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Flame balls in mixing layers

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ABSTRACT

We present a study of flame balls in a two-dimensional mixing layer with one objective being to derive an ignition criterion (for triple-flames) in such a non-homogeneous reactive mixture. The problem is formulated within a thermo-diffusive single-reaction model and leads for large values of the Zeldovich number β to a free boundary problem. The free boundary problem is then solved analytically in the asymptotic limit of large values of the Damköhler number, which represents a non-dimensional measure of the (square of the) mixing layer thickness. The explicit solution, which describes a non-spherical flame ball generalising the classical Zeldovich flame balls (ZFB) to a non-uniform mixture, is shown to exist only if centred at a single location. This location is found to be precisely that of the leading-edge of a triple-flame in the mixing layer, and typically differs from the location of the stoichiometric surface by an amount of order β^{-1} depending only on a normalised stoichiometric coefficient Δ .

The thermal energy of the burnt gas inside the flame ball is used to derive an expression for the minimum energy E_{min} (of an external spark say) required for successful ignition. In particular, it is found that the presence of the inhomogeneity increases E_{min} compared to the homogeneous case. For a stoichiometrically balanced mixture, corresponding to $\Delta = 0$, the relative increase in the ignition energy is found to be proportional to β^2/Da , i.e. to the square of the Zeldovich number and to the reciprocal of the Damköhler number Da . More generally, for arbitrary value of Δ , the minimum ignition energy is found to correspond to that of the Zeldovich flame ball in a uniform mixture at the local conditions prevailing at the location of the leading edge of the triple-flame, plus a positive amount depending on Δ which is again proportional to β^2/Da . In short, the analysis provides a possible criterion for successful ignition in a non-homogeneous mixture by determining the minimum energy required (E_{min}) and the most favourable location (that of the leading-edge of a triple-flame) where it should be deposited.

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1. Introduction

In order to ignite a reactive mixture by means of an external source such as an electric spark, the energy of the source must exceed a minimum critical value. In homogeneous premixed mixtures, theoretical studies are available relating this critical value to the thermal energy contained inside a non-propagating spherical flame known as Zeldovich flame ball (ZFB), which is a stationary solution of the reaction–diffusion equations. In non-uniform reactive mixtures on the other hand, there seems to be no theoretical studies in the literature addressing the same ignition problem and describing analytically the corresponding non-propagating solutions. These non-spherical solutions, generalising ZFBs to

non-homogeneous mixtures, are the subject of the current investigation and we shall also refer to them as flame balls.

In homogeneous premixed reactive gases, flame balls were predicted by Zeldovich about seventy years ago as time-independent spherically symmetric solutions of the heat conduction and diffusion equations [1]. An important feature of these solutions is that they are unstable under adiabatic conditions [1,2] and that additional physical mechanisms need to be taken into account for them to be stable. Such stabilising mechanisms have been found to include volumetric heat-loss [3,4], conductive heat-loss to walls [5] or some weakly non-uniform flow fields [6]. In fact, much of the recent theoretical work on flame balls has been motivated by the observation of such apparently stable structures in lean hydrogen–air mixtures in the experiments by Ronney and coworkers under micro-gravity conditions [7,8]. More recently, ZFBs have been shown to be a special case of stationary spherical flames, termed *generalised flame balls* by Daou et al. [9], surrounding a point

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source/sink of hot inert gas. The existence and stability of these flames have been investigated in [9], and it was found, in particular, that they can have positive, zero or negative burning speeds, with zero speeds characterising ZFBs. It is worth noting that, in addition to the significance of flame balls as a possible mode of combustion, such as for drifting flame balls [10–12], they are also significant in ignition problems involving heat addition by an external source [1,2,13–15]. In this context in its most fundamental form, they may indeed serve to estimate the minimum energy to be deposited by the source for successful ignition whereby an initially formed hot kernel generates an outwardly propagating premixed-flame front [1].

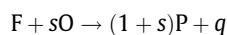
In a non-homogeneous reactive mixture, such as the two-dimensional mixing layer between non-premixed reactants considered in this study, successful ignition by an external source should also lead to flame fronts propagating away from the source, as observed numerically in [16]. Given that the mixture varies from fuel-lean to fuel-rich conditions across the mixing layer, the fronts are expected to be those of two triple-flames travelling in opposite directions. Such triple-flames, first observed experimentally by Philips [17], are now well-understood combustion structures due to several dedicated studies which followed the pioneering analytical work by Ohki and Tsuge [18] and Dold and coworkers [19,20]. Indeed, several aspects of triple-flames have been to date investigated including gas-expansion [21–24], preferential diffusion [25,26], heat losses [27–29], reversibility of the chemical reaction [30,31] and the presence of a parallel flow [32,33]. We shall not discuss here the relatively vast literature on triple-flames, but refer the interested reader to the review paper [34] or to [35], for further references. We would like to point out, however, that despite the wealth of investigations available, there seems to be no analytical studies dedicated to the ignition of triple-flames based on an extension of the concept of ZFBs to reactive mixing-layers. Some important results with some relevance to our investigation can be found in the literature nevertheless. For example, in the context of edge-flames (in premixed and non-premixed mixtures), two dimensional structures termed flame tubes, flame strings or 2D-spots depending on the author, have been identified in two-dimensional counterflows, both experimentally [36] and numerically [37–40]. These structures although resembling ZFBs are also different given that their existence hinges on the presence of strain, as mentioned in [38] where it is also argued that the cellular instability of the planar flame may be at their origin. Of more direct relevance to this paper are two numerical studies by Jackson and Buckmaster [41] and Lu and Ghosal [42]. The first study is carried out in a two-dimensional unstrained mixing layer, a configuration to be adopted in this paper and that we have used in two previous publications on triple-flames [32,33]. The second study is conducted in an strained mixing layer corresponding to an axisymmetric counterflow configuration. Two types of combustion structures are addressed in [41], namely *flame isolas*, finite-size regions of burning surrounded by a non-reacting mixture similar to the flame balls investigated in this paper, and *flame holes*, regions of local extinction on a diffusion flame discussed previously in [43]. The authors of [41] describe the expansion or shrinkage of these structures and the dependence of the propagation speed of their edges on the Damköhler number and their instantaneous radius. Similar structures are investigated in [42], termed *flame discs* and *flame holes*, with particular focus on their temporal evolution and dynamics. Among the findings, a critical hole (disc) radius depending on the strain rate is determined which corresponds to non-propagating holes (discs). In particular, the critical disc radius is found numerically to be an increasing function of the strain rate, and this is argued to indicate that a minimum source energy is required for successful ignition in a mixing layer. We shall provide an explicit formula for such minimum ignition energy valid in the

framework of our asymptotic study. Before proceeding, it is worth mentioning that our study differs from [41,42] in several important aspects which include the following: (1) It is asymptotic and analytical. (2) the stoichiometric coefficient is an important parameter in our study and not fixed; this is important since the flame balls/discs/isolas are not necessarily centered at the location of the diffusion flame, as we shall see, contrary to the stoichiometrically balanced case considered in [41,42]. (3) Our problem is posed as a free boundary problem and in particular we do not use the one-dimensional diffusion flame to provide boundary conditions for our flame balls as done in [41], may be as an approximation, in order to anchor the flame at one boundary.

The paper is organised as follows. We begin by describing the thermo-diffusive model adopted and formulating the corresponding problem in Section 2. This is followed by an asymptotic analysis which is based on a compact reformulation of the problem derived in Section 3 in the limit of infinitely large activation energy of the chemical reaction. The reformulated problem is shown to reduce to a neat free boundary problem which is solved analytically in Sections 4 and 5. The results and their physical implications are discussed in Section 6 with concluding remarks given in Section 7.

