The effect of Lewis number variation on combustion waves in a model with chain-branching reaction

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Abstract

In this paper we investigate the linear stability and properties of the travelling premixed combustion waves in a model with two-step chain-branching reaction mechanism in the adiabatic limit in one spatial dimension. It is shown that the Lewis number for fuel has a significant effect on the properties and stability of premixed flames, whereas the Lewis number for the radicals has only quantitive (but not qualitative) effect on the combustion waves. We demonstrate that when the Lewis number for fuel is less than unity the flame speed is unique and is a monotonically decreasing function of the dimensionless activation energy. The combustion wave is stable and exhibits extinction for finite values of activation energy as the flame speed decreases to zero. For fuel Lewis number greater than unity the flame speed is a double-valued function. The slow solution branch is shown to be unstable whereas the fast solution branch is either stable or exhibits the onset of pulsating instabilities via the Hopf bifurcation. The evolution of these instabilities leads to flame extinction.

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1 Introduction

Premixed combustion waves in models with two-step kinetic reaction schemes have been studied for some time. They provide an alternative to both the classical one-step reaction mechanism, which has been a cornerstone of combustion theory, and models with detailed kinetic mechanisms of the reaction. Two-step models can, on one hand, describe the complexity of real multi-step combustion reactions more adequately than one-step models, and on the other hand, still be relatively simple for comprehensive general analysis.

One of the most interesting types of two-step models are the models with chain-branching reaction. Hydrocarbon flames normally produce a pool of radicals through the branching reaction steps. These radicals later recombine to produce heat and products. The chain-branching reaction mechanism cannot be adequately described by an overall reaction. Therefore the two-step chain-branching reaction models are more popular than other two-step kinetic mechanisms.

The first two-step chain branching reaction model was introduced by Zeldovich in 1948 [1] and was later analyzed by Liñán in 1971 [2] using the activation energy asymptotic (AEA). Therefore this model is usually referred to as the Zeldovich-Liñán model. The model comprises a chain branching reaction $A + B \rightarrow 2B$, and chain-breaking (or recombination) reaction $B + B + M \rightarrow 2P + M$, where A is the fuel, B is the radical, P is the product, and M is a third body. It is assumed that the first reaction has a large activation energy and is isothermic whereas, the recombination reaction has zero activation energy and is exothermic. The condition of an isothermic first reaction was subsequently relaxed in [3].

In [2] it was shown that there are three flame regimes in the Zeldovich-Liñán model: fast, slow and intermediate recombination regimes. In the fast recombination regime the production of radicals by the branching step is much slower than the consumption of radicals through the recombination step, which has the following consequences: the chain-branching and chain-recombination take place in the same thin reaction zone, the concentration of radicals is asymptotically small, and therefore the steady state approximation can be applied to it. In the slow recombination regime, the concentration of radicals is $O(1)$ and all radicals are produced in a thin reaction zone. However the consumption of radicals proceeds in a long scale region greater or comparable to the convection-diffusion region. In the intermediate recombination regime the rates of the branching and termination reactions are comparable. The concentration of radicals is neither asymptotically small nor it is $O(1)$ and the recombination reaction takes place in a region, which is much thicker than the branching reaction zone, but much thinner than the convection-diffusion region.

Using the above arguments various asymptotic expansions have been introduced in different flame zones. The resulting asymptotic differential equations are then solved either analytically or numerically depending on the complexity of the system of equations appearing as a results of asymptotic analysis. The model considered in [2, 3] does not include heat loss and the response curves obtained in these papers are single valued functions. In [4] the Zeldovich-Liñán model with heat loss to the surroundings was considered by using the AEA. It was demonstrated that the flame speed as a function of other parameters of the problem is a C-shaped function which exhibits turning point type extinction condition similar to that predicted by the one-step nonadiabatic model [5].

In a number of investigations [6, 7, 8, 9], the influence of stretch on premixed flame for the Zeldovich-Liñán model was studied. The authors considered several distinguished limits in order to examine the problem in terms of AEA either analytically or semi-analytically. As a result it was found that the flame response to stretching depended upon the particular flame regime i.e. slow, fast or intermediate recombination.

The stability of flames in the Zeldovich-Liñán model has not been investigated. It appears that the problem of stability of the combustion waves cannot be treated effectively by using the AEA for this model. The complexity of the stability analysis increases significantly as the num-

ber of reactions involved in the kinetic scheme increases in comparison to models with one-step kinetic mechanism. The other important property of the Zeldovich-Liñán model is the nonlinear dependence of the reaction terms on the concentration of reactants, which is usually avoided in the one-step formulation. In order to investigate the stability of the flames with chain-branching reaction mechanism, a simplified version of the Zeldovich-Liñán model was introduced recently [10]. In this model the order of the recombination reaction was reduced by one so that the resulting kinetic scheme is written as $A + B \rightarrow 2B$, $B + M \rightarrow P + M$. The reduction of the order of the chain-breaking reaction makes the dependence of the rection terms on the concentration of radicals in the governing equations linear, which allows the problem to be treated by using the AEA. The speed of the combustion wave was determined as a function of the parameters of the problem and was shown to be C-shaped in the nonadiabatic case. For the adiabatic case the expression derived in [10] suggests a unique flame speed. For the case of the reactant Lewis number less than one the analysis in [10] predicted that the wave can lose stability due to the emergence of cellular instabilities.

In our earlier paper [11] we have investigated the properties of the model introduced in [10] in the adiabatic case and in the limit of equal diffusivity of the reactant, the radical and heat. In contrast to [10] the activation energy in [11] is taken to be $O(1)$ (not an infinite number). As noted in [7] this is a reasonable assumption for real flames like the hydrogen oxidation flame. We also used a different nondimensionalization, which enables us to make more convenient comparisons between one- and two-step models. In [11] the properties of the traveling wave solutions are investigated in detail by means of numerical simulation. It was demonstrated that the speed of the combustion wave as a function of parameters is single valued. We have found that for finite activation energy there is a residual amount of reactant left behind the traveling combustion wave which is not used in the reaction [12]. This makes the problem similar to the nonadiabatic one-step premixed flames. The other characteristic of the model considered in [11] which makes the similarity between the adiabatic two-step reaction and the nonadiabatic one-step system even stronger is that, at certain parameter values the combustion wave exhibits extinction. However, this extinction only occurs at zero flame speed. This mainly distinguishes the one- and two-step adiabatic models, since in the former case the wave can propagate for any parameter values and even in the nonadiabatic case the speed of combustion wave does not vanish for finite parameter values, whereas this is not the case for the two-step models. The route to extinction in this model was investigated in detail in [13]. It was shown that the flame speed as a function of activation energy approaches zero in a linear fashion. The stability of the traveling combustion waves was also investigated. For equidiffusional case the flame is stable over an extensive parameter range which correlates with the results in [10].

The equidiffusional approximation used in [11, 13] makes the analysis of the problem easier. However this choice of parameters reduces the applicability of the results to real flames with chain-branching reaction mechanism, which can be characterized by various values of Lewis numbers for both radicals, L_B, and fuel, L_A. This is especially true for the stability analysis, since the flame stability is expected to depend substantially on these parameters $L_{A,B}$ [10]. The aim here is to investigate the effect of varying the Lewis number for both the fuel and radicals on the properties of the premixed combustion waves and their stability. We are predominantly interested in the qualitative changes of the flame speed and stability character. Our investigation focuses on several values of Lewis numbers taken in such a way so as to cover all possible qualitative relations between the values of L_A and L_B. More detailed studies on this topic is a subject of our future work, which we discuss briefly in the conclusion.

2 Model

We consider an adiabatic model of premixed flame propagating in one spatial dimension that includes the following two steps: autocatalytic chain branching $A + B \rightarrow 2B$ and recombination

$B + M \to C + M$. Here A is the fuel, B is the radical, C is the product, and M a third body. It is assumed that all the heat of the reaction is released during the recombination stage and the chain branching stage does not produce or consume any heat. According to [11], the governing equations for the non-dimensional temperature, u, concentration of fuel, v, and radicals, w, can be written in the nondimensional form as

$$\begin{aligned} u_t &= u_{xx} + rw, \\ v_t &= L_A^{-1} v_{xx} - \beta v w e^{-1/u}, \\ w_t &= L_B^{-1} w_{xx} + \beta v w e^{-1/u} - r\beta w, \end{aligned} \tag{1}$$

where x and t are the dimensionless spatial coordinate and time respectively, L_A and L_B are the Lewis numbers for fuel and radicals respectively, β is the dimensionless activation energy of the chain-branching step (which corresponds to the definition for the one step model [12]), r is the ratio of the characteristic time of the recombination and branching steps (which cannot be reproduced in one-step formulation of the flame kinetics).

Equations (1) are considered subject to the boundary conditions

$$\begin{aligned} &u = 0, \quad v = 1, \quad w = 0 \quad \text{for} \quad x \to \infty, \\ &u_x = 0, \quad v_x = 0, \quad w = 0 \quad \text{for} \quad x \to -\infty. \end{aligned} \tag{2}$$

On the right boundary we have cold ($u = 0$) and unburned state ($v = 1$), where the fuel has not been consumed yet and no radicals have been produced ($w = 0$). The ambient temperature is taken to be equal to zero. On the left boundary ($x \to -\infty$) neither the temperature of the mixture nor the concentration of fuel can be specified. We only require that there is no reaction occurring so the solution reaches a steady state of (1). Therefore the derivatives of u, v are set to zero and $w = 0$ for $x \to -\infty$.

The solution to the problem (1), (2) is sought in the form of a traveling wave $u(x,t) = u(\xi)$, $v(x,t) = v(\xi)$, and $w(x,t) = w(\xi)$, where a coordinate in the moving frame, $\xi = x - ct$, is introduced and c is the speed of the traveling wave. Substituting the solution of this form into the governing equations we obtain

$$\begin{aligned} &u_{\xi\xi} + cu_\xi + rw = 0, \\ &L_A^{-1} v_{\xi\xi} + cv_\xi - \beta v w e^{-1/u} = 0, \\ &L_B^{-1} w_{\xi\xi} + cw_\xi + \beta v w e^{-1/u} - r\beta w = 0. \end{aligned} \tag{3}$$

The boundary conditions (2) can be modified if we multiply the first equation in (3) by β, add it to the second and third equations in (3) and integrate it once with respect to ξ over $(-\infty, +\infty)$. This yields a condition: $\lim_{\xi\to-\infty} S = \lim_{\xi\to+\infty} S$, where $S = \beta u + v + w$. Combining this condition with (2) results in

$$\begin{aligned} &u = 0, \quad v = 1, \quad w = 0 \quad \text{for} \quad \xi \to \infty, \\ &u = \beta^{-1}(1-\sigma), \quad v = \sigma, \quad w = 0 \quad \text{for} \quad \xi \to -\infty, \end{aligned} \tag{4}$$

where σ denotes the residual amount of fuel left behind the wave and is unknown until a solution is obtained.

In order for the traveling combustion wave solution to (1) to exist several conditions have to be satisfied. The reactions in front of the wave, in the preheat region, and behind the wave, in the product region, should be in chemical equilibrium. The latter restriction implies that the

radicals cannot be produced in these regions effectively. In other words the production of radicals is overwhelmed by the consumption of the radicals, which are governed by the second and the third terms in the right hand side of the last equation in (1). In front of the flame, the temperature is low and the branching reaction term, which has the Arrhenius form for the temperature dependence, vanishes. Therefore any small concentration of radicals would exhibit a fast depletion due to the recombination and diffusion. In the product region, the reaction must be completed therefore the branching term is less than the recombination term in the third equation of (1), that is

$$r > \sigma \exp\left(\frac{-\beta}{1-\sigma}\right). \tag{5}$$

Condition (5) was first derived in [13] and it defines the region in the parameter space where the travelling combustion waves exist. Once this condition is violated the combustion waves exhibit extinction. In the reaction zone the production of radicals due to branching reaction is dominating the depletion due to the recombination therefore $v\exp(-1/u)$ is greater than r. In other words, it is the reaction region where all radicals are produced and the concentration of the radicals vanishes in front and behind of this region. It has to be noted here that in the reaction zone the diffusion of radicals becomes important therefore the production term has to balance both the recombination and diffusion of radicals, which strengthens the requirement $v\exp(-1/u) > r$. The maximum value for $v\exp(-1/u)$ can be estimated as $\exp(-\beta/(1-\sigma))$. Therefore we have to require $r < \exp(-\beta/(1-\sigma))$ in order for the travelling wave solution to exist. The increase of the activation energy results in the decrease of $\exp(-\beta/(1-\sigma))$. Therefore if r is fixed for certain value of β, the latter condition is violated and the travelling solution exhibits extinction. The extinction phenomenon for the travelling combustion wave can also be understood from the following consideration. Once the activation energy of the branching reaction is increased, the burning temperature is decreasing. This results in the decrease of the radicals production due to the Arrhenius type of the branching reaction rate dependence. Since less radicals are produced, w decreases and as a result less heat is released during the recombination step, which again is related to the decrease of the burning temperature and the flame speed. In contrast to the Zeldovich-Liñán model, the decrease of the radicals concentration does not change the balance between the rates of branching and recombination reactions since both are proportional to w and at certain values of β the production of radicals becomes completely dominated by the consumption of radicals in the reaction zone due to both recombination and diffusion resulting in the extinction of the travelling combustion wave. Therefore we conclude that extinction of the combustion wave is inevitable in the model considered in this paper even in the adiabatic case when the activation energy becomes sufficiently large.

3 Results

The properties of the traveling combustion waves are investigated numerically by solving the equations (3) subject to boundary conditions (4). We used a standard shooting algorithm with a fourth order Runge-Kutta integration scheme first and then the results were corrected by employing the relaxation algorithm. The detailed description of these methods can be found in [11, 12]. The control parameters used in our investigation are the Lewis numbers for the fuel and radicals. We take $L_{A,B}$ one order of magnitude smaller than, greater than, or equal to unity i.e. either $L_{A,B} = 0.1$, 1.0 or 10. This allows us to cover all possible combinations for the diffusivities of heat and reactants in order to understand the extent of the impact of these parameters on both the properties and stability of the combustion wave solutions to the model.

The flame stability has been investigated by using the Evans function method [14] implemented with the use of compound matrix approach [12]. However in order to carry out such an investigation the methods outlined in [12] have to be generalized for a model with two step reaction mechanism.

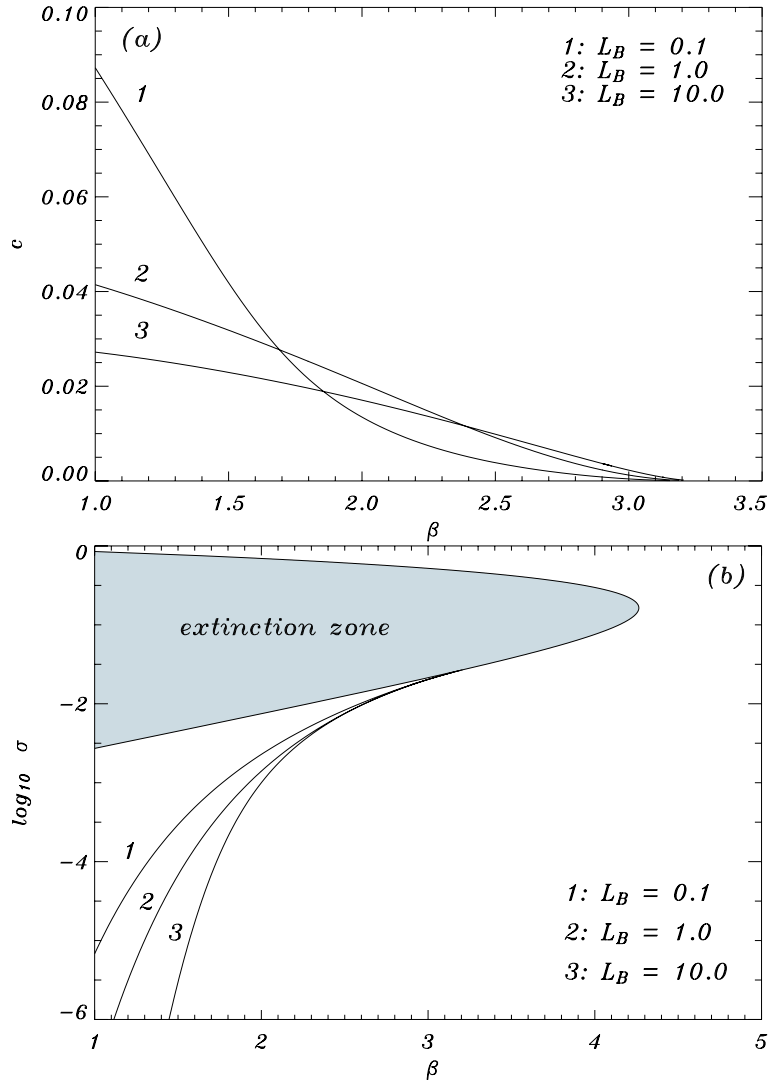

Figure 1: The dependence of (a) the flame speed c and (b) the residual amount of fuel σ on the dimensionless activation energy β for $L_A = 0.1$ and various values of the Lewis number for radicals L_B as shown in figure.

From the point of view of the compound matrix method the difference between the one-step model considered in [12] and the current model with two-step reaction mechanism is in the increased number of governing equations. This substantially changes the topology of the linear stability problem in the compound matrix method formulation for the case of the two-step model and makes the methods described in [12] not applicable to the case considered here. In order to overcome this difficulty we used an exterior algebra formulation of the Evans function method and generalized our approach to make it applicable to models with arbitrary number of the governing equations as described in [15] .

In figure 1 the properties of the traveling solution to (3) for small Lewis number for fuel ($L_A = 0.1$) are summarized. Figure 1(a) shows the flame speed as a function of the dimensionless activation energy. For parameter L_B varying from 0.1 to 10 the wave speed $c(\beta)$ is a monotonically decaying function exhibiting extinction for β around 3.2. The flame speed decreases to zero according to the quadratic law i.e. $c \sim (\beta - \beta_e)^2$ as the dimensionless activation energy approaches the extinction value β_e. The individual curves plotted for $L_B = 0.1$, 1.0 and 10 possess the same quantitive behaviour although the values of the flame speed change with varying L_B. It is interesting to note

that the location of the point of extinction in the parameter space does not depend on the Lewis number for radicals. However this issue has to be investigated separately. In figure 1(b) the residual amount of fuel σ (in logarithmic scale) is plotted as a function of β. The dependence of σ on β is a single-valued function. The shaded region shows the parameter values, where condition (5) is violated and the travelling wave solutions cease to exist. For parameter values sufficiently away from the extinction condition the residual amount of fuel is negligible. Almost all the fuel is converted to radicals with no fuel leakage observed. As we increase β and approach the extinction point, σ becomes significant. At the extinction condition the residual amount of fuel reaches its maximum value corresponding to the boundary of the shaded region i.e. the extinction condition defined by (5). Regardless of L_B the dependences $c(\beta)$ and $\sigma(\beta)$ show qualitatively similar behaviour for $L_A < 1$. Moreover the $\sigma(\beta)$ curves always lie below the lower boundary of the shaded region. The extinction value for the dimensionless activation energy is less than the maximum value of β_m which corresponds to the boundary of the shaded region or the extinction curve i.e. a set of β and σ for which the condition $r = \sigma \exp[-\beta/(1-\sigma)]$ is satisfied. However as L_A is increased the extinction value of the activation energy, β_e, moves closer to the maximum value β_m and coalesces with it when $L_A = 1$. All solution branches were found to be stable.

For the case $L_A = 1$, the structure of the travelling solution branch in the parameter space changes. In figure 2(a) the dependence of the flame speed, c, on β is plotted for $L_B = 0.1$, 1.0, and 10. Although $c(\beta)$ is still a monotonic function approaching zero as β reaches some critical value $\beta_e = 4.2...$ corresponding to extinction, the behaviour of the wave speed is different compared to $L_A < 1$ near the point of extinction. Namely, the flame speed decreases to zero according to the linear law: c is proportional to $(\beta - \beta_e)$ for β close to β_e in contrast to the quadratic dependence for $L_A < 1$. Similar results were obtained analytically in [13]. Qualitatively the dependence of $c(\beta)$ is the same for the Lewis number for radicals varying in two orders of magnitude from 0.1 to 10. The extinction occurs when the activation energy approaches the value β_m corresponding to the turning point of the extinction curve. This extinction condition does not depend on the variation of L_B. The dependence of σ on β is plotted in the figure 2(b) for $L_B = 0.1$, 1.0, and 10. The residual amount of fuel as a function of β is monotonic. The curve $\sigma(\beta)$ lies below the shaded region for all values of $\beta < \beta_m$. For activation energies sufficiently smaller than β_m the value of σ is negligible and there is no fuel leakage and all the fuel is converted into radicals in the course of the reaction. Closer to the extinction point, when $\beta \approx \beta_m$, the residual amount of fuel becomes considerable and fuel leakage occurs. At the point of extinction, when $\beta = \beta_m$, the curve $\sigma(\beta)$ becomes tangent to the extinction curve in preparation to a more drastic change of the solution branch behaviour for $L_A > 1$. The stability of the travelling wave solutions was investigated by using the Evans function method. All solution branches were found to be stable for $L_A = 1.0$.

The dependence of the flame speed, c, on β for $L_A = 10$ and various values of L_B is shown in the figure 3 (a). In figure 3(b) the residual amount of fuel as function of β is presented for the same parameter values. In both figures the stable solution branches are plotted with the solid line and the dotted line represents the unstable branches. As seen from the figures both $c(\beta)$ and $\log_{10}\sigma(\beta)$ become C-shaped for $L_A > 1$. The flame speed and the residual amount of fuel as functions of the dimensionless activation energy are double valued. There are either two solutions travelling with different flame speed or no solution exists. For small values of β the fast solution branch is stable and is characterized by a negligibly small residual amount of fuel. As the activation energy is increased, the residual amount of fuel grows and it becomes significant as the turning point of $c(\beta)$ and $\sigma(\beta)$ is approached (for both curves the turning point is reached simultaneously). It is important to note that in the case of $L_A > 1$, the extinction condition is reached for values of β larger than β_m – the maximal activation energy for the shaded region. The slow solution branch (lower branch) is always unstable. It is characterized by a considerable fuel leakage so that the curve $\sigma(\beta)$ is lying above the shaded region for all allowable values of β. As we move along the slow solution branch by decreasing β from the turning point value for $c(\beta)$, the flame speed decreases

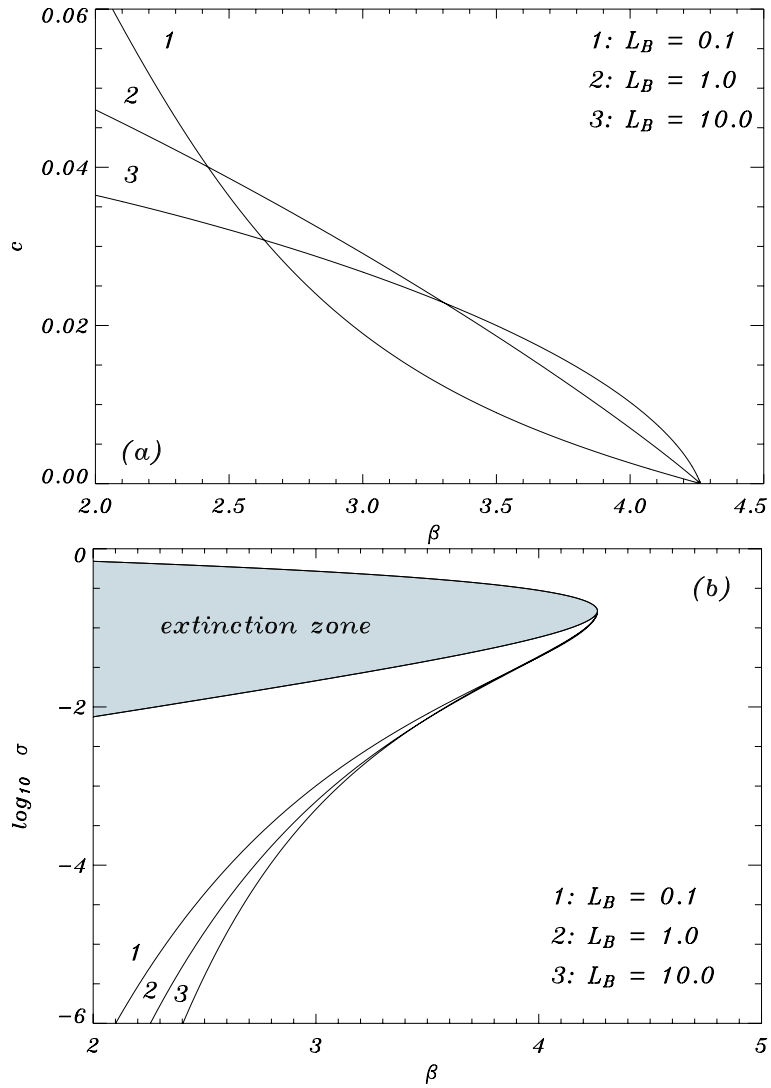

Figure 2: The dependence of (a) the flame speed c and (b) the residual amount of fuel σ on the dimensionless activation energy β for $L_A = 1.0$ and various values of the Lewis number for radicals L_B as shown in figure.

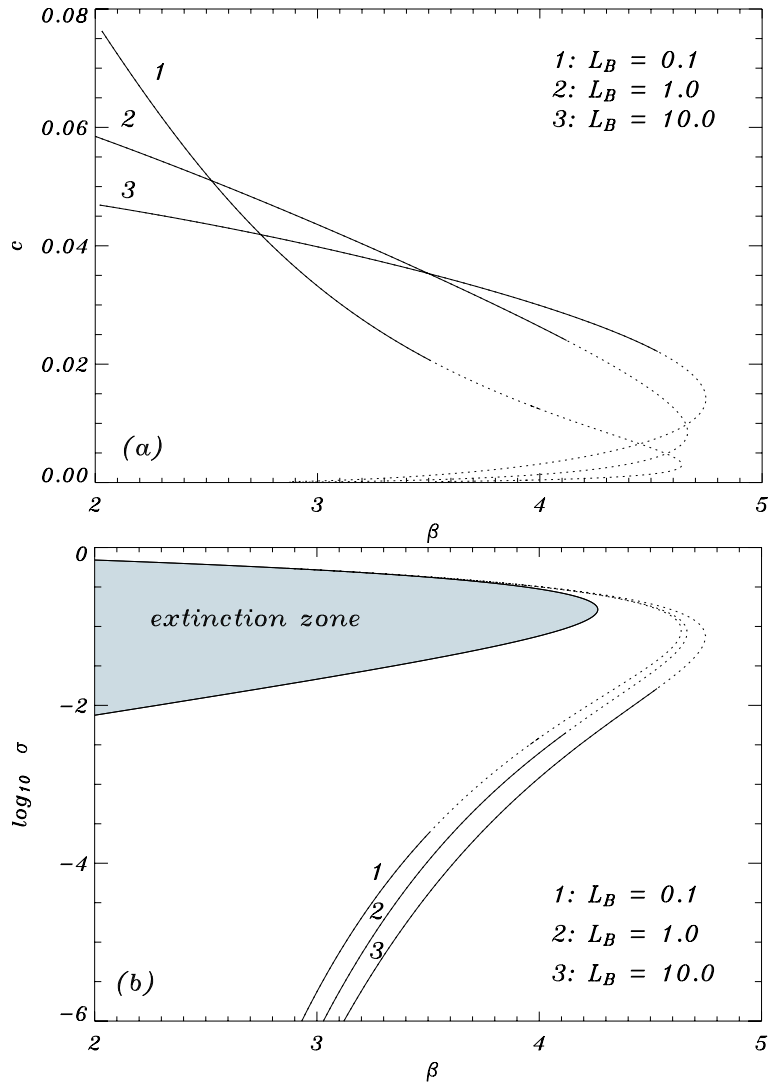

Figure 3: The dependence of (a) the flame speed c and (b) the residual amount of fuel σ on the dimensionless activation energy, β, for $L_A = 10$ and various values of the Lewis number for radicals L_B as shown in figure. Solid lines denote stable solutions, whereas dotted lines represent unstable solutions.

and at a certain value β_e the speed becomes zero. The behaviour of $c(\beta)$ follows the quadratic law i.e. $c \sim (\beta - \beta_e)^2$ as the dimensionless activation energy approaches the extinction value β_e. The variation of L_B does not affect either the qualitative behavior of the solution branches in the parameter space or the flame stability.

The stability of the travelling wave solutions for $L_A = 10$ have been investigated using the Evans function method. The fast solution branch is found to be stable for small values of the activation energy. As β is increased the fast solutions lose stability before reaching the turning point due to the emergence of the pulsating instabilities via a Hopf bifurcation. This scenario of the loss of stability has been observed for L_B ranging from 0.1 to 10. The locations of the Hopf bifurcation move closer to the turning point as the value of L_B increases.

We also investigate the stability of the fast solution branch by direct integration of the governing equations (1) which are solved on a sufficiently large coordinate domain with the boundary conditions (2) imposed at the edge points of the spatial grid. For our numerical algorithm we use the method of splitting with respect to physical processes. That is, initially we solve the set of ordinary differential equations which describes the temperature and the species concentration variations due to the branching and recombination reactions by using the fourth order Runge-Kutta algorithm. Next, equations of mass transfer for fuel and radicals are solved with the Crank-Nicholson method with the second order approximation in space and time. The initial conditions for the numerical scheme are taken in the form of a travelling wave solution of (3). In order to investigate the stability of the solutions the initial profiles are perturbed. The perturbation is added by multiplying the initial radical concentration profiles by a factor which is taken to be close to unity. If the solution is stable the perturbation disappears and the initial profile converges to the autowave solution travelling with the speed c obtained from numerical integration of (3). If the travelling wave solution is unstable then the small perturbation of the initial profiles contain portions of the eigenmodes giving rise to instability which grow exponentially with time. As a result either a new solution emerges, such as pulsating waves, or the flame extinguishes.

According to the linear stability analysis the rate of convergence/divergence of the solution of (1) to an autowave is given as $e^{-\lambda t}$ where λ is a spectral parameter which can be found by the Evans function method (see [12] for more details on the linear stability analysis). The real part of λ describes how fast the perturbations grow ($Re(\lambda) > 0$) or decay ($Re(\lambda) < 0$) with time and the imaginary part of the spectral parameter defines the frequency with which the perturbations oscillate in time.

The results of direct integration of (1) are presented in figure 4 for $L_A = 10$, $L_B = 1.0$ and two values of the dimensionless activation energy (a) $\beta = 4.0$ and (b) $\beta = 4.22$. The stability analysis of the travelling wave solutions presented in figure 3(a, b) (curves 2) for the given parameter values suggests that the solution is stable for $\beta < 4.1225...$ and it loses stability for larger values of β due to the occurrence of Hopf bifurcation. For the case $\beta = 4.0$, figure 4(a), we integrate the governing equations (1) with the perturbed initial conditions and we found that the solution profiles converge quickly to a travelling wave solution. The speed of the wave was found to be equal to the flame velocity obtