THE CRUCIAL ROLE OF THE LEWIS NUMBER IN JET IGNITION

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ABSTRACT

During the early phase of the transient process following a hydrogen leak into the atmosphere, a contact surface appears separating hot air from cold hydrogen. Locally, the interface is approximately planar. Diffusion occurs, potentially leading to ignition. This process was analyzed by Liñán and Crespo (1976) for Lewis number unity and Liñán and Williams (1993) for Lewis number less than unity. In addition to conduction, these processes are affected by expansion due to the flow, which leads to a temperature drop. If chemistry is very temperature-sensitive, then the reaction rate peaks close to the hot region, where relatively little fuel is present. Indeed the Arrhenius rate drops rapidly as temperature drops, much more so than fuel concentration. However, the small fuel concentration present close to the air-rich side depends crucially upon the balance between fuel diffusion and heat diffusion, hence the fuel Lewis number. For Lewis number unity, the fuel concentration present due to diffusion is comparable to the rate of consumption due to chemistry. If the Lewis number is less than unity, fuel concentration brought in by diffusion is large compared with temperature-controlled chemistry. For a Lewis number greater than unity, diffusion is not strong enough to bring in as much fuel as chemistry would be able to burn, and combustion is controlled by fuel diffusion. In the former case, combustion occurs faster, leading to a localized ignition at a finite time determined by the analysis. As long as the temperature drop due to the expansion associated with the multidimensional nature of the jet does not lower significantly the reaction rate up to that point, ignition in the jet takes place. For fuel Lewis number greater than unity, first, the reaction rate is much lower. Second, chemistry does not lead to a defined ignition. Eventually, expansion will affect the process and ignition does not take place. In summary, it appears that the reason why hydrogen is the only fuel for which jet ignition has been observed is a Lewis number effect, coupled with a high speed of sound hence a high initial temperature discontinuity.

1. INTRODUCTION

Spontaneous ignition of hydrogen jets was first studied by Wolański & Wójcicki [1], although [2] there are reports of earlier observations. Recent work includes experiments [3, 4] and simulations [5, 6, 7]. Potential role of expansion due to shock multidimensionality leading to a temperature drop has been suggested [7]. However, as discussed in detail by Astbury & Hawksworth [2], the exact mechanism resulting in ignition of transient hydrogen jets from a high pressure source remains to be determined.

Initially, the structure of the system is well described as a shock tube (Riemann) problem, if in a one-dimensional geometry [3] (i.e., inside a tube), or its multidimensional equivalent [8]. Chemistry needs fuel and oxidant to be present, at a location where temperature is high enough for the reaction rate to be significant. Thus one should expect that if ignition occurs, it will happen in the neighborhood of the contact surface produced by the shock tube problem, separating hydrogen cooled by expansion from the high pressure source from air warmed though the leading shock. For lower pressure ratios, this interface is Rayleigh-Taylor stable; however, in the planar shock tube problem, for higher pressure ratios, cold hydrogen becomes heavier than hot air on the other side of the contact surface, which is then unstable; even then, the density ratio remains close to 1 thus the instability is rather weak. In any event, the instability will affect the contact surface unevenly, with some regions largely unaffected.

The initially nearly planar interface between hot air and cold hydrogen evolves under the combined effects of diffusion and advection due to flow including instability. However, over the time window required for ignition (determined by the analysis), the thickness of the diffusion layer remains small compared with its radius. As a result, a one-dimensional model remains realistic and meaningful. An analysis by Liñán & Crespo [9], and by Liñán & Williams [10], focuses upon ignition precisely at such an interface. However their results will be affected by expansion due to the three-dimensional shock structure, which lowers temperature, potentially inhibiting ignition. The analysis reveals crucial differences in behavior depending upon the Lewis number. For Lewis number below a threshold close to unity, the value of which is precisely determined by the analysis, a mechanism similar to the classical thermal explosion mechanism of Frank-Kameneskii takes place, as long as the expansion rate is below a critical value, also determined by the analysis. However, if the Lewis number is above the threshold, initial ignition occurs as a slow process characterized by a front progressing from the warm air side into the fuel, which is eventually quenched by expansion.

Physically, since the reaction rates are very temperature-sensitive, then in the spontaneous ignition problem, ignition should take place very nearly in the hot air region. That hydrogen diffuses much more effectively than heat (i.e. a small Lewis number) is then crucial in bringing enough fuel to sustain the chemical reaction. (Likewise in the case of an oxidant with Lewis number smaller than unity, one might expect that a jet of oxidant from a high pressure source into fuel might result in jet ignition.)

The role of the Lewis number in extinction of stretched flames is well-known. However, to the authors' knowledge, its role in spontaneous ignition of hydrogen jets had not been identified previously.

