

which is known as the Mathieu equation. An extensive analysis of the solutions of the Mathieu equation may be found i.e. in [7]. Whittaker's method [8] can be selected to obtain its solutions. The methodology considers a solution for the Mathieu equation of the type,

$$Y(z) = e^{i\upsilon z}\sum_{k=-\infty}^{+\infty} c_{2k}e^{i2kz}. \tag{10}$$

Substituting this in (9), after some manipulation (details can be found in the references [9] and [10]) it is found that to be a solution, the variable υ should fulfill the condition

$$\cos\left(\pi\upsilon\right) = 1 - \Delta(0,a,q,k)\left(1 - \cos\left(\pi\sqrt{a}\right)\right) \tag{11}$$

where $\Delta(0,a,q,k)$ is the determinant of a $(2k+1)_x(2k+1)$ tridiagonal matrix with $\Delta(0,a,q,k)_{ii}=1$, $\Delta(0,a,q,k)_{i(i-1)}= \Delta(0,a,q,k)_{i(i+1)}=\gamma_{2(k-i)}$ with $\gamma_{2k}=q(2k-\upsilon)^2-a$. The Sträng method [10] allows calculating $\Delta(0,a,q,k)$ with the recursion formula, $\Delta(0,a,q,k)=\beta_{2k}\Delta(0,a,q,k-1)-\alpha_{2k}\beta_{2k}\Delta(0,a,q,k-2)+ +\alpha_{2k}\alpha^2_{2(2k-1)}\Delta(0,a,q,k-3)$ with $\alpha_{2k}=\gamma_{2k}\gamma_{2(k-1)}$ and $\beta_{2k}=1-\alpha_{2k}$ and q,a real numbers. This implies that γ_{2k} is real for all k natural, so that $\Delta(\upsilon,a,q,k)$ is also a real number. Additionally, a is also real and so $\cos(\pi a^{1/2})=\cosh(\pi|a|^{1/2})$ if $a<0$ which means that the cosine remains always real. We may nevertheless study the solutions of υ, $\upsilon=u+wi$. Thus, $\cos(\pi(u+wi))=\cos(\pi u)\cosh(\pi w)-i\sin(\pi u)\sinh(\pi w)$ and then $\cos(\pi u)\cosh(\pi w)-i\sin(\pi u)\sinh(\pi w)= 1-\Delta(0,a,q,k)(1-\cos(\pi a^{1/2}))$. From the previous considerations, it is possible to separate into real and complex parts to obtain

$$\cos(\pi u)\cosh(\pi w) = 1 - \Delta(0,a,q)\left(1 - \cos\left(\pi\sqrt{a}\right)\right), \quad \sin(\pi u)\sinh(\pi w) = 0. \tag{12}$$

The solutions of the complex part are $u=0+k\pi$, or $w=0$. So, υ is purely real or complex. Therefore, the solutions of the real part in (12) are, if $w=0$, $u=\pi^{-1}\operatorname{acos}(1- \Delta(0,a,q,k)(1-\cos(\pi a^{1/2})))$, and if $u=0+k\pi$, $\cosh(\pi w)=\pm(1-\Delta(0,a,q,k)(1-\cos(\pi a^{1/2})))$ and so, w is found to be

$$w = \pm\frac{1}{\pi}\operatorname{acosh}\left(\left|1 - \Delta(0,a,q)\left(1 - \cos\left(\pi\sqrt{a}\right)\right)\right|\right). \tag{13}$$

Thus, if $|1-\Delta(0,a,q,k)(1-\cos(\pi a^{1/2}))|\le 1$, υ (see eq. (10)) is real, and otherwise complex. Re-calling the definition of the solution and substituting (10) in (8)

$$Y(z) = e^{-wz}e^{iuz}\sum_{k=-\infty}^{+\infty} c_{2k}e^{i2kz} \Rightarrow F = e^{-\kappa z}e^{iky}Y(z) = e^{(-w-\kappa)z}e^{i(uz+ky)}\sum_{k=-\infty}^{+\infty} c_{2k}e^{i2kz}, \tag{14}$$

that is stable in the case $-w<\kappa$. Due to the plus-minus solution of w a necessary condition for the stability is that,

$$|w| < \kappa \Rightarrow \pi^{-1}\operatorname{acosh}\left(\left|1 - \Delta(0,a,q)\left(1 - \cos\left(\pi\sqrt{a}\right)\right)\right|\right) < \beta/\left(\omega\alpha\right), \tag{15}$$

a condition that we anticipate could be used later for an enhanced analysis of the stability.