2. Formulation

We consider a reactive mixture in a channel of width $2L$ extending to infinity along the X-direction and Y-direction, the latter being perpendicular to the plane of the figure; see Fig. 1. The walls of the channel are assumed to be porous and the concentrations of fuel and oxidizer are maintained fixed at the walls. This setup has been used in previous publications such as [32,33,41]. Although such a configuration may be difficult to achieve experimentally, it is adopted here in order to construct a simple theoretical model for flame balls in a mixing layer. The combustion is represented by a single irreversible one-step reaction of the form



where F denotes the fuel, O the oxidizer and P the products. The quantity s denotes the mass of oxidizer consumed and q the heat released, both per unit mass of fuel. We consider a thermo-diffusive approximation with constant density and constant transport properties. The governing equations in dimensional form can be written in the form

$$\frac{\partial T}{\partial t} = D_T \Delta T + \frac{q}{c_p} \frac{\tilde{\omega}}{\rho} \quad (1)$$

$$\frac{\partial Y_F}{\partial t} = D_F \Delta Y_F - \frac{\tilde{\omega}}{\rho} \quad (2)$$

$$\frac{\partial Y_O}{\partial t} = D_O \Delta Y_O - s \frac{\tilde{\omega}}{\rho}. \quad (3)$$

Here T , Y_F and Y_O are respectively the temperature and the mass fractions of the fuel and oxidizer. In addition, D_F , D_O , and D_T denote the diffusion coefficients of the fuel, the oxidizer, and heat respectively, and are taken to be constants. The quantities ρ and c_p denote the density and the heat capacity. The reaction rate $\tilde{\omega}$, defined as the mass of fuel consumed per unit volume and unit time, is assumed to obey an Arrhenius law

$$\tilde{\omega} = B \rho^2 Y_F Y_O \exp(-E/RT), \quad (4)$$

where B and E/R represent, respectively, the (constant) pre-exponential factor and the activation temperature.

The conditions as $|X^2 + Y^2| \rightarrow \infty$ correspond to the frozen solution independent of X and Y , which is given by

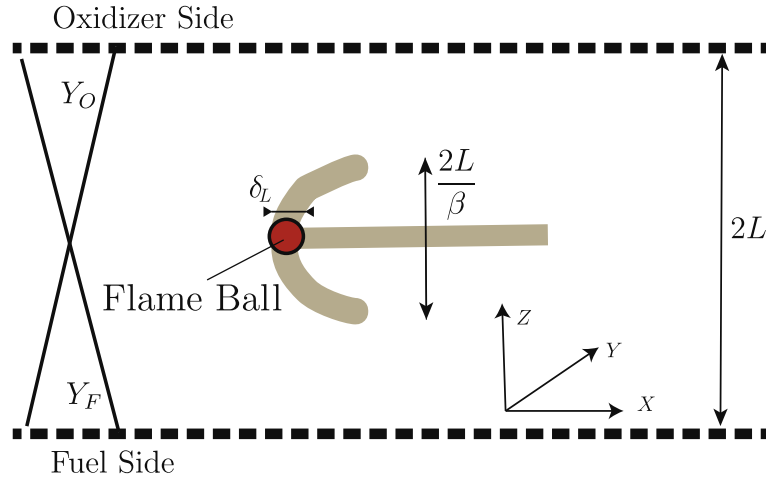


Fig. 1. A schematic illustration of a flame ball in a non homogeneous domain between two porous wall. The mass fractions are prescribed by $Y_F = Y_{F,F}$ and $Y_O = 0$ on the fuel side, and $Y_F = 0$ and $Y_O = Y_{O,O}$ on the oxidizer side.

$$T = T_u, \quad (5a)$$

$$Y_F = \frac{Y_{F,F}}{2} \left(1 - \frac{Z}{L}\right), \quad (5b)$$

$$Y_O = \frac{Y_{O,O}}{2} \left(1 + \frac{Z}{L}\right), \quad (5c)$$

where $Y_{F,F}$ and $Y_{O,O}$ refer to the mass fraction of the fuel side and the oxidizer side respectively, and T_u refers to the temperature on both sides as well as in the unburnt mixture; thus, the lateral boundary conditions are also given by (5) with $Z \rightarrow \pm L$ for all X and Y .

For large activation energies, the region which may be able to sustain significant heat generation is centred around the stoichiometric surface. In the frozen mixture far away from the flame ball, i.e. for $X^2 + Y^2 \rightarrow \infty$, this surface is located at $Z = Z_{st}$ where Z_{st} is determined from $Y_O = sY_F$ and Eqs. (5) to be

$$\frac{Z_{st}}{L} = \frac{S - 1}{S + 1}, \quad (6)$$

where $S \equiv sY_{F,F}/Y_{O,O}$ is a normalised stoichiometric coefficient (or a global equivalence ratio).

Next, we write the governing equations in terms of the scaled quantities

$$y_F = \frac{Y_F}{Y_{F,st}}, \quad y_O = \frac{Y_O}{Y_{O,st}}, \quad \theta = \frac{T - T_u}{T_{ad} - T_u}, \quad (7)$$

where the subscript 'st' indicates values at ($X^2 + Y^2 \rightarrow \infty, Z = Z_{st}$) and where $T_{ad} \equiv T_u + qY_{F,st}/c_p$ is the adiabatic flame temperature.

To non-dimensionalise the problem, we follow [26] and select as unit length $L_r \equiv L/\beta$, that is (half) the ratio between the mixing layer thickness and the Zeldovich number $\beta \equiv E(T_{ad} - T_u)/RT_{ad}^2$. This length scale represents the typical radius of curvature of the triple-flame (sketched in the figure) which can propagate in the mixing layer [26,32,33]; it is also, as we shall see later, the appropriate scale to measure the distance between the location where the flame balls are centered and the stoichiometric surface. As a unit time we choose the diffusion time L_r^2/D_T . Then, on substituting (7) into Eqs. (1)–(3), we obtain the non-dimensional problem

$$\frac{\partial \theta}{\partial t} = \Delta \theta + \epsilon_L^{-2} \omega \quad (8)$$

$$\frac{\partial y_F}{\partial t} = \text{Le}_F^{-1} \Delta y_F - \epsilon_L^{-2} \omega \quad (9)$$

$$\frac{\partial y_O}{\partial t} = \text{Le}_O^{-1} \Delta y_O - \epsilon_L^{-2} \omega, \quad (10)$$

in terms of the coordinates $x = \beta X/L$, $y = \beta Y/L$, and $z = \beta(Z - Z_{st})/L$. Here

$$\epsilon_L \equiv \frac{\delta_L}{L_r} = \frac{\beta D_T}{L S_L}, \quad (11)$$

where δ_L and S_L are the thickness and speed of the laminar planar stoichiometric flame which are related by $\delta_L = D_T/S_L$; thus ϵ_L represents the thickness of the planar stoichiometric flame measured with the reference length L/β . In fact ϵ_L is related to the Damköhler number Da that we define as the ratio between the diffusion time across the mixing layer L^2/D_T and the flame transit time δ_L^2/D_T by

$$Da \equiv \frac{L^2}{\delta_L^2} = \beta^2 \epsilon_L^{-2}. \quad (12)$$

Taking for S_L its leading order value for large β , namely,

$$S_L = \sqrt{\frac{4\text{Le}_F \text{Le}_O}{\beta^3} Y_{O,st} (\rho D_T) B \exp(-E/RT_{ad})}, \quad (13)$$

the non-dimensional reaction rate ω takes the form

$$\omega = \frac{\beta^3}{4\text{Le}_F \text{Le}_O} y_F y_O \exp\left(\frac{\beta(\theta - 1)}{1 + \alpha(\theta - 1)}\right), \quad (14)$$

where $\alpha = (T_{ad} - T_u)/T_{ad}$.

The boundary conditions are

$$\theta = 0 \quad (15a)$$

$$y_F = 1 - \gamma_F \frac{z}{\beta} \quad (15b)$$

$$y_O = 1 + \gamma_O \frac{z}{\beta}, \quad \text{as } x^2 + y^2 \rightarrow \infty, z \rightarrow -\frac{\beta}{\gamma_O} \text{ or } z \rightarrow \frac{\beta}{\gamma_F}, \quad (15c)$$

where

$$\gamma_F = \frac{1+S}{2} \quad \text{and} \quad \gamma_O = \frac{1+S}{2S}. \quad (16)$$

The problem now is fully formulated by Eqs. (8), with the boundary conditions (15). A main aim when tackling this problem is to find its stationary solutions (flame balls) and examine their stability. In particular, one would like to determine the domain of existence of these stationary solutions, and the corresponding profiles of θ, y_F , and y_O , in terms of $\text{Le}_F, \text{Le}_O, S, \epsilon_L, \beta$ and α . This can be carried out numerically in the general case. For the sake of an analytical treatment, however, we consider in this paper the problem in the limiting case $\beta \rightarrow \infty$ where a compact reformulation can be derived

and then solved analytically using perturbation methods for the determination of the stationary solutions. We shall restrict our focus on this task in the present study leaving a more general treatment, necessarily numerical, to further investigations. In particular, we shall not examine herein the stability of the stationary solutions or their temporal evolution to propagating triple-flames, nor the propagation of the triple-flames. In fact, triple-flame propagation in this configuration has been addressed in our recent publications carried out in more general contexts allowing for the presence of an imposed flow [32,33] or for variable density and buoyancy effects [24].

3. The large activation energy asymptotic limit

3.1. A β -free reformulated problem

In this section, we derive a compact formulation valid in the distinguished limit $\beta \rightarrow \infty$ with $\epsilon_L = \mathcal{O}(1)$. The analysis is restricted to near-equidiffusion flames for which

$$Le_F \sim 1 + \frac{l_F}{\beta} \quad \text{and} \quad Le_O \sim 1 + \frac{l_O}{\beta},$$

where l_F and l_O are the reduced Lewis numbers of the fuel and oxidizer respectively. In this limit, the reaction zone is confined to an infinitely thin sheet that we shall call the flame surface, which is given by $F(t, x, y, z) = 0$, say. A reformulation of the problem free from the presence of β can then be derived. To this end, we expand the dependent variables in terms of β^{-1} in the form

$$\theta = \theta^0 + \frac{\theta^1}{\beta} + \dots, \quad y_F = y_F^0 + \frac{y_F^1}{\beta} + \dots, \quad y_O = y_O^0 + \frac{y_O^1}{\beta} + \dots$$

In the reaction zone and in the burnt gas, we assume that $\theta^0 = 1$ and $y_F^0 = y_O^0 = 0$, which leads to

$$\begin{aligned} \theta &= 1 + \frac{\theta^1}{\beta} + \dots, \quad y_F = \frac{y_F^1}{\beta} + \dots, \quad y_O \\ &= \frac{y_O^1}{\beta} + \dots, \quad \text{in the burnt gas.} \end{aligned} \quad (17)$$

The reaction term can be eliminated from Eqs. (8)–(10) by using the variables $Z_F \equiv \theta + y_F$ and $Z_O \equiv \theta + y_O$ which when substituted into (8)–(10) lead to

$$\frac{\partial Z_F}{\partial t} = \Delta Z_F - \frac{l_F}{\beta} \Delta y_F, \quad (18)$$

$$\frac{\partial Z_O}{\partial t} = \Delta Z_O - \frac{l_O}{\beta} \Delta y_O. \quad (19)$$

The variables Z_F and Z_O can be expanded as

$$Z_F = Z_F^0 + \frac{Z_F^1}{\beta} + \dots, \quad Z_O = Z_O^0 + \frac{Z_O^1}{\beta} + \dots, \quad (20)$$

but since $\theta^0 + y_F^0 = 1$ and $\theta^0 + y_O^0 = 1$ everywhere, one obtains

$$\begin{aligned} Z_F^0 &= \theta^0 + y_F^0 = 1, & Z_F^1 &= \theta^1 + y_F^1, \\ Z_O^0 &= \theta^0 + y_O^0 = 1, & Z_O^1 &= \theta^1 + y_O^1. \end{aligned}$$

Introducing the dependent variables $h \equiv \theta^1 + y_F^1$ and $k \equiv \theta^1 + y_O^1$, and substituting (17) and (20) into Eqs. (8), (18) and (19) yield the governing equations for θ^0, h and k in the form

$$\frac{\partial \theta^0}{\partial t} = \Delta \theta^0, \quad (21)$$

$$\frac{\partial h}{\partial t} = \Delta h + l_F \Delta \theta^0, \quad (22)$$

$$\frac{\partial k}{\partial t} = \Delta k + l_O \Delta \theta^0, \quad (23)$$

which are to be solved on both sides of the reaction sheet where $F \neq 0$, with the boundary conditions

$$\theta^0 = 0, \quad h = -\gamma_F z, \quad k = \gamma_O z \quad \text{as} \quad x^2 + y^2 + z^2 \rightarrow \infty. \quad (24)$$

The jump conditions at $F = 0$ are

$$[\theta^0] = [h] = [k] = 0, \quad (25a)$$

$$\left[\frac{\partial h}{\partial n} \right] = -l_F \left[\frac{\partial \theta^0}{\partial n} \right], \quad \left[\frac{\partial k}{\partial n} \right] = -l_O \left[\frac{\partial \theta^0}{\partial n} \right], \quad (25b)$$

$$\epsilon_L \left[\frac{\partial \theta^0}{\partial n} \right] = \sqrt{1 + \frac{|h-k|}{2}} \exp\left(\frac{h+k-|h-k|}{4}\right), \quad (25c)$$

where n is coordinate normal to the flame surface $F = 0$ pointing towards the unburnt gas. It can be noted that these jump conditions can be derived following the methodology described in [31]; see also [44, p. 39].

3.2. Stationary solutions

We first note that Eqs. (22) and (23) are in fact valid across the reaction sheet and that they are clearly satisfied, for time-independent problems, by h and k given by

$$h = -l_F \theta^0 - \gamma_F z \quad \text{and} \quad k = -l_O \theta^0 + \gamma_O z. \quad (26)$$

We also note that these expressions satisfy the far field boundary condition (24) and the jump conditions (25a) and (25b), and that they are in fact known in the burnt gas domain, Ω say, since $\theta^0 = 1$ there. Thus the problem is reduced to a single equation for θ^0 , to be solved outside Ω , with the solution required to vanish in the far field, and to satisfy two boundary conditions on the reaction sheet, i.e. on the unknown boundary $\partial\Omega$ of Ω . More precisely, introducing the notation $\psi \equiv \theta^0$ henceforth (while reserving for θ its usual meaning in the spherical coordinate system to be used shortly), we have to solve

$$\Delta \psi = 0 \quad \text{in} \quad R^3 \setminus \Omega \quad (27)$$

$$\psi = 0, \quad \text{as} \quad |r| \rightarrow \infty \quad (28)$$

$$\psi = 1, \quad \frac{\partial \psi}{\partial n} = -\bar{\mathcal{F}} \quad \text{on} \quad \partial\Omega. \quad (29)$$

Here $\bar{\mathcal{F}}$ is an explicit function of $(z; \epsilon_L, S, l_F, l_O)$ given by

$$\begin{aligned} \bar{\mathcal{F}} &= \epsilon_L^{-1} \left\{ 1 + \left| \frac{l_F - l_O}{2} + \frac{(S+1)^2}{4S} z \right| \right\}^{1/2} \\ &\quad \times \exp \left\{ \min \left(-\frac{l_F}{2} - \frac{S+1}{4} z, -\frac{l_O}{2} + \frac{S+1}{4S} z \right) \right\}, \end{aligned}$$

on using (25c) and (26). An important simplification occurs if we assume that $l_F = l_O$, equal to l , say, since these parameters can then be absorbed into ϵ_L . With this assumption¹ adopted henceforth we have

$$\bar{\mathcal{F}} = \epsilon^{-1} \mathcal{F}(z; S), \quad (30)$$

where²

$$\mathcal{F} = \left\{ 1 + \frac{(S+1)^2}{4S} |z| \right\}^{1/2} \exp \left\{ \min \left(-\frac{S+1}{4} z, \frac{S+1}{4S} z \right) \right\}, \quad (31)$$

¹ Note that the assumption $l_F = l_O$ also implies that the diffusion flame location coincides with that of the stoichiometric surface given by (6) in the Burke–Schumann limit $Da \rightarrow \infty$; see e.g. [26].

² The argument of the exponential can also be written as

$$\frac{(1-S^2)}{8S} z - \frac{(1+S)^2}{8S} |z|.$$

and

$$\epsilon \equiv \epsilon_L \exp \frac{l}{2}. \tag{32}$$

Parenthetically, we note that ϵ thus defined represents the non-dimensional radius of the classical Zeldovich flame ball at the stoichiometric conditions, whereas ϵ_L represents the non-dimensional planar flame thickness as defined in (11). This follows from the fact that the dimensional radius of the Zeldovich flame ball, δ_Z say, is given by

$$\delta_Z = \delta_L \exp \frac{l}{2}, \tag{33}$$

as can be confirmed from the results to be derived (Eq. (42) below) or shown independently.

4. Analytical Solution for $S = 1$

We shall seek an analytical solution of the problem (27)–(31) valid for small values of ϵ , i.e. for large values of the Damköhler number Da . It must be emphasised however that a non-trivial feature of the problem, as we shall confirm in the next section, is that the flame ball is centred at a location z_0 which needs to be determined as part of the solution. It is simpler therefore to begin with the stoichiometrically balanced case $S = 1$, for which we expect $z_0 = 0$ based on the symmetry of the temperature field with respect to the plane $z = 0$. We introduce a spherical coordinate system (r, θ, ϕ) centred at the origin and rescaled such that $(x, y, z) = \epsilon(r \sin \theta \cos \phi, r \sin \theta \sin \phi, r \cos \theta)$. The rescaling amounts to using as a new reference length the Zeldovich flame ball radius δ_Z given in (33) instead of $L_r \equiv L/\beta$. The rescaled problem is still given by (27)–(29) with $\bar{\mathcal{F}}$ modified to read

$$\bar{\mathcal{F}} = (1 + \epsilon r |\cos \theta|)^{1/2} \exp \left(-\frac{\epsilon r |\cos \theta|}{2} \right). \tag{34}$$

We look for axisymmetric solutions (independent of the azimuthal angle ϕ) and assume that the flame ball boundary $\partial\Omega$ is described by

$$r = R(\theta; \epsilon) \quad (\text{or more simply } r = R(\theta)).$$

In terms of the coordinates (r, θ) , the problem takes the form

$$\Delta\psi = 0 \quad \text{for } r > R(\theta), \tag{35}$$

$$\psi = 0 \quad \text{as } r \rightarrow \infty, \tag{36}$$

$$\psi = 1, \quad \frac{\partial\psi}{\partial r} = -\left(1 + \frac{R'^2}{R^2}\right)^{-1/2} \bar{\mathcal{F}} \quad \text{at } r = R(\theta), \tag{37}$$

where primes denote differentiation with respect to θ , $\bar{\mathcal{F}}$ is given by (34), and

$$\Delta\psi = \frac{1}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial\psi}{\partial r} \right) + \frac{1}{r^2 \sin \theta} \frac{\partial}{\partial \theta} \left(\sin \theta \frac{\partial\psi}{\partial \theta} \right).$$

We note that the last boundary condition is obtained by using the equality

$$\frac{\partial\psi}{\partial n} = \left(1 + \frac{R'^2}{R^2}\right)^{1/2} \frac{\partial\psi}{\partial r}, \tag{38}$$

valid at $r = R(\theta)$. This can be justified by noting that if $G \equiv r - R(\theta)$ then a unit outwardly-pointing vector normal to the flame surface $G = 0$ is given by

$$\mathbf{n} = \frac{\nabla G}{|\nabla G|} = \frac{\mathbf{e}_r - \frac{R'}{R} \mathbf{e}_\theta}{\left(1 + \frac{R'^2}{R^2}\right)^{1/2}}$$

so that

$$\begin{aligned} \frac{\partial\psi}{\partial n} &= \mathbf{n} \cdot \nabla\psi = \mathbf{n} \cdot \left\{ \mathbf{e}_r \frac{\partial\psi}{\partial r} + \frac{\mathbf{e}_\theta}{r} \frac{\partial\psi}{\partial \theta} \right\}_{r=R} \\ &= \left(1 + \frac{R'^2}{R^2}\right)^{-1/2} \left\{ \frac{\partial\psi}{\partial r} - \frac{R'}{R^2} \frac{\partial\psi}{\partial \theta} \right\}. \end{aligned} \tag{39}$$

Furthermore, since $\psi = 1$ on the surface $r = R(\theta)$, $d\psi = 0$ for a displacement on the surface. In other words, $\psi_r dr + \psi_\theta d\theta = 0$ for a displacement such that $dr = R'(\theta)d\theta$. Therefore $\psi_\theta = -R'(\theta)\psi_r$ on the surface showing that (38) follows from (39).

Returning to the problem (35)–(37), our aim is to find solutions $\psi(r, \theta)$ and $R(\theta)$. We shall determine two-term approximations of these for small values of ϵ . We first note that (34) implies that

$$\bar{\mathcal{F}} = 1 - b \epsilon^2 R^2 \cos^2 \theta + o(\epsilon^2) \quad \text{with } b = \frac{1}{4}. \tag{40}$$

We therefore write expansions in the form

$$R(\theta) = R_0 + \epsilon^2 R_1(\theta) + \dots, \quad \psi(r, \theta) = \psi_0(r) + \epsilon^2 \psi_1(r, \theta) + \dots$$

which we substitute into (35)–(37). The leading order problem is then

$$\begin{aligned} \frac{1}{r^2} \frac{d}{dr} \left(r^2 \frac{d\psi_0}{dr} \right) &= 0 \quad (r > R_0); \quad \psi_0 = 0 \text{ as } r \rightarrow \infty; \\ \psi_0 &= 1, \quad \frac{d\psi_0}{dr} = -1 \text{ at } r = R_0. \end{aligned} \tag{41}$$

Its solution is clearly given by

$$R_0 = 1, \quad \psi_0 = \frac{1}{r} \tag{42}$$

and corresponds to the classical Zeldovich flame ball in a uniform mixture.

To write the problem for ψ_1 , we first transfer the boundary conditions given at $r = R = 1 + \epsilon^2 R_1$ to the fixed location $r = 1$ by using Taylor expansions for small ϵ . From the condition $\psi = 1$ at $r = R$ we obtain $\psi_1(r = 1) + \psi_0'(1)R_1 = 0$, hence $\psi_1(r = 1) = R_1$. Similarly, using (37) and (40), we obtain the relation $\psi_{1r}(r = 1) + \psi_0''(1)R_1 = b \cos^2 \theta$; but since $\psi_0''(1) = 2$ and $R_1 = \psi_1(r = 1)$, this implies that $\psi_{1r} + 2\psi_1 = b \cos^2 \theta$ at $r = 1$. Finally the problem for ψ_1 is given by

$$\Delta\psi_1 = 0 \quad (r > 1) \tag{43}$$

$$\psi_1 = 0 \quad \text{as } r \rightarrow \infty \tag{44}$$

$$\frac{\partial\psi_1}{\partial r} + 2\psi_1 = b \cos^2 \theta \quad \text{at } r = 1 \tag{45}$$

$$R_1 = \psi_1(r = 1) \tag{46}$$

The Laplace differential equation and the first two boundary conditions are sufficient to determine ψ_1 ; the last condition allows then determination of R_1 . Indeed, the general solution of (43) satisfying (44) is given by

$$\psi_1 = \sum_{n=0}^{\infty} \frac{A_n P_n(\cos \theta)}{r^{n+1}}, \tag{47}$$

involving Legendre Polynomials P_n . Applying the boundary condition (45) implies that

$$\sum_{n=0}^{\infty} (1 - n) A_n P_n(\cos \theta) = b \cos^2 \theta \tag{48}$$

from which we find³

³ using $\cos^2 \theta = \frac{1}{3}(P_0(\cos \theta) + 2P_2(\cos \theta))$ since $P_0 = 1, P_1(x) = x$ and $P_2(x) = \frac{1}{2}(3x^2 - 1)$.