2. PHYSICAL MODEL

In the structure of the flow behind a three dimensional expansion [8], the region of interest is what, in an inviscid and non-conducting large scale model, appears as a contact surface separating air heated by the leading shock from expanded hydrogen. Locally, diffusion will take place. As long as the thickness of the diffusion layer is small compared with the radius of curvature, the geometry remains approximately planar. It is convenient to introduce a frame of reference attached to the contact surface in the large scale problem, and to focus on the local small scale problem, taking diffusion into account. Because the shock weakens as it expands, this local frame is non-inertial, but one readily shows that the resulting pressure gradients are limited to a fraction of the mean pressure of the order of the Mach number based upon local velocities, which are small. Thus, momentum results in a pressure that is time-dependent but spatially uniform at leading order. The time dependency can then be obtained from the large scale problem; here it is taken to be an unspecified but known function of time. This is precisely the same approach as in Maxwell & Radulescu [7], who implemented a numerical solution, using a realistic kinetic scheme.

For the planar model to be realistic, there must be regions where the diffusion layer is not overly affected by Rayleigh-Taylor instability. At the pressures of interest, the behavior of hydrogen may depart from the ideal gas. Using a Noble-Abel model [11, 12, 13], the relationship between pressure and velocity across an expansion wave is unchanged in relation to results for ideal gas, and so is the pressure ratio. However, writing the Nobel-Abel equation of state as $p(1 - b\rho) = \rho RT$, the density ratio becomes

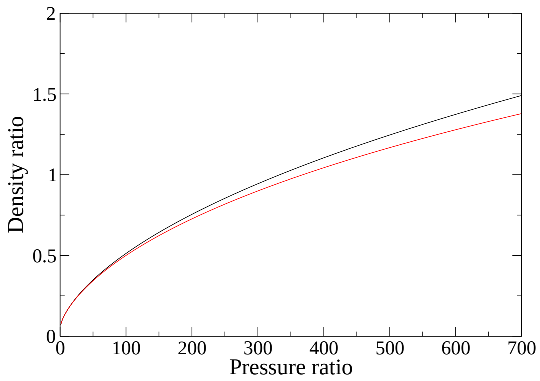

Figure 1: Density ratio across the contact surface in the one dimensional shock tube problem. Upper line, ideal gas law for both hydrogen and air; lower curve: hydrogen is a Noble-Abel fluid with b = 0.00775 m^3/kg, air is ideal gas.

$$\frac{\rho_{H_2}}{\rho_{air}} = \frac{R_{air}T_{air}}{R_{H_2}T_{H_2} + bp} \tag{1}$$

Figure 1 shows how the density ratio across the diffusion layer depends upon the pressure ratio. It shows that for higher pressures the ratio becomes larger than unity, so that the layer becomes unstable, and that even for large pressures, the ratio remains close to 1. Taking both hydrogen and air as ideal gases, the transition occurs for a pressure ratio of approximately 335, and 370 using a Noble-Abel model for hydrogen, which results in a density ratio of 1.38 for a pressure ratio 700. Otherwise, in the neighborhood of the contact surface, real gas effects should not be significant. For an analysis of the effect of curvature on the instability, see [8].

The problem is thus reduced to the formulation of Liñán & Crespo [9], but in an environment in which pressure is dropping. The physical model still includes conservation of mass, energy and species, namely fuel and oxidant. The kinetic model is assumed to be single step Arrhenius. As a result, the problem is described by dimensionless conservation laws:

$$\frac{\partial \rho}{\partial t} + \frac{\partial(\rho u)}{\partial x} = 0 \tag{2}$$

$$\rho\frac{\partial T}{\partial t} + \rho u\frac{\partial T}{\partial x} - \frac{\gamma - 1}{\gamma}\frac{dp}{dt} = \frac{\partial}{\partial x}\rho\alpha\frac{\partial T}{\partial x} + Q\dot{m} \tag{3}$$

$$\rho\frac{\partial y_O}{\partial t} + \rho u\frac{\partial y_O}{\partial x} = \frac{\partial}{\partial x}\rho D_O\frac{\partial y_O}{\partial x} - \nu\dot{m} \tag{4}$$

$$\rho\frac{\partial y_F}{\partial t} + \rho u\frac{\partial y_F}{\partial x} = \frac{\partial}{\partial x}\rho D_F\frac{\partial y_F}{\partial x} - \dot{m} \tag{5}$$

with

$$\dot{m} = k\rho y_O^a y_F^b \exp\frac{-T_a}{T} \tag{6}$$

Here the fluid has been taken as an ideal gas with constant specific heats. Mass fractions are y_F and y_O, respectively for fuel and oxidant. Thermodynamic state variables such as pressure p, density ρ, temperature T, and thermal diffusivity α have been scaled by some reference value (say, initial values in the hot air side), time by a yet to be specified time scale τ, the heat release Q, by c_p times the reference temperature, and the rate multiplier k, by $1/\tau$. The diffusion coefficients D_O and D_F have been scaled by the thermal diffusivity at the reference state. The coordinate x has been scaled by the square root of the product τ times the reference thermal diffusivity. ν accounts for the stoichiometric ratio. The Lewis numbers $Le_F = \alpha/D_F$ and $Le_O = \alpha/D_O$ are assumed constant, and diffusivity α is taken to be inversely proportional to the square of density. T_a is the activation temperature scaled by the reference temperature. Typically, T_a is quite large, so that the reaction rate becomes very temperature-sensitive. The solution below will be obtained specifically for $T_a >> 1$.

Boundary conditions for temperature and mass fractions correspond to the values associated with the contact surface in the large scale problem, taking the effect of expansion into account, respectively at $x \to \pm\infty$. Initial conditions at $t = 0$ correspond to a jump consistent with the contact surface. In effect, the problem has been reduced to a one-dimensional diffusion/advection/chemistry.

Before attempting to solve, it is useful to reduce the problem to a formulation that, in the absence of chemistry, is self-similar, introducing the similarity variable

$$\eta = \frac{1}{\rho\sqrt{\alpha t}}\int_0^x \rho dx \tag{7}$$

Then,

$$4t\frac{\partial y_F}{\partial t} - 2\eta\frac{\partial y_F}{\partial \eta} = \frac{1}{Le_F}\frac{\partial^2 y_F}{\partial \eta^2} - \frac{4tT}{p}\dot{m} \tag{8}$$

$$4t\frac{\partial y_O}{\partial t} - 2\eta\frac{\partial y_O}{\partial \eta} = \frac{1}{Le_O}\frac{\partial^2 y_O}{\partial \eta^2} - \frac{4tT\nu}{p}\dot{m} \tag{9}$$

$$4t\frac{\partial T}{\partial t} - 2\eta\frac{\partial T}{\partial \eta} = \frac{\partial^2 T}{\partial \eta^2} + \frac{4t(\gamma-1)T}{\gamma p}\frac{dp}{dt} + \frac{4tTQ}{p}\dot{m} \tag{10}$$

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.

This completes the formulation. Next, to determine whether ignition will occur, the following approach is implemented. So far, the time τ which has been used to scale time remains unspecified. It is clear that for sufficiently short times, the effect of chemistry will remain negligible. If the chemical source term is negligible, then the problem above admits a closed form solution, which is readily obtained.

Next, in order to determine whether ignition will take place, time will be scaled such that the chemical reaction contributes to only small temperature changes, such that temperatures will only increase by a fraction of the order of the inverse activation temperature T_a which is assumed to be large. This will allow to write the effect of chemistry as a perturbation added to the frozen flow solution. Finally, the behavior of the solution to the perturbation problem will determine whether ignition takes place.

3. CHEMICALLY FROZEN FLOW

If the chemical source term is neglected, and in the absence of expansion, the solution to the problem outlined in the previous section is self-similar. The solution only depends upon time through the similarity variable η and takes the form of an error function. The pressure drop term adds a particular solution to the homogeneous problem. Accounting for the pressure drop, one finds that the frozen solution is

$$y_{Of} = \frac{1 + \mathrm{erf}(\eta\sqrt{Le_O})}{2}, \quad y_{Ff} = \frac{1 - \mathrm{erf}(\eta\sqrt{Le_F})}{2} \tag{11}$$

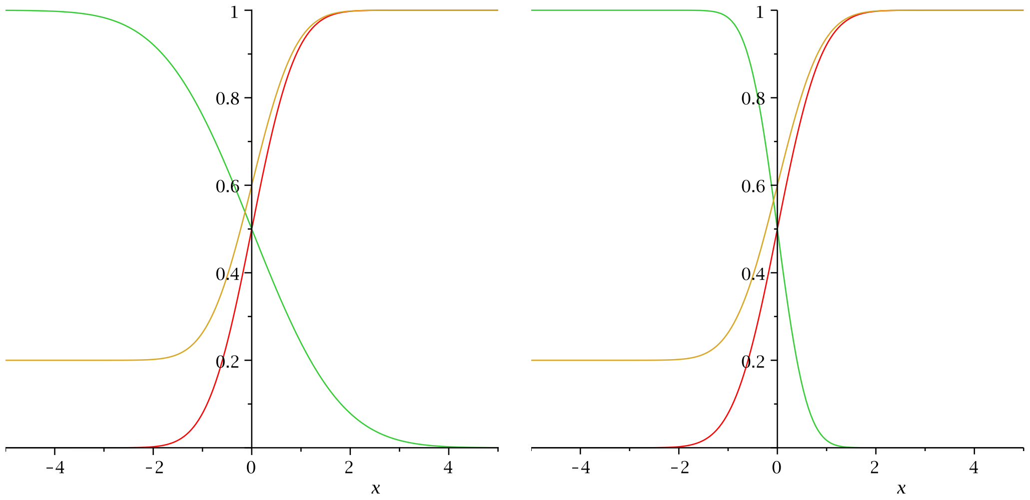

Figure 2: Frozen flow solution, vs. η, $T_O = 1$, $T_F = 0.2$, $Le_O = 1.0$. Left: $Le_F = 0.5$, right: $Le_F = 1.5$. Temperature goes from 0.2 on left to 1.0 on right, fuel from 0 to 1, oxidant from 1 to 0.

$$T_f = \left\{ T_O + \frac{(T_O - T_F)[\mathrm{erf}(\eta) - 1]}{2} \right\} p^{(\gamma-1)/\gamma} \tag{12}$$

Typical solutions to the diffusion problem are shown in Fig. 2. The effect of the Lewis number in the region, on the air side, where temperature is high enough to support chemistry, is quite clear; fuel concentration is significantly lower for the higher Lewis number. Figure 3 explores the consequence on local reaction rates. For a Lewis number 1.5, the peak in the rate is approximately 10% of the rate for $Le_F = 0.5$. On this basis alone, it becomes clear that the fuel mass diffusivity is important.

