Influence of Lewis Number and Expansion on Jet Ignition

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1   Introduction

Jet ignition, consisting in ignition during the early transient part of the release process of a high pressure gaseous fuel such as hydrogen into the atmosphere, was first studied by Wolański & Wójcicki [1], although there are earlier anecdotal reports [2]. The phenomenon has an ambiguous role in hydrogen safety, clearly increasing risks in certain circumstances, but potentially lowering the risk of detonation in other situations. Experimental evidence mentioned for instance in a review by Astbury & Hawksworth [2] shows the size of the release opening to be a crucial parameter, with ignition being more likely for larger openings. While there are a number of possible explanations for the observations, potentially the most likely is that the combined effect of the heat and mass diffusion processes and chemistry, at the initially sharp contact surface, that separates expanded hence cooled hydrogen, and compressed, hence hot air, leads to ignition. Even for an axissymmetric configuration, the shock structure is complex [3]. Realistic models will require resolving both the shock structure, and the diffusive phenomena; early numerical attempts have been performed [4] however the problem remains at the limit of current computational capability because of the range of scales of interest. However, locally, the process approaches a planar geometry, which will be meaningful as long as the radius is large compared with the thickness. For constant pressure and planar geometry, this process was analyzed by Liñán & Crespo [5] for unity Lewis number, single step Arrhenius kinetics, in the limit of high activation energy. The case of Lewis number less than one was briefly touched in an appendix by Liñán and Williams [6]. For constant pressure, ignition always occurs; the first part of the analysis identifies the ignition time. Here, that analysis is extended to account for the expansion due to the evolution of the shock structure, and Lewis numbers both smaller and greater than unity are considered. However, it has been proposed [7] that expansion due to the multidimensional nature of the shock structure, which will lower

temperature hence slowing down the chemistry, may play a role in defining a critical regime. The analysis shows that ignition can only take place for Lewis numbers ≤ 1. It then determines the time scale on which expansion must take place and the ignition time; a critical value of the expansion rate is identified, separating conditions in which ignition is inhibited and conditions in which ignition will take place. For Lewis numbers greater than unity, an even significantly slower expansion will unconditionally inhibit ignition.

2 Formulation

The formulation follows closely from that of Liñán & Crespo [5]. The physical model includes conservation of mass, momentum and energy. The former leads to introducing a mass-weighed spatial coordinate, while the second leads to pressure being spatially uniform, given that in a frame of reference instantaneously attached to the diffusion region, Mach numbers are small. As a result, the expansion, absent in Liñán & Crespo's model, is also spatially uniform expansion, given that the diffusive length is small compared with the length scales associated with the shock structure. Initially, there is little chemistry thus the solution is reduced to the diffusion process: this is the (chemically) frozen solution. The ignition time is defined as the time required until a temperature perturbation of the order of the inverse activation energy goes to infinity, assuming it does; otherwise ignition is inhibited.

In the absence of expansion, the frozen flow solution is self-similar, but expansion, being time-dependent, sets a time hence a length scale. Leaving these unspecified yet, the planar problem reduces to the energy equation, and diffusion equations for fuel and oxidant.

Under the assumption that diffusivities are inversely proportional to the square of density, using a density weighed spatial coordinate x related to the original coordinate $\hat{x}$ by

$$x = \frac{1}{\rho\sqrt{\alpha}} \int_0^{\hat{x}} \rho d\hat{x} \qquad (1)$$

in which α is the heat diffusivity, and denoting the Lewis numbers as Le_O and Le_F, respectively for oxidant and for fuel, the problem is reduced to diffusion equations, for temperature and mass fraction, with sources due to chemistry and, in the energy equation, for the pressure fluctuation. Then using the similarity variable $\eta = x/2\sqrt{t}$ of the homogeneous problem, the dimensionless frozen problem becomes

$$4t\frac{\partial y_F}{\partial t} - 2\eta\frac{\partial y_F}{\partial \eta} = \frac{1}{Le_F}\frac{\partial^2 y_F}{\partial \eta^2} \qquad (2)$$

$$4t\frac{\partial y_O}{\partial t} - 2\eta\frac{\partial y_O}{\partial \eta} = \frac{1}{Le_O}\frac{\partial^2 y_O}{\partial \eta^2} \qquad (3)$$

$$4t\frac{\partial T}{\partial t} - 2\eta\frac{\partial T}{\partial \eta} = \frac{\partial^2 T}{\partial \eta^2} + \frac{4t(\gamma-1)T}{\gamma p}\frac{dp(t/t_0)}{dt} \qquad (4)$$

in which y_O is the oxidant mass fraction. y_F is the fuel mass fraction, T is temperature, Le is the Lewis number, p is a dimensionless pressure, and t_0 is the ratio of the time scale associated with expansion to the yet arbitrary time scale otherwise used. At $t = 0$, $p = 1$. Boundary conditions for $\eta \to -\infty$ are $y_O \to 0$, $y_F \to 1$ and $T \to T_F$. For $\eta \to -\infty$ the boundary conditions are $y_O \to 1$, $y_F \to 0$ and $T \to T_O$. Initial conditions match the boundary conditions. In the absence of expansion, the solution is self-similar and only depends upon time through η. However, accounting for the pressure term, one finds

$$y_O = \frac{1 + \mathrm{erf}(\eta\sqrt{Le_O})}{2}, \quad y_F = \frac{1 - \mathrm{erf}(\eta\sqrt{Le_F})}{2}, \quad T = \left\{ T_O + \frac{(T_O - T_F)[\mathrm{erf}(\eta) - 1]}{2} \right\} p^{(\gamma-1)/\gamma} \qquad (5)$$

3 Perturbation

In order to determine if ignition will occur, perturbations of the order of the inverse activation energy are considered, using the notation y'_O, y'_F and T'. The similarity variable is rescaled by $[1 - \mathrm{erf}(\eta)]/2 = \epsilon\xi/(1 - T_F/T_O)$, in which $\epsilon = T_O/T_a << 1$, with T_a being the activation temperature. This places the focus close to the hot end of the diffusion region, where temperature only departs from the hot side temperature by a perturbation of the order of the inverse activation energy; this is the region in the flow where chemistry is fastest and where oxidant concentration is largest. However relatively little fuel is present, which slows down chemistry although not enough to balance the effect of temperature. Using the rescaled coordinate, one finds that for ξ of order unity, the frozen flow solution becomes

$$y_O = 1 - \frac{(\eta\sqrt{\pi})^{Le_O - 1}}{2\sqrt{Le_O}} \left(\frac{2T_O\epsilon\xi}{T_O - T_F} \right)^{Le_O}, \quad Y_F = \frac{(\eta\sqrt{\pi})^{Le_F - 1}}{2\sqrt{Le_F}} \left(\frac{2T_O\epsilon\xi}{T_O - T_F} \right)^{Le_F} \tag{6}$$

$$T = T_O(1 - \epsilon\xi)p^{(\gamma-1)/\gamma} \tag{7}$$

Thus if $Le_F = 1$, the contribution to the fuel and temperature perturbations due to advection-diffusion are both at order ϵ so that Liñan's formulation is recovered. However, if $Le_F < 1$, the fuel concentration due to diffusion in the region where temperature has only dropped from the hot side temperature by order ϵ is then of order ϵ^{Le_F}; this is larger than consumption by chemistry, which becomes negligible. For $Le_F > 1$, the opposite occurs; the fuel that diffusion brings in is still at order ϵ^{Le_F}, but this is now of magnitude smaller than ϵ, i.e. what chemistry at order ϵ would require; since it is impossible to burn fuel that is not present, chemistry is limited to order ϵ^{Le_F}, although in the Arrhenius exponential, the temperature drop of order ϵ due to diffusion remains. Clearly, different formulations are required for these three different cases.

In all three cases, when setting the time scale so as to balance chemistry and diffusion, the transient term becomes small compared with diffusion. As a result, the perturbation equations are reduced to a quasi-steady formulation, in which time now only appears as a parameter. After elimination, and neglecting smaller order terms, for $Le_F - 1 = O(-1/\log \epsilon)$, the perturbation of order ϵ, denoted as T' and y'_F, becomes [5]:

$$\frac{\xi^2}{t} \frac{d^2 T'}{d\xi^2} = -\left[\xi + l\left(1 - \frac{T_F}{T_O} \right) y'_F \right]^b \exp\left(T' - \alpha t - \xi \right) \tag{8}$$

$$Q\frac{\xi^2}{t} \frac{d^2 y'_F}{d\xi^2} = \left[\xi + l\left(1 - \frac{T_F}{T_O} \right) y'_F \right]^b \exp\left(T' - \alpha t - \xi \right) \tag{9}$$

in which the temperatures T_O and T_F refer to the respective initial values, Q is the heat release, b is the reaction order for fuel, and $\epsilon^{Le_F - 1} = 1/l$ with l of order unity. Pressure has been assumed to drop slowly with respect to time scaled by τ, such that

$$p = 1 - \epsilon\alpha t, \quad \alpha = -\frac{\gamma - 1}{\gamma} \frac{dp}{dt} \tag{10}$$

so that $\alpha > 0$ when pressure drops.