$$A_0 = \frac{b}{3}, \quad A_2 = -\frac{2b}{3}, \quad A_n = 0 (n > 2), \quad A_1 \text{ arbitrary.}$$

The arbitrariness of A_1 indicates that there are infinitely many solutions to the perturbation problem, obtained by arbitrary choosing A_1 . However, only $A_1 = 0$ insures the symmetry of the solution with respect to the plane $z = 0$ (or $\theta = \frac{\pi}{2}$) which we expect based on the symmetry of the original problem. Therefore we shall adopt this symmetry condition and set $A_1 = 0$. It is interesting to note that a similar non-uniqueness of the solution of the perturbation problem without additional condition being imposed has been noted in [10] in the context of flame ball drift. With the coefficients A_i being determined, we have on substituting into (47) and then using (46)

$$\psi_1 = \frac{b}{3} \left(\frac{1}{r} - \frac{2P_2(\cos \theta)}{r^3} \right) \quad \text{and} \quad R_1 = \frac{b}{3} (1 - 2P_2(\cos \theta)). \quad (49)$$

From (42) and (49) a two-term expansion is now available for ψ and R and is given by

$$\begin{aligned} \psi &= \frac{1}{r} + \frac{b\epsilon^2}{3} \left(\frac{1}{r} + \frac{1}{r^3} - \frac{3\cos^2 \theta}{r^3} \right) \quad \text{and} \\ R &= 1 + \frac{b\epsilon^2}{3} (2 - 3\cos^2 \theta) \end{aligned} \quad (50)$$

with $b = 1/4$ in this particular case $S = 1$.

5. Analytical solution in the general case

As mentioned in the previous section, a non-trivial feature of the problem is that the flame ball is centred at a location z_0 which needs to be determined as part of the solution. This location is determined by the function $\mathcal{F}(z)$ defined in (31), as we shall show.

First, expand $\mathcal{F}(z)$ in a Taylor series near z_0

$$\mathcal{F} = \mathcal{F}(z_0) + \mathcal{F}_z(z_0)(z - z_0) + \mathcal{F}_{zz}(z_0) \frac{(z - z_0)^2}{2} + \dots$$

and define

$$\epsilon \equiv \frac{\epsilon_L}{\mathcal{F}(z_0)} \exp \frac{l}{2}. \quad (51)$$

Next use spherical coordinates centred at $(x, y, z) = (0, 0, z_0)$ and rescaled such that $(x, y, z - z_0) = \epsilon(r \sin \theta \cos \phi, r \sin \theta \sin \phi, r \cos \theta)$. As can be checked, the rescaling amounts to using as a new reference length $\delta_z/\mathcal{F}(z_0)$, where δ_z is the Zeldovich flame ball radius corresponding to the stoichiometric conditions at $z = 0$ introduced in (33).

Finally, with $r = R(\theta)$ denoting the flame surface as before, the problem becomes

$$\Delta \psi = 0 \quad \text{for} \quad r > R \quad (52)$$

$$\psi = 0 \quad \text{as} \quad r \rightarrow \infty \quad (53)$$

$$\psi = 1,$$

$$\frac{\partial \psi}{\partial r} = - \left(1 + \frac{R^2}{r^2} \right)^{\frac{1}{2}} \left(1 + a\epsilon R \cos \theta - b\epsilon^2 R^2 \cos^2 \theta + o(\epsilon^2) \right) \text{ at } r = R, \quad (54)$$

where

$$a = \frac{\mathcal{F}_z(z_0)}{\mathcal{F}(z_0)} \quad \text{and} \quad b = -\frac{\mathcal{F}_{zz}(z_0)}{2\mathcal{F}(z_0)}. \quad (55)$$

We now write expansions in powers of ϵ in the form

$$R(\theta) = R_0 + \epsilon R_1(\theta) + \dots \quad \psi(r, \theta) = \psi_0(r) + \epsilon \psi_1(r, \theta) + \dots$$

which we introduce into (52)–(54).

The leading order problem is then found to be exactly as before, given by (41), with its solution (ψ_0, R_0) determined by (42).

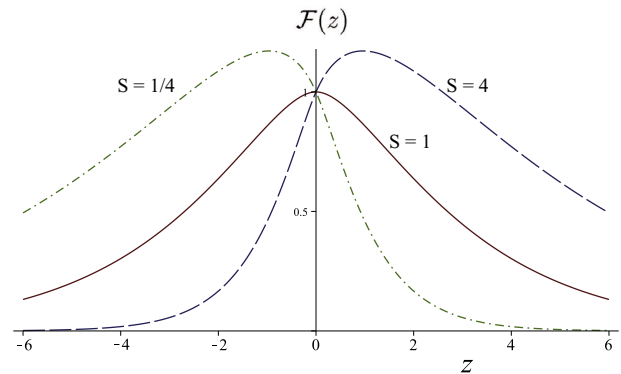


Fig. 2. $\mathcal{F}(z)$ versus z for $S = \frac{1}{4}, 1$, and 4.

The problem in the next approximation is found to be given by (43)–(46), except that (45) should now be modified to read

$$\frac{\partial \psi_1}{\partial r} + 2\psi_1 = -a \cos \theta \quad \text{at} \quad r = 1.$$

This modification leads to

$$\sum_{n=0}^{\infty} (1 - n) A_n P_n(\cos \theta) = -a \cos \theta, \quad (56)$$

instead of Eq. (48). So we must have

$$A_n = 0 \quad (n \neq 1) \quad \text{and} \quad 0 \times A_1 = -a.$$

Clearly the problem has no solution unless $a = 0$, that is $\mathcal{F}_z(z_0) = 0$, in view of (55). This condition appears as a solvability condition and means that a flame ball may only exist if it is centred on an extremum of the function $\mathcal{F}(z)$. Assuming that z_0 is such an extremum, the boundary condition (54) simplifies on setting $a = 0$, and we are back to the problem solved in the previous section whose solution to $\mathcal{O}(\epsilon^2)$ is given by (50), except that b must be determined now from (55).

All that remains to be done is to find z_0 as an extremum of $\mathcal{F}(z)$ and hence b . An elementary function study shows that $\mathcal{F}(z)$ possesses a unique extremum, as illustrated in Fig. 2 for selected values of S . More precisely, we find that

$$z_0 = \begin{cases} \frac{2(1-S)}{(1+S)^2} & \text{for } S \leq 1 \\ \frac{2S(1-S)}{(1+S)^2} & \text{for } S \geq 1 \end{cases}, \quad (57)$$

which determines the location at which the flame ball must be centred versus S . From the expression of z_0 it also follows that

$$\mathcal{F}(z_0) = \begin{cases} \sqrt{\frac{1+S}{2S}} \exp \left\{ \frac{S-1}{2(1+S)} \right\} & \text{and } b = \begin{cases} \frac{(1+S)^2}{16} & \text{for } S \leq 1 \\ \frac{(1+S)^2}{16S^2} & \text{for } S \geq 1 \end{cases} \end{cases}, \quad (58)$$

two quantities which completes the specification of ϵ in (51) and the two-term approximation (50) for $\psi(r, \theta)$ and $R(\theta)$.

The volume of the flame ball, V say, given by

$$V \equiv 2\pi \int_0^\pi \int_0^{R(\theta)} r^2 \sin \theta dr d\theta, \quad (59)$$

can then be evaluated using the expression for $R(\theta)$ in (50). The result is given below and is used in discussing minimum ignition energies.

6. Discussion and implication of the results

6.1. Summary of main results

The analysis above has shown that flame balls can exist only if centred at a single location z_0 which is determined by the stoichiometric coefficient S as given by (57). In fact, it is convenient to introduce the modified stoichiometric coefficient

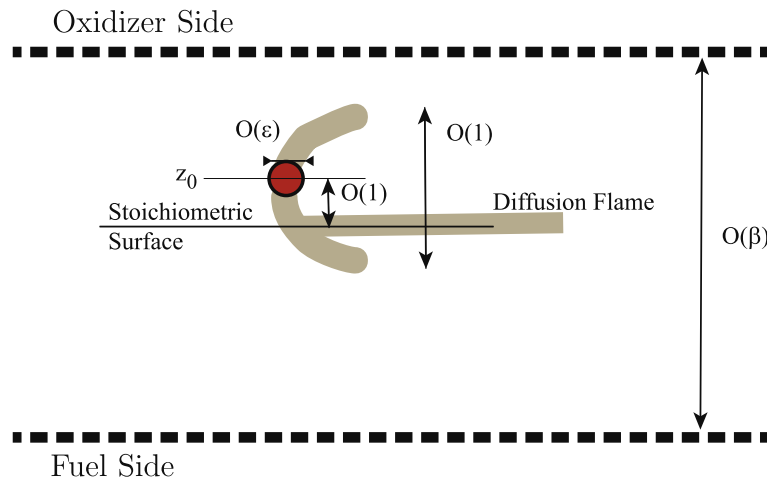


Fig. 3. Illustration of the scales involved in the asymptotic analysis.

$$\Delta \equiv \frac{S-1}{S+1}, \tag{60}$$

in terms of which the results can be expressed in a more compact form. This coefficient represents the non-dimensional stoichiometric location $\bar{z}_{st} \equiv Z_{st}/L$ on account of Eq. (6) which can be written as $\bar{z}_{st} = \Delta$.

In terms of Δ Eq. (57) can be expressed as

$$z_0 = -\Delta(1 + |\Delta|). \tag{62}$$

The analysis has also provided two-term approximations given by (50) for the temperature field $\psi(r, \theta)$ around the flame ball and for the ball radius $R(\theta)$. Using the expression for $R(\theta)$ in (59), a two-term approximation for the burnt gas volume V can be obtained. To sum up, it is found that

$$\psi = \frac{1}{r} + \frac{b\epsilon^2}{3} \left(\frac{1}{r} + \frac{1}{r^3} - \frac{3\cos^2\theta}{r^3} \right) \tag{63}$$

$$R = 1 + \frac{b\epsilon^2}{3} (2 - 3\cos^2\theta) \tag{64}$$

$$V = \frac{4\pi}{3} (1 + b\epsilon^2). \tag{65}$$

It should be noted that these formulas depend on z_0 , and hence on S , as a consequence of the dependence of ϵ and b on z_0 exhibited in (51), (55) and (58). This dependence can be conveniently expressed in terms of Δ by rewriting (51) and (58) in the form

$$\mathcal{F}_0 = \frac{\exp\left(-\frac{|\Delta|}{2}\right)}{(1-|\Delta|)^{\frac{1}{2}}}, \quad b = \frac{1}{4(1+|\Delta|)^2}, \quad \epsilon = \frac{\epsilon_L}{\mathcal{F}_0} \exp\frac{l}{2}, \tag{66}$$

where the shorthand notation $\mathcal{F}_0 \equiv \mathcal{F}(z_0)$ has been introduced.

6.2. Scales, notation, and relation to triple-flames

At this point, a remark is worth making in order to avoid confusion, namely that \bar{z}_{st} , z_0 and r in Eqs. (61)–(63) are coordinates non-dimensionalised using the scales L , L/β and δ_Z/\mathcal{F}_0 , respectively. These three scales are all needed to characterise the mixing length, the location of the flame ball centre, and the flame ball radius, respectively, as schematically depicted in Fig. 3 where L/β is taken as unit length.⁴ Note that we have also sketched in the figure the tri-

⁴ Of course, there is also a fourth scale pertaining to the reaction zone thickness of the flame ball, which is β times smaller than its radius. Although not depicted, this has been taken into account in the derivation of the jump conditions (25) valid in the limit $\beta \rightarrow \infty$.

ple-flame which can be encountered in this configuration and which has been investigated analytically and numerically in recent publications [32,33] in a more general context allowing for the presence of a parallel flow. As can be found in these publications when $\epsilon \ll 1$, the triple-flame has a thin premixed front with radius of curvature of the order of L/β , local burning speed $S_L\mathcal{F}(z)$, local thickness $\delta_L/\mathcal{F}(z)$ and a leading-edge located at z_0 given by (62). An important first consequence of the present analysis is therefore that flame balls may exist only if centred at the location of the leading-edge of the triple-flame which may exist in the non-uniform mixture considered. A second consequence is that $\delta_Z/\mathcal{F}(z)$ can be interpreted as a local Zeldovich flame ball radius, i.e. the radius of the spherical flame ball pertaining to a uniform mixture at the conditions prevailing at z . In particular, δ_Z/\mathcal{F}_0 is the radius of the Zeldovich flame ball at the conditions prevailing at z_0 ; this is consistent with Eq. (64) which implies that $R \rightarrow 1$ as $\epsilon \rightarrow 0$. Note that our notations also imply that $z = \beta(\bar{z} - \bar{z}_{st})$, with bars indicating that the coordinate is made non-dimensional using L as reference length, i.e. $\bar{z} \equiv Z/L$. Hence, the location of the flame ball centre measured with L is given by

$$\bar{z}_0 = \bar{z}_{st} + \frac{z_0}{\beta},$$

that is, using (61) and (62),

$$\bar{z}_0 = \Delta - \frac{\Delta(1+|\Delta|)}{\beta}. \tag{67}$$

We are now ready to discuss the main implications of the results, beginning with the stoichiometrically balanced case $S = 1$, then describing the effect of varying S , and finishing with conclusions related to the minimum energy required for successful ignition.

6.3. The stoichiometrically balanced case $S = 1$

Shown in Fig. 4 are temperature contours in the near field outside the flame ball (bounded by the solid thick inner line) in the x - z plane for $S = 1$ and $\epsilon = 0.9$. For comparison the spherical Zeldovich flame ball is also represented by a dashed circle. The figure illustrates the deviation from sphericity which is especially clear close to the flame and becomes weaker for $r \gg 1$. The flame ball deformation from a spherical shape increases with ϵ , as illustrated in Fig. 5. The figure shows how the non-uniformity in concentrations lead to an extension of the flame ball in the horizontal direction and a shrinkage it in the vertical direction. This deformation leads to a change in the volume of the burnt gas inside the ball, which in turn has implications concerning the minimum energy required for ignition as will be discussed below.

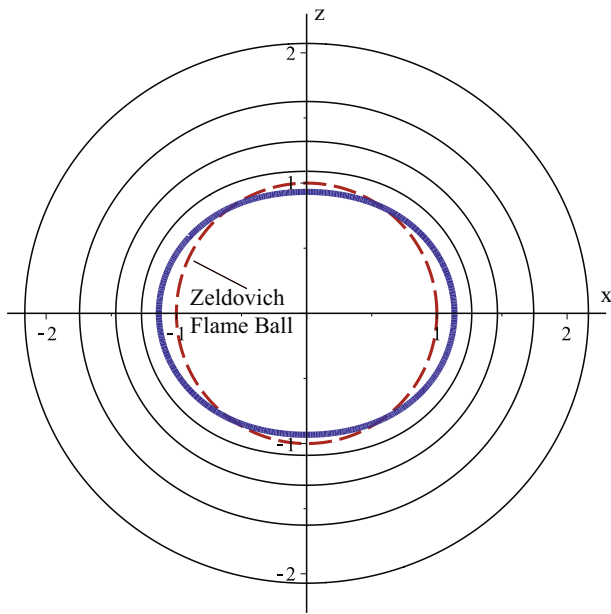


Fig. 4. Temperature contours in the near field outside the flame ball (bounded by the solid thick inner line) in the x - z plane for $S = 1$ and $\epsilon = 0.9$. The contours are represented by five solid lines corresponding to values of ψ uniformly equidistributed between 1 (the thick inner line) and 0.5 (the outer). For comparison the spherical Zeldovich flame ball is also represented by a dashed circle.

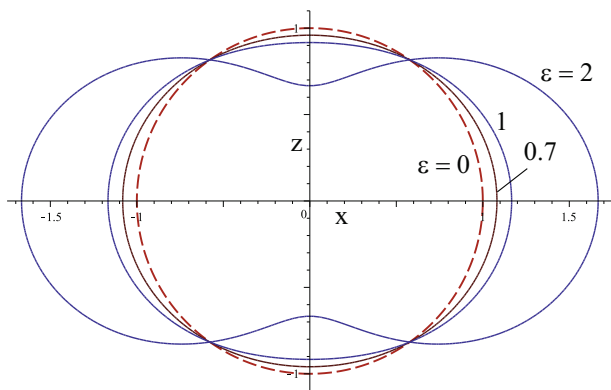


Fig. 5. Flame ball shape in the x - z plane for $S = 1$ and selected values of ϵ . The dashed circle pertaining to $\epsilon = 0$ represents the spherical Zeldovich flame ball.

6.4. Effect of the stoichiometry of the reaction

The location of the stoichiometric surface (or that of the diffusion flame), \bar{z}_{st} , and the location of the flame ball centre, \bar{z}_0 , vary with S according to (61) and (67) as shown in Fig. 6 where they are plotted versus $\Delta = (S - 1)/(S + 1)$. For $S = 1$ or $\Delta = 0$, both locations coincide midway between the fuel and oxidizer boundaries, $\bar{z}_0 = \bar{z}_{st} = 0$. For $S > 1$, the flame ball centre sits between the diffusion flame and the fuel (or lower) boundary, $-1 < \bar{z}_0 < \bar{z}_{st} < 1$. For $S < 1$, it sits between the diffusion flame and the oxidizer boundary, $-1 < \bar{z}_{st} < \bar{z}_0 < 1$. It can be noted that the distance between the flame ball centre and the diffusion flame is an increasing function of $|\Delta|$ of order β^{-1} . In particular, we have $\bar{z}_{st} \rightarrow -1$, $\bar{z}_0 \rightarrow -1 + 2\beta^{-1}$ as $\Delta \rightarrow -1$ and $\bar{z}_{st} \rightarrow 1$, $\bar{z}_0 \rightarrow 1 - 2\beta^{-1}$ as $\Delta \rightarrow 1$.

The change in the locations of \bar{z}_{st} and \bar{z}_0 just described is the dominant effect of varying S on the solution as long as ϵ remains sufficiently small. It should be noted, however, that the value of ϵ also varies with S ; these variations, can be accounted for, if needed, using the third equation in (66).

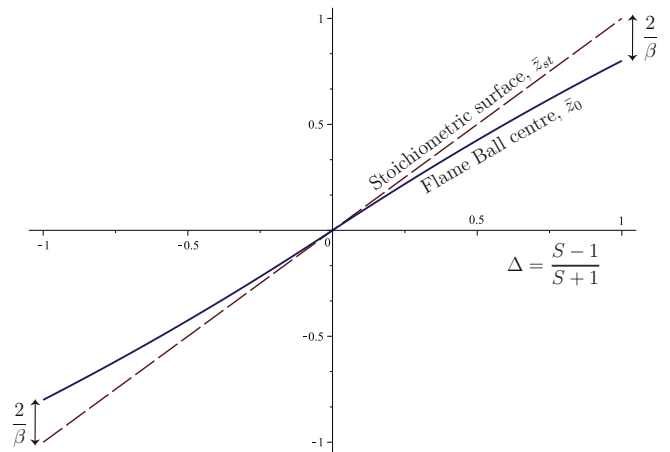


Fig. 6. Location of the Flame Ball centre, \bar{z}_0 , and location of the stoichiometric surface (and the diffusion flame), \bar{z}_{st} , versus the stoichiometric parameter $\Delta \equiv \frac{S-1}{S+1}$.

6.5. Minimum ignition energy

In studies on ignition in uniform premixed reactive mixtures, the thermal energy contained in a Zeldovich flame ball has been used to estimate the energy (of a spark say) required for successful ignition, see e.g. [1,2,13–15]. If V_Z is the dimensional volume of the Zeldovich flame ball in the mixture and T_{ad} is the temperature of the burnt gas enclosed, then

$$E_Z = \rho c_p (T_{ad} - T_u) V_Z. \quad (68)$$

In particular, if the uniform mixture is such that its reactants have the same concentrations as those prevailing at the stoichiometric surface in our problem, then $V_Z = (4\pi/3)\delta_z^3$ where δ_z is the radius of the Zeldovich flame ball at the stoichiometric conditions given in (33). Henceforth E_Z will be taken to correspond to these conditions and will be referred to as the *minimum ignition energy of the stoichiometric uniform mixture*.

Let us by analogy use the energy enclosed in our flame balls as an estimate for the minimum ignition energy required in our non-uniform mixture. To this end, we note that the dimensional volume of our flame ball corresponding to the non-dimensional volume V given in (65) is $V_B = VV_Z/\mathcal{F}_0^3$; this is simply because the length scale used to obtain (65) is δ_z/\mathcal{F}_0 as mentioned in Section 5. Corresponding to this volume is a dimensional energy

$$E_B = \rho c_p (T_{ad} - T_u) V_B, \quad (69)$$

given that the temperature inside the flame ball is equal to T_{ad} , neglecting non-dimensional temperature variations of order β^{-1} .

From (65), (68) and (69), it follows that

$$\frac{E_B}{E_Z} = \frac{V_B}{V_Z} = \frac{1}{\mathcal{F}_0^3} (1 + b\epsilon^2), \quad (70)$$

where the coefficients \mathcal{F}_0 , b and ϵ depend on the stoichiometric coefficient Δ as given by (66). More explicitly, we obtain

$$\frac{E_B}{E_Z} = e^{\frac{3|\Delta|}{2}} (1 - |\Delta|)^{\frac{3}{2}} \left\{ 1 + \frac{e^{|\Delta|+1} (1 - |\Delta|) \beta^2}{4(1 + |\Delta|)^2 \text{Da}} \right\}, \quad (71)$$

where Eq. (12), namely $\epsilon_L^2 = \beta^2/\text{Da}$, has been used to introduce the Damköhler number Da . For illustration, plots of E_B/E_Z versus $|\Delta|$ based on (71) are shown in Fig. 7 for $l = 0$ and three selected values of ϵ_L .

Formula (71) is in fact an important result since it may serve as a possible criterion for successful ignition in our non-uniform reactive mixture. We shall simply highlight in what follows a few

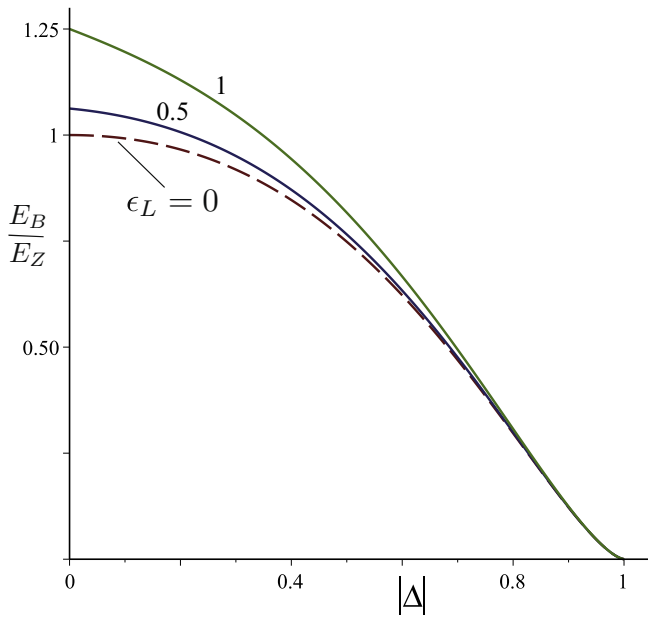


Fig. 7. $\frac{E_B}{E_Z}$ versus $|\Delta|$ for three selected values of ϵ_L (and $l = 0$).

implications of this formula. In doing so, it is convenient to refer to E_B as the *minimum ignition energy of the non-uniform mixture*, denoted by E_{min} in the abstract and conclusion sections, by analogy to our reference above to E_Z as being *the minimum ignition energy of the stoichiometric uniform mixture*.