4. PERTURBATION DUE TO CHEMICAL REACTION

4.1. Approach

Next, focusing upon ignition, perturbations of the order of the inverse activation energy are considered, using the symbol $\epsilon = T_O/T_a << 1$ (ϵ is related to the reciprocal of the Zel'dovich number). The time scale τ such that chemistry affects the problem at order ϵ will be determined.

Ignition should occur in the region (in space-time) where the diffusion process above leads to temperature departing from the hot air value by a fraction of order ϵ. To resolve that region, where the independent similarity variable η becomes large, η needs to be rescaled, replacing it by the local (time-dependent) space variable ξ, of order unity in that region:

$$\frac{1 - \mathrm{erf}(\eta)}{2} = \frac{\epsilon\xi}{(1 - T_F/T_O)} \tag{13}$$

This is the region where chemistry is fastest and where oxidant concentration is very close to 1. However relatively little fuel is present, which slows down chemistry although possibly not enough to balance the effect of temperature. Using the rescaled coordinate ξ, one finds that the frozen flow solution becomes

$$y_{Of} = 1 - \frac{(\eta\sqrt{\pi})^{Le_O - 1}}{2\sqrt{Le_O}} \left(\frac{2T_O\epsilon\xi}{T_O - T_F} \right)^{Le_O} \tag{14}$$

$$y_{Ff} = \frac{(\eta\sqrt{\pi})^{Le_F - 1}}{2\sqrt{Le_F}} \left(\frac{2T_O\epsilon\xi}{T_O - T_F} \right)^{Le_F} \tag{15}$$

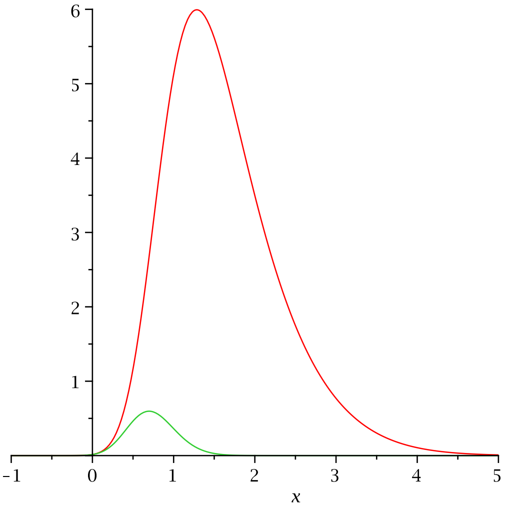

Figure 3: Reaction rates based upon the frozen flow solution vs. η, same parameters as in Fig. 2, $k\rho = 10^6$, $T_a = 10.$ and both a and $b = 1$. Higher curve: $Le_F = 0.5$, lower curve, $Le_F = 1.5$.

$$T_f = T_O(1 - \epsilon\xi)p^{(\gamma-1)/\gamma} \tag{16}$$

The latter equation shows, as expected, a temperature departing from T_O by order ϵ. As to mass fractions, y_O is close to unity. However, obviously, close to the air side, the fuel concentration is small, of order ϵ^{Le_F}. It is of note that **the magnitude** of the fuel concentration in the region of interest depends upon the Lewis number, being larger than ϵ, the magnitude of the temperature perturbation, if the Lewis number is smaller than unity, and otherwise if Le_F is greater than 1. Only for Le_F close to unity is the reactant concentration of order ϵ. Thus one needs to develop three different solutions for these three cases.

Under the high activation energy approximation, using the expansions $T = T_f + \epsilon T'$ and (further below) $y_F = y_{Ff} + \epsilon y'_F$, the exponential in the Arrhenius rate becomes

$$\exp\frac{-T_a}{T} = \exp\frac{-1}{\epsilon[(1-\epsilon\xi)p^{(\gamma-1)/\gamma} + \epsilon T']} = \exp\frac{-p^{-(\gamma-1)/\gamma}(1 + \epsilon\xi - \epsilon p^{-(\gamma-1)/\gamma}T')}{\epsilon[1 - \epsilon^2(\xi^2 - p^{-2(\gamma-1)/\gamma}T'^2)]} \tag{17}$$

If pressure drops by more than order ϵ over a time of order τ, then the magnitude of the reaction rate drops over a time of order τ, effectively quenching any incipient chemistry so that ignition will not take place. Likewise, if pressure drops less than by order ϵ over a time of order τ, the process is no longer affected by the pressure drop. A balance will thus occur for pressure changing by order ϵ over a time τ, thus one writes $p = p(\epsilon t)$. If pressure was scaled by its initial value so that $p(0) = 1$, using Taylor series,

$$p^{-(\gamma-1)/\gamma} = 1 - \epsilon t\frac{\gamma-1}{\gamma}\left.\frac{dp}{dt}\right|_0 = 1 + \epsilon\sigma t \tag{18}$$

The symbol σ, with positive value when pressure drops, is defined as

$$\sigma = -\frac{\gamma-1}{\gamma}\left.\frac{dp}{dt}\right|_0 \tag{19}$$

Neglecting contributions of higher order in ϵ, the Arrhenius exponent becomes

$$\exp\frac{-T_a}{T} = \exp\frac{-1}{\epsilon}\exp(T' - \xi - \sigma t) \tag{20}$$

The constant factor $\exp -1/\epsilon$ will be absorbed in the time scale τ, as defined below. The temperature perturbation T', the effect of diffusion ξ, and expansion σt all contribute at the same order. The former results in an increasing reaction rate while the latter two slow it down.