3 Particularization for H_2-air mixtures in normal conditions

At this point, it would be convenient in order to fix ideas and draw initial conclusions, to particularize the equations appearing in the previous section to obtain results for H_2-air mixtures at normal conditions. Due to the wide flammability limits of hydrogen-air mixtures [11], an evaluation of the stability characteristics of this gas with respect to the *acoustic* and *parametric* instability is very significant. The meaningfulness is accentuated by the fact that the emphasis on the very significant investigations that have until now experimentally analyzed the *acoustic* and *parametric* instability have been dedicated to hydrocarbons, i.e., propane [2] and methane [4][12], and only recently data has become available for hydrogen [13]. From the theoretical point of view no monographic study has been yet devoted to this gas. Recall that the wide flammability limits of hydrogen-air mixtures leads to a range of concentrations of practical interest which covers conditions in which $Le<1$ $Ma<0$ (lean) as well as *Le>1 Ma>0* (rich) resulting in the widest possible extent (for 7.5% and 60 % vol H_2-Air mixtures respectively: Ma=-2.6/1.5; Le= 0.3/2.3; Ze=5.5/3.7; U_L=0.05/1.8 m/s; flame thickess = 53 10^{-5}/4.4 10^{-5} m).

The frequency of the acoustic perturbation ω is a free parameter of our system. Due to the limited space of this extended summary only a single excitation frequency, $\omega=1000$ Hz has been considered for the analysis. Figure 1 contains the results of the application of the method for fuel concentrations of 7.5, 12.5, 15., 30., 45., 60. vol % H_2 at normal conditions The diagrams represent the growth rate in a color scale for different combinations of flame surface wavenumbers (abscissa) and reduced velocities (ordinate). Only positive growth rates are plotted. The stable regions (pairs $(k,U_a/U_l)$ in which the growth rate σ is less than 0) are plotted in violet. Black lines separate stable regions from unstable. The diagram corresponding to 7.5% H_2 (upper left) shows no separation between the *acoustic* and *parametric* instability. Inside the figure corresponding to 12.5 % H_2 the *acoustic* instability (appears at the bottom left corner, still with a low growth rate) and the *parametric* instability (appearing on the left of the figure) are already clearly identifiable. The plots corresponding to 15 and 30 vol % H_2 show the appearance of the *acoustic* instability region with a growing intensity. Moreover, they depict a reduction of the overlap between the *acoustic* and the *parametric* instability. In the picture related to 45 vol % H_2, a horizontal stripe free of instability appears for intensities U_a/U_l of around 4.5. This band grows with an increased fuel concentration as it is shown in the plot for 60% vol H_2. The absence of overlapping between both instabilities not only prevents the spontaneous developing of the *acoustic* instability in the *parametric* one but tends to the suppression of the DL instability (as higher U_a/U_l values produce negative growth rates). It is also interesting to underline the differences appearing in Figure 1 in the growth rate between the *acoustic* and the *parametric* instability. These differences are remarkable for lean and rich mixtures where the ratio between them reaches two orders of magnitude.

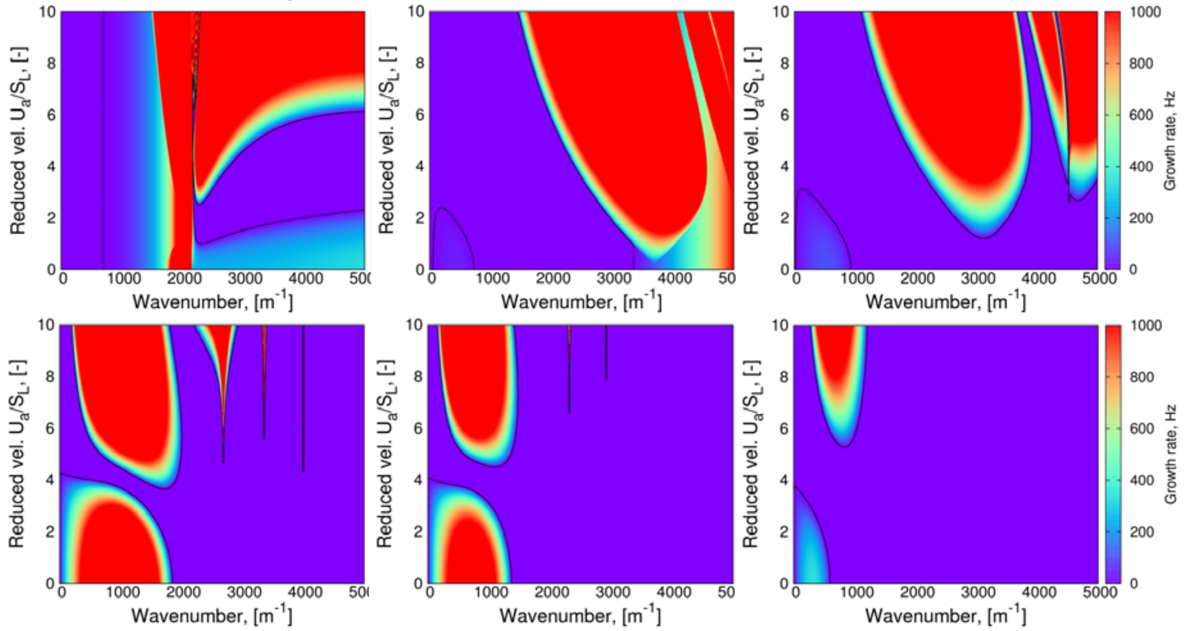

Figure 1. Stability graphs for H_2-air mixtures at normal conditions. Excitation frequency of 1000 Hz. From left to right and from top to bottom 7.5, 12.5, 15, 30, 45, 60 vol % H_2, corresponding to diagrams (a) to (e)

3 Existence of instability for all intensities of acoustic perturbation

The analytic results summarized in the stability condition (15) allow postulating the existence of a range of wavenumbers in which the flame surface results to be instable for all intensities of acoustic perturbation.

The solutions of the stability problem in which the variable κ is negative characterize a significant phenomenon. Clearly, the stability condition represented by the equation (15) cannot be fulfilled. As a result, these gaseous mixtures are unstable independently of the intensities of the amplitude of the perturbation, U_a. Due to this characteristic, the flames in which κ is negative may be prone to couple with the perturbations produced by waves, reflections, etc. with adequate frequency suffering significant accelerations and increasing its riskiness.

Let us study the conditions in which κ can be negative, and thus, re-call the definition of $\kappa=U_L kB(k)/\omega A(k)$. The variables U_L, k and ω are always positive. A and B (2) can be positive or negative depending on the values of Ma, J, θ, L, and k. The conditions in which $sign(A)\neq sign(B)$ denote the domain we are looking for (instability). Because of the nature of the integrand in (5), the Markstein number can be written as

$$Ma = \theta J/(\theta-1) + 0.5Ze(Le-1)\left|\int_0^1 h(\vartheta)ln(\vartheta)/(1+\vartheta(\theta-1))d\vartheta\right|. \tag{16}$$

Applying (16) to the definitions of A and B, equation (2), and writing in a more convenient form, the zeros of A and B, k_{0A} and k_{0B} can be immediately found to be

$$k_{0A} = -\left((\theta-1)L0.5Ze(Le-1)\left|\int_0^1 h(\vartheta)ln(\vartheta)/\left(1+\vartheta(\theta-1)\right)d\vartheta\right|/(\theta+1)\right)^{-1}, \tag{17}$$

$$k_{0B} = -\left(\theta L\left(\int_0^1 h(\vartheta)/\left(1+\vartheta(\theta-1)\right)d\vartheta + 0.5Ze(Le-1)\left|\int_0^1 h(\vartheta)ln(\vartheta)/\left(1+\vartheta(\theta-1)\right)d\vartheta\right|\right)\right)^{-1}, \tag{18}$$

where the condition $k=k_{0A}$ represents resonance.

From equations (17) and (18), $Le-1<0,$ results to be a necessary condition for k_{0A}, $k_{0B},$to be both positive. If $Le-1<0$ implies k_{0A} positive anyway and two cases are possible k_{0B} also positive or not. In any case, for $k>k_{0A}$ the variable A is always negative resulting in unstable conditions for an interval of wavenumbers that is henceforth determined. In the former case, in which both k_{0A}, k_{0B} are positive, there will exist an interval of wavenumbers, suppose $k_{0A}<k_{0B}$, in which $\forall k\in (k_{0A}, k_{0B})$ the flame is unstable. For the latter case, if only k_{0A} is positive, the interval of instability will extend to $k\in (0, k_{0A})$.or to $k\in (k_{0A}, \infty)$. This is shown in Figure 2, which contains the particularization of the variables k_{0A}, k_{0B} for H_2-air mixtures. For mixtures under 21.% vol H_2 both k_{0A}, k_{0B} are positive while for richer mixtures only k_{0A} is positive.

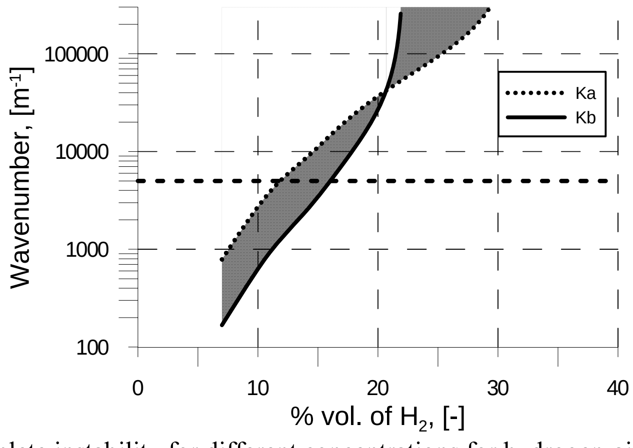

Figure 2. Bands of complete instability for different concentrations for hydrogen-air mixtures under normal conditions.

The original formulation of Pelce and Clavin [5] for perturbed flame fronts contains the assumption of large scale wrinkling in its derivation. Therefore, the analysis performed here should be restricted to wavenumbers $k<<2\pi/L$ (bellow the horizontal dashed line in Figure 2). For the cases in which both k_{0A}, $k_{0B}<<2\pi/L$ or at least one of them fulfills the inequality, the wavenumber range of instability resulting on the form $0<k_{0A}<k<k_{0B}<<2\pi/L$ or $0<k_{0A}<k<<2\pi/L$ is a characteristic of the gaseous mixture. Particularly, it is independent of the value of the excitation frequency ω. Additionally, κ depends inversely on the frequency of the perturbation.

These findings allow us to re-interpret the information contained in the Figure 1. The upper left (7.5 vol % H_2) diagram, it is represented the situation in which both $k=k_{0B}$ (left) and $k=k_{0A}$ (right, resonance) lines are visible (vertical lines). For lean mixtures, the range of instability is centered in relatively low wavenumbers making the band completely included in the region with physical sense (represented). In the figure corresponding to 12.5 % H_2 only a small part of the $k=k_{0B}$ line is visible. The resonance lays in higher wavenumbers than the ones shown already in the region $k<2\pi/10L$.

For those gaseous mixtures in which the resonance exists, k_{0A} positive, and has physical significance, $k_{0A}<<2\pi/L$, it represents a mechanism of instantaneous flame turbulization. The fact that for $k=k_{0A}$ the growth rate is infinite suggests, in analogy with Hooke's Law, the existence of a missing term in the derivation of equation (1) that will limit the growth rate and constitute a limitation in the validity of the analysis performed. The probability that the resonance lying in the interval $(0, 2\pi/10L)$ is physically meaningful would be enhanced for mixtures with a higher temperature and pressure, as L is inversely proportional to these magnitudes.

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