Adding Eqs. (8) and (9), one finds that $T' + Qy'_F = 0$. One defines $\beta = c_p(T_O - T_F)l/Q$, i.e. the ratio of the hot air temperature to the adiabatic flame temperature of the stoichiometric mixture initially at the cold fuel temperature, which in the current context should be < 1. Finally, introducing the time parameter $\Delta = t \exp -\alpha t$, the problem becomes:

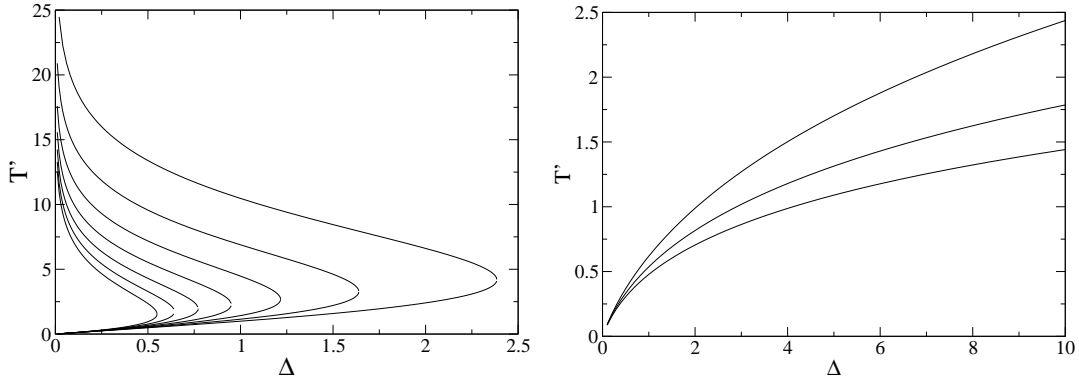$$-\frac{\xi^2}{\Delta} \frac{d^2 T'}{d\xi^2} = (\xi - \beta T')^b \exp\left(T' - \xi \right) \tag{11}$$

Figure 1: Left: Solution T' for $\xi \to \infty$ to Eq. (11) for β from 0 to 0.6 at intervals 0.1, from left to right. Critical values of Δ correspond to the turning point, at the right end of each curve, where the two branches meet. Right: $\beta = 1.1$, 1.2 and 1.4. $b = 1$.

For $Le_F < 1$ with $Le_F - 1$ of order unity, a similar procedure yields the equation

$$\frac{-\xi^{2-bLe_F}}{\Delta}\frac{d^2T'}{d\xi^2} = \exp\left(T' - \xi\right) \tag{12}$$

Finally, for $Le_F > 1$ and with a departure of order unity, the perturbation of order ϵ^{Le_F} becomes

$$\frac{-\xi^2}{\Delta}\frac{d^2T''}{d\xi^2} = (\xi - \beta T'')^b \exp -\xi \tag{13}$$

In all cases, time has been scaled by

$$\tau = \left(1 - \frac{T_F}{T_O}\right)^{bLe_F}\frac{(2\eta^*)^{2+b(1-Le_F)}Le_F^{b/2}\epsilon^{2-bLe_F}}{\pi^{b(Le_F-1)/2}Qk}\exp\frac{1}{\epsilon} \tag{14}$$

η^* being defined as the solution to

$$\eta^{*2} = -\log\frac{2\eta^*\epsilon\sqrt{\pi}}{1 - T_F/T_O} \tag{15}$$

In all three cases, boundary conditions are $T'(0) = 0$ for $\xi = 0$, since far away no fuel is present thus no chemistry occurs and from matching to the main diffusion zone, $dT'/d\xi \to 0$ for $\xi \to \infty$.

In all cases, a second order ordinary differential equation in ξ needs being solved, in which time only appears as a parameter. If the rate of pressure change is order unity, then chemistry becomes exponentially small and ignition will not occur. If it is smaller than ϵ then its effect is negligible, the problem is reduced to either the problem of Liñán & Crespo [5] for $Le = 1$, or similar problems. The critical regime is identified when taking the rate of pressure decay to be of order ϵ.

Equation (11) is precisely the same equation obtained by Liñán & Crespo [5], but for a somewhat broader definition of Δ which is no longer simply equal to time but equals $t \exp -\alpha t$.

In all three cases, a two point boundary value problem in which time appears as a parameter needs being solved numerically; a shooting method is used. Because of the singularity at $\xi = 0$, the equivalent system of first order ordinary differential equations does not satisfy a Lipschitz condition. A local linearization allows
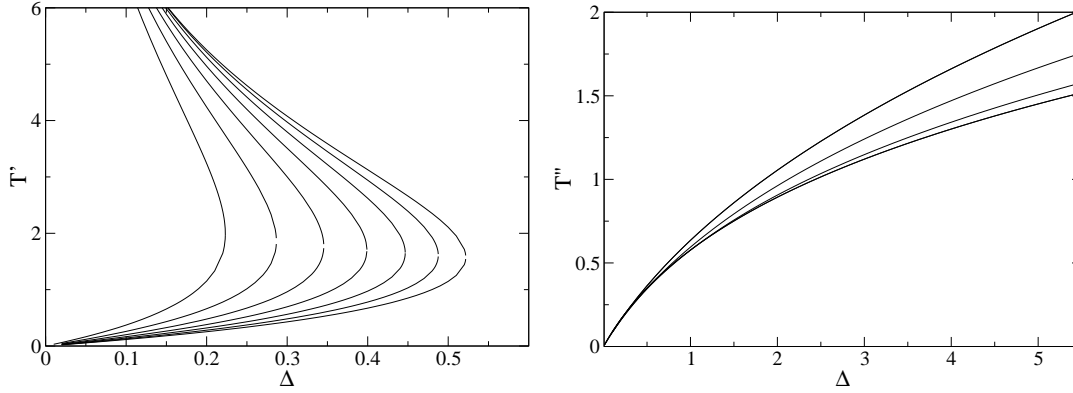
Figure 2: Solutions T' for $\xi \to \infty$ to Eqs. (12) and (13), left, for Le_F from 0.3 to 0.9 at intervals 0.1, from left to right. Right, $Le_F = 1.1, 1.2, 1.4$ and 1.6 from bottom to top. $b = 1$.

eliminating one integration constant, that multiplies a mode that grows large for $\xi \to 0$. A shooting method or an iterative technique adjusts the second integration constant in the linearized formulation.

As shown in [5] and in Fig. 1, for $\beta < 1$, the problem of Eq. (11) admits two solutions for $\Delta < \Delta^*$ and no solution for larger values. The upper branch has no physical meaning [5]. For $\Delta \to \Delta^*$, the scaling leading to Eq. (11) breaks down and chemistry becomes very fast, indicating ignition. In the absence of expansion, it follows that ignition occurs at $t = \Delta^*$. As similar situation occurs with respect to the fuel Lewis number, as Fig 2 shows. For $Le_F < 1$, Eq. (12) admits two solutions for Δ below the turning point, at a critical value Δ^*, behind which no solution exists. In contrast, for $Le_F > 1$, single solutions exist for all values of Δ, no matter how large.

In contrast with cases in which pressure stays constant, here $\Delta(t) = t \exp(-\alpha t)$ experiences a maximum value $\Delta_{\max}(\alpha) = 1/\alpha e$, for $t = 1/\alpha$. For higher values of Δ there is no time satisfying $\Delta = t \exp -\alpha t$. Thus, for $Le_F < 1$ or for $\beta < 1$ if $Le_F = 1$, ignition will occur if $\Delta^*(Le_F)$ or $\Delta^*(\beta) \leq 1/\alpha e$, the critical value α^* of the expansion rate being then given by

$$\alpha^* = \frac{1}{e\Delta^*} \tag{16}$$

the dimensional value of which being then readily calculated by reintroducing the time scale τ from Eq. (14).

For an expansion faster than this critical value, ignition does not occur. Likewise, for $Le_F > 1$ or for $\beta > 1$ if $Le_F = 1$, expansion will always eventually lead to a distributed drop of temperature larger than the inverse activation energy before the ignition process is complete. (The process is described in detail by Liñán and Crespo [5] for the latter case, but similar for $Le_F > 1$.)

The analysis shows that the reaction rate peaks close to the hot air side of the diffusion layer. For fuel Lewis number close to unity, the rate at which fuel is consumed is of the same order as the rate at which it is supplied by diffusion. For smaller Lewis numbers, consumption is negligible to supply by diffusion so that temperature becomes limiting. however, for larger Lewis numbers, chemistry could consume more fuel than diffusion provides, resulting in much slower chemistry, leading to a negligible temperature increase.

In the context of jet ignition, there is a clear relationship between the size of the leak and the expansion rate, with a larger hole leading to a slower expansion. Thus the current result provides an explanation for the experimental observation whereby larger hydrogen leaks are more likely to ignite [1, 2, 8].

Moire importantly, however, the analysis results in a very different behavior for small and large fuel Lewis number, with a critical value

$$Le_{F,\text{critical}} = 1 - \frac{\log[c_p(T_O - T_F)/Q]}{\log \epsilon} \tag{17}$$

such that for smaller Le_F, a well-defined ignition behaving as a thermal explosion will take place in finite time, the value of which is determined by the analysis, as long as the rate of expansion stays below a critical value. For larger Le_F, however, a slower and more progressive process takes place, that is eventually and unconditionally quenched by expansion. This provides a fundamental explanation as to why jet ignition has only been observed for hydrogen.

Acknowledgments

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