If $Le_F = 1$, Liñán's formulation is recovered [9]. In all three cases ($Le_F < 1$, close to 1 or > 1), as in Liñán, when setting the time scale so as to balance chemistry and diffusion, the transient term becomes small compared with diffusion. As a result, in all three cases the perturbation equations are reduced to a quasi-steady formulation, in which time only appears as a parameter. Likewise, anticipating results below, the expression for the time scale τ leading to a balance between diffusion and reaction is also the same in all cases:

$$\tau = \left(1 - \frac{T_F}{T_O}\right)^{bLe_F} \frac{(2\eta^*)^{2+b(1-Le_F)} Le^{b/2} \epsilon^{2-bLe_F}}{\pi^{b(Le_F-1)/2} Qk} \exp \frac{1}{\epsilon} \tag{21}$$

in which the factor $\exp 1/\epsilon$ is due to the Arrhenius rate. The large number η^* is defined as the root of

$$\eta^{*2} = -\log \frac{2\eta^* \epsilon \sqrt{\pi}}{1 - T_F/T_O} \tag{22}$$

When ξ is of order unity, η is obviously large -see Eq. (13)- while η^* only differs from η by order unity, allowing for replacement of the variable η by the constant value η^*, resulting in smaller order differences, which can be neglected.

4.2. Le_F close to unity

One readily verifies that if $Le - 1 = O(-1/\log \epsilon)$ then the fuel concentration in the region where ξ is of order unity is of order ϵ. Thus one introduces a reduced Lewis number l, of order unity, defined by

$$\epsilon^{Le_F - 1} = 1/l \tag{23}$$

After elimination, setting τ as per Eq. (21), the value providing for balance between diffusion and reaction, the perturbation equations of order ϵ respectively to energy and fuel equations, become [9]:

$$\frac{1}{4\eta^2} \frac{\partial T'}{\partial t} + \frac{\xi^2}{t} \frac{\partial^2 T'}{\partial \xi^2} = -\left[\xi + l\left(1 - \frac{T_F}{T_O}\right) y'_F\right]^b \exp\left(T' - \sigma t - \xi\right) \tag{24}$$

$$\frac{1}{4\eta^2} \frac{\partial y'_F}{\partial t} + \frac{\xi^2}{t} \frac{\partial^2 y'_F}{\partial \xi^2} = \frac{1}{Q}\left[\xi + l\left(1 - \frac{T_F}{T_O}\right) y'_F\right]^b \exp\left(T' - \sigma t - \xi\right) \tag{25}$$

Since $\eta >> 1$, the first (transient) term in Eqs. (24) and (25) is negligibly small compared with the other terms, and the problem is reduced to the quasi-steady formulation

$$\frac{\xi^2}{t} \frac{d^2 T'}{d\xi^2} = -\left[\xi + l\left(1 - \frac{T_F}{T_O}\right) y'_F\right]^b \exp\left(T' - \sigma t - \xi\right) \tag{26}$$

$$\frac{\xi^2}{t} \frac{d^2 y'_F}{d\xi^2} = \frac{1}{Q}\left[\xi + l\left(1 - \frac{T_F}{T_O}\right) y'_F\right]^b \exp\left(T' - \sigma t - \xi\right) \tag{27}$$

These are ordinary differential equations in ξ in which time only appears as a parameter. They are combined as follows. Adding Eqs. (26) and (27), one finds that $T' + Qy'_F = 0$. One defines β as 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:

$$\beta = \frac{c_p(T_O - T_F)l}{Q} \tag{28}$$

Finally, introducing the time parameter

$$\Delta = t \exp -\sigma t \tag{29}$$

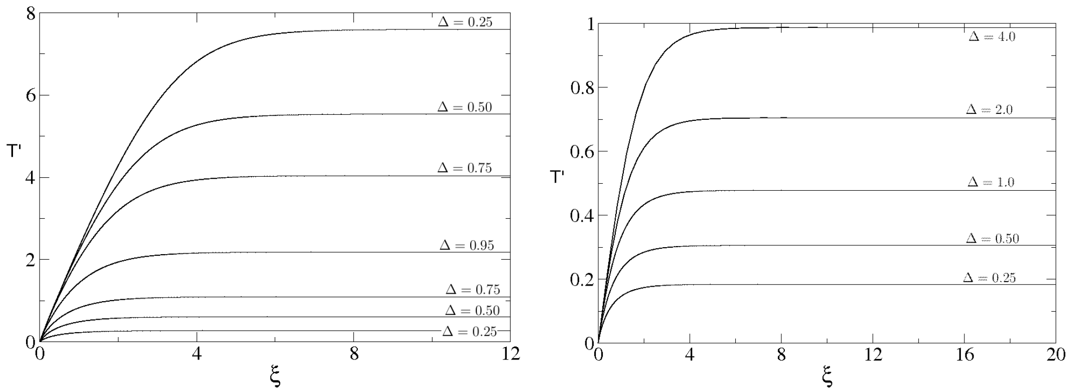

Figure 4: Solutions to the perturbation equation, Eq. (30). Left, for $\beta = 0.3$; right, for $\beta = 1.8$; $b = 1$.

the problem becomes:

$$\frac{-\xi^2}{\Delta}\frac{d^2T'}{d\xi^2} = (\xi - \beta T')^b \exp(T' - \xi) \tag{30}$$

This is precisely the same equation obtained by Liñán and Crespo [9], although the symbols Δ and β are now more broadly defined, accounting for the expansion in Δ which in Liñán and Crespo was simply time, and for the departure from unity of the Lewis number, in β. Boundary conditions are $T'(0) = 0$ for $\xi = 0$ (which corresponds to $x \to \infty$), 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 $\xi \to \infty$.

The problem has been reduced to a second order two point boundary value problem in ξ, in which time only appears through parameter Δ. This problem calls for a numerical solution. It is readily transformed into a two dimensional first order problem that is solved using a shooting method. However, because of the presence of the factor ξ^2 on the left hand side, the resulting problem does not satisfy a Lipschitz condition at $\xi = 0$ so that one needs to use a perturbation in order to implement initial conditions, eliminating one integration constant, which multiplies a mode that grows large for $\xi \to 0$ which is inconsistent with the initial condition. The same solution as in Liñán and Crespo [9] is recovered. For each set of data, .i.e. for given values of β and Δ, the solution increases monotonically from $T' = 0$ at $\xi = 0$, approaching a limit $T'(\xi \to \infty)$ as ξ becomes large, as shown in Fig. 4. The behavior for large ξ is consistent with the right boundary condition, which imposes the slope to approach zero.

These solutions look similar in all cases. However, on the left plot, for $\beta < 1$, the curves, which initially moved higher for higher Δ, eventually move lower, while on the right, for $\beta > 1$, the values of $T'(\xi \to \infty)$ continue growing as Δ increases. This difference becomes more apparent when plotting values of $T'(\xi \to \infty)$, vs. Δ, as shown in Fig. 5.

As shown in [9] and in Fig. 5, for $\beta < 1$, the solution to the problem of Eq. (30) changes significantly between the case of $\beta < 1$ and $\beta > 1$. For $\beta < 1$, the figure on the left shows that two solutions exist for values of the time parameter Δ below a critical value Δ^* that depends upon β, and no solution is found for larger values of Δ. The upper branch has no physical meaning [9]. For $\Delta \to \Delta^*$, the scaling leading to Eq. (30) breaks down since close to the turning point Δ^* the rate at which the solution depends upon Δ, hence on time becomes large. Thus chemistry becomes very fast, since the rate of growth of the perturbation becomes infinite. This indicates that ignition has taken place.

For $\beta < 1$, in the absence of expansion, it follows that ignition occurs unconditionally at $t = \Delta^*$. Including expansion, however, the relationship between Δ and time is now related by Eq. (29). As shown on Fig. 6, for a given value of σ, for Δ below a maximum value Δ_{max}, there are two times

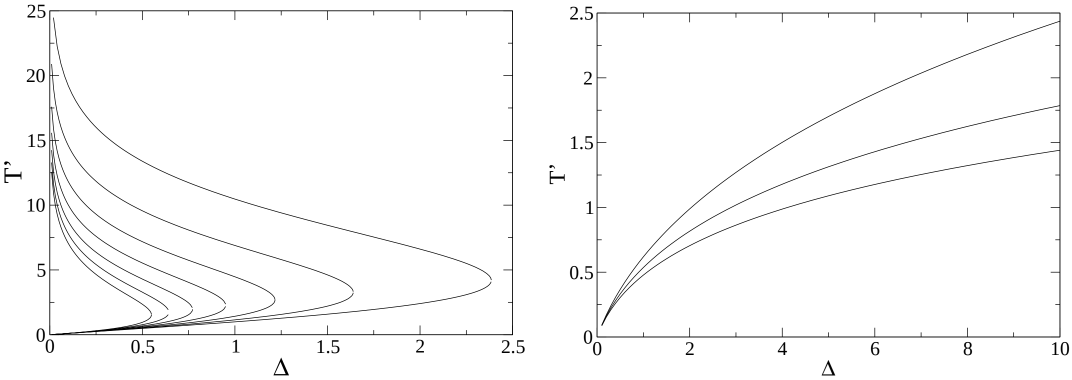

Figure 5: Case Le_F close to unity. Left: Solution T' for $\xi \to \infty$ to Eq. (30) for β from 0 to 0.6 at intervals 0.1, from left to right. Right: $\beta = 1.1$, 1.2 and 1.4. $b = 1$.