We begin with the stoichiometrically balanced case $S = 1$ or $\Delta = 0$, for which (71) simplifies to

$$\frac{E_B}{E_Z} = 1 + \frac{e^l \beta^2}{4 \text{Da}}, \quad (72)$$

or, equivalently,

$$\frac{E_B}{E_Z} = 1 + \frac{e^l \epsilon_L^2}{4} = 1 + \frac{\epsilon^2}{4}.$$

This shows that the minimum ignition energy of the non-uniform mixture E_B is higher than that of the uniform mixture E_Z by a relative amount which is proportional to the square of the Zeldovich number and inversely proportional to the Damköhler number. It can be noted that E_B approaches E_Z in the limit of infinitely large Da , or more precisely of vanishingly small values of the non-uniformity parameter ϵ_L (or ϵ). This makes sense from the physical point of view since in this case, $S = 1$, E_B is the thermal energy inside a non-spherical flame ball centred at the stoichiometric surface, while E_Z is that inside of a (spherical) Zeldovich flame ball in a homogeneous mixture corresponding to the reactants concentration at the same stoichiometric surface. It is natural therefore that the non-spherical flame ball solution tends to the Zeldovich solution as the non-uniformity measured by ϵ_L becomes weaker. This is not true when $S \neq 1$ since then $\bar{z}_0 \neq \bar{z}_{st}$. In the general case, we may write

$$\frac{E_B}{e^{\frac{3|\Delta|}{2}(1-|\Delta|)^{\frac{3}{2}} E_Z}} = 1 + \frac{e^{|\Delta|+l}(1-|\Delta|) \beta^2}{4(1+|\Delta|)^2 \text{Da}} \quad (73)$$

showing that $E_B \rightarrow E_Z^0$ in the limit $\epsilon_L \rightarrow 0$ where

$$E_Z^0 \equiv e^{\frac{3|\Delta|}{2}(1-|\Delta|)^{\frac{3}{2}}} E_Z.$$

In fact, E_Z^0 , which can also be written as $E_Z/\mathcal{F}^3(z_0)$ on using the first expression in (66), can be interpreted as the energy inside a Zeldo-

vich flame ball corresponding to a uniform mixture at the condition prevailing at z_0 . To see this, let us define $\tilde{E}(z)$ as being the thermal energy inside a Zeldovich flame ball in a uniform mixture at the local conditions prevailing at z and let us refer to such a ball by the shorthand notation $\text{ZFB}(z)$. The radius of $\text{ZFB}(z)$ is $\delta_Z/\mathcal{F}(z)$ as argued in Section 6.2 and the temperature of the burnt gas inside it is T_{ad} , neglecting non-dimensional temperature variations of order β^{-1} for $z \sim 1$. Therefore $\tilde{E}(z) = (4\pi/3)\rho c_p(T_{ad} - T_u)\delta_Z^3/\mathcal{F}^3(z)$, that is

$$\tilde{E}(z) = E_Z/\mathcal{F}^3(z), \quad (74)$$

confirming our interpretation for $E_Z^0 = \tilde{E}(z_0)$. In fact, E_Z^0 is clearly the minimum of $\tilde{E}(z)$, given that z_0 is the location of the maximum of $\mathcal{F}(z)$. In other words, z_0 is the location of the Zeldovich flame ball $\text{ZFB}(z)$ of minimum energy (and radius) in the mixing layer. Returning to (73), it is seen that the energy of the flame ball in the non-uniform mixture E_B is equal the energy of the Zeldovich ball at the location of the leading edge of a triple-flame to leading order plus a positive correction depending on the stoichiometric coefficient and the Lewis number, which is proportional to the square of the Zeldovich number and inversely proportional to the Damköhler number. The findings suggest that a necessary condition for successful ignition is that the energy deposited exceeds E_B and that the most favourable location for energy deposit is z_0 , given that $\tilde{E}(z) \geq \tilde{E}(z_0)$.

7. Concluding remarks

A thermo-diffusive model for flame balls in a mixing layer has been analysed theoretically in the limit of large Zeldovich number β . The analysis has led to a free boundary problem outside a (burnt-gas) domain whose unknown boundary represents an infinitely-thin reaction zone. The free boundary problem has been solved analytically using perturbation methods in the asymptotic limit of large Damköhler number. The solutions have provided explicit formulas determining the free boundary and the temperature field outside it. These solutions, which represent non-spherical flame balls generalising the classical Zeldovich flame balls to non-uniform mixtures, are shown to exist only if the flame ball is centred at a single location z_0 . This location differs from the location of the stoichiometric surface \bar{z}_s by an amount of order β^{-1} , and both locations depend simply on a normalised stoichiometric coefficient Δ , see Eqs. (60), (61) and (67). In fact, it is found that z_0 is precisely the position of the leading-edge of a triple-flame in the mixing layer.

In analogy with the homogeneous case, the thermal energy of the burnt gas inside the flame ball has been used to derive an expression for the minimum energy E_{min} (of an external spark say) required for successful ignition. In particular, it is found that the presence of the inhomogeneity increases the minimum ignition energy required compared to the homogeneous case. For a stoichiometrically balanced mixture, corresponding to $\Delta = 0$, the relative increase in the ignition energy is found to be proportional to the square of the Zeldovich number and to the reciprocal of the Damköhler number, see formula (72) with $E_{min} \equiv E_B$. More generally, for arbitrary value of Δ , the minimum ignition energy is found to correspond to that of the Zeldovich flame ball in a uniform mixture at the local conditions prevailing at z_0 , i.e. at the location of the leading edge of the triple flame, plus a positive amount depending on Δ which is again proportional to the square of the Zeldovich number and to the reciprocal of the Damköhler number, see formula (73). In summary, the analysis provides a possible criterion for successful ignition in a non-homogeneous mixtures by determining the minimum energy required and the location z_0 , argued to be the most favourable for ignition, where it should be deposited.

We close the paper by noting that the study is a first theoretical step towards a better understanding of ignition in non-homogeneous mixtures. The focus of the paper was on deriving a free-boundary problem and providing an analytical description of its stationary solutions where feasible. These solutions are expected to be unstable, as observed in related numerical studies [41,42], but this aspect, as well as the temporal evolution into propagating triple-flames, is not considered here. The stability aspect will be however the subject of further studies which will allow the inclusion of stabilising effects such as volumetric heat-losses (which are known to have a significant influence on flame balls [3,4] as well as on triple-flames [27–29]), or a flow-field (as in related studies such as [6,45] or [46]). Finally, three interesting open questions for further investigation raised by one of the reviewer of the paper, to whom we are grateful, are the following. (1) How would a convective field superimposed onto the non-homogeneity of the mixture (like in previous studies [32,33]) affect the existence/stability of the flame balls? (2) How may the uniqueness of the solution, enforced by the symmetry condition mentioned at the end of Section 4, be influenced by considering other configurations like e.g. in the presence of a convective, laminar or even lightly turbulent, field? (3) Can the dynamic ignition process, or the transit of the (typically unstable) flame balls into propagating triple-flames, be described by time-dependent evolution equations characterising the flame shape, similar to the non-linear integro-differential equation for the flame ball radius derived by G. Joulin in the homogeneous case [13]?

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