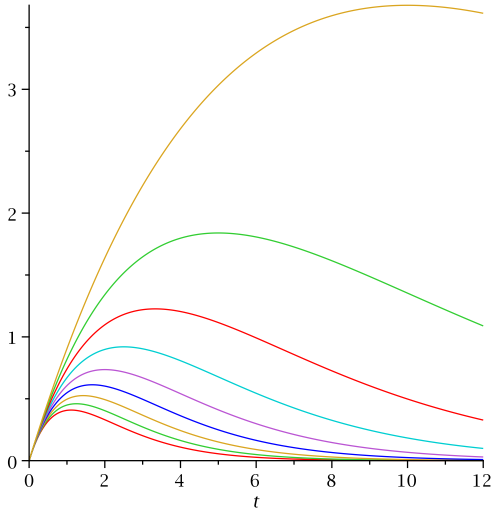

Figure 6: Relationship between Δ and time, for $\sigma = 0.9$, 0.8, 0.7, 0.6, 0.5, 0.4, 0.3, 0.2 and 0.1 from bottom to top

yielding the same value of Δ. Only the smallest one is physically meaningful, given that the process proceeds in time starting at $t = 0$. However for $\Delta > \Delta_{max}$, there is no longer any time associated with Δ. The maximum value, obtained for $t = 1/\sigma$, is $\Delta_{max} = 1/e\sigma$. As $t \to 1/\sigma$, the rate at which Δ increases with time becomes zero. Thus the reaction becomes progressively slower and effectively stops.

It is thus found that if $\beta < 1$, for ignition to take place, i.e. for Δ to reach Δ^*, it must be that $\Delta^* \leq \Delta_{max}$. Thus the critical expansion rate for a given value of $\beta < 1$ is

$$\sigma^*(\beta) = \frac{1}{e\Delta^*(\beta)} \tag{31}$$

For $\beta > 1$ however, a different situation occurs. The plot on the right in Fig. 5 shows that, as Δ increases, $T'(\infty)$ now increases monotonically, although the rate of increase is monotonically decreasing. On the right side of Eq. (30), the quantity $\xi - \beta T'$ to the power b, corresponds to fuel mass fraction, which vanishes when the fuel is fully burnt, i.e. at equilibrium. Such an equilibrium, i.e. $T' = \xi/\beta$, is a solution to Eq. (30); it satisfies the boundary condition at $\xi = 0$ (which corresponds to $x \to \infty$) but not the boundary condition toward the diffusion zone. The right plot in Fig. 4 shows a solution approaching equilibrium for ξ close to zero, and growing as Δ, hence time, increases. This behavior corresponds to a combustion front moving progressively from the air side into the diffusion layer, with no runaway ignition ever taking place at a finite Δ. However, in the presence of expansion, this process will end

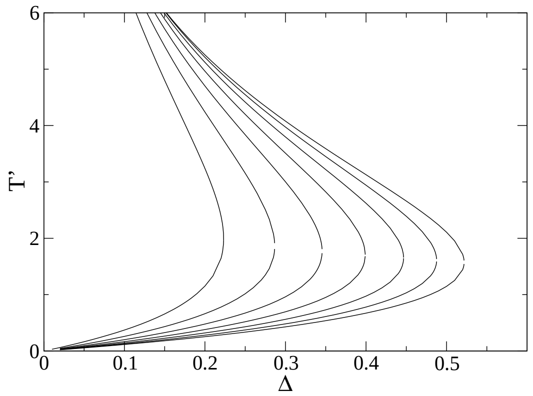

Figure 7: $Le_F < 1$. Solutions T' for $\xi \to \infty$ to Eq. (33), for Le_F from 0.3 to 0.9 at intervals 0.1, from left to right. $b = 1$.

unconditionally when $\Delta \to \Delta_{max}$, i.e. at time $t = 1/\sigma$, beyond which ignition does not proceed.

In summary, the analysis shows that for $\beta > 1$, i.e. for a fuel Lewis number above a critical value Le_F^*, ignition will not occur, even if expansion is very small. This critical value is given by

$$Le_F^* = 1 - \frac{\log c_p(T_O - T_F)/Q}{\log \epsilon} \tag{32}$$

However, if the fuel Lewis number is below that threshold, ignition will take place as long as the expansion rate is below the critical value $1/e\Delta^*(\beta)$.

4.3. Le_F smaller than unity

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, the rate at which diffusion bring fuel into the ignition zone is, according to Eq. (15) of order ϵ^{Le_F}, which is now much larger than the rate at which temperature-limited chemistry consumes fuel, of order ϵ. Thus now consumption becomes negligible compared to supply. Thus the factor representing the role of fuel concentration in the reaction rate simply becomes $\xi^{Le_F b}$ times a constant absorbed in the time scale. A procedure similar to the one above for Le_F of order unity yields the equation

$$\frac{-\xi^{2-bLe_F}}{\Delta}\frac{d^2T'}{d\xi^2} = \exp\left(T' - \xi\right) \tag{33}$$

This result is mentioned briefly in an appendix in [10]; conditions of validity and implications are only briefly discussed.

Boundary conditions remain the same as above. Again, a numerical solution is obtained, yielding the results for $T'(\xi \to \infty)$ shown in Fig. 7.

Here, the results are similar to the case above (Le_F close to unity), for $\beta < 1$. Ignition will take place as long as the expansion rate is below a critical value, given by the same expression as above. However, the time scale, given by Eq. (21), decreases rapidly with Le_F. Thus a similar dimensionless σ^* will actually correspond to a much stronger actual expansion, and ignition happens significantly earlier for fuels with smaller Lewis number. This is the case most relevant to hydrogen jet ignition. The numerical value for the time scale based upon Eq. (21) are comparable with results from numerical simulation [5, 6, 7].

4.4. Le_F greater than unity

The formulation above assumed chemistry to take place at order ϵ, entailing consumption of fuel at that order. However if the fuel Lewis number is greater than unity, diffusion is no longer strong enough to

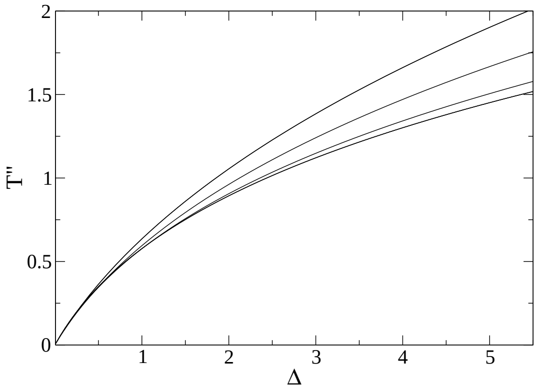

Figure 8: $Le_F > 1$. Solutions T' for $\xi \to \infty$ to Eq. (34), $Le_F = 1.1$, 1.2, 1.4 and 1.6 from bottom to top. $b = 1$.

provide that concentration, but only values of order ϵ^{Le_F}. Since the reaction cannot consume fuel that is not present, although temperatures continue departing from the hot side temperature by order ϵ, the reaction rate is now limited by fuel concentration, to a temperature increase that is negligible compared with the drop due to diffusion and expansion.

As a result, one needs to consider perturbations at order ϵ^{Le_F}, limited by fuel concentration, denoted as T" and y". Using the same approach as above, a quasi-steady equation is obtained in which the effect of temperature increase due to the reaction in the Arrhenius term is now negligible:

$$\frac{-\xi^2}{\Delta}\frac{d^2T"}{d\xi^2} = (\xi - \beta T")^b \exp -\xi \tag{34}$$

Again, 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 this case again, solutions to the problem of Eq. (34) for $T'(\xi \to \infty)$ are single-valued and a solution exists for all values of Δ, no matter how large, just as in the case for Le_F close to unity, when $\beta > 1$. This case is clearly not conducive to ignition since eventually the effect of expansion will quench the chemistry.

5. CONCLUSION

The current analysis focused upon ignition in the diffusion layer that appears at the interface between shock-heated air and hydrogen cooled by expansion during the transient phase of the jet ignition process. Results show the process to be quite sensitive to the fuel Lewis number. If the fuel Lewis number is less than a critical value close to unity, then as long as the rate of expansion due to the multidimensional shock topology remains below a specific threshold determined by the analysis, ignition takes place as a runaway mechanism. The analysis also determines the finite ignition time, with numerical value comparable with numerical simulation results [5, 6, 7]. If the fuel Lewis number is larger than the critical value, instead of a runaway ignition, a reaction front moves from the warm air side into the diffusion region, but no matter how slow, expansion eventually quenches the front in finite time. The critical Lewis number value for which transition between these two regimes occurs was also determined.

The significance of the fuel Lewis number is due to the following. If chemistry is very temperature sensitive, then the reaction rate peaks close to the hot air side of the diffusion layer, where there is ample oxygen to support ignition, but very little hydrogen, supplied from the cold side by mass diffusion. The Lewis number is the ratio heat diffusion/mass diffusion. A fuel with low Lewis number, such as hydrogen, will diffuse much faster, resulting in a significantly higher concentration at the location where temperature is close to the hot side temperature, where ignition is expected to occur. 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 compared with supply by diffusion, which is large compared with the inverse activation temperature. However, for larger Lewis numbers, chemistry could consume more fuel than diffusion provides, resulting in much slower chemistry; the temperature increase due to the reaction is then small compared with the drop due to diffusion and expansion.

Advective mixing such as turbulence and/or Rayleigh-Taylor instability might be of some help in igniting fuels with **high Lewis numbers**. However, advection affects equally heat and mass diffusion, resulting in a turbulent Lewis number equal to unity. Thus in hydrogen jets, turbulence will likely hamper ignition rather than promoting it, and ignition will likely occur along quiescent sections of the diffusion layer.

Furthermore, 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 as to why only hydrogen jets appear to ignite, in contrast with hydrocarbon fuels, and also confirmation of an explanation [7] of the experimental observation whereby larger hydrogen leaks are more likely to ignite [1, 2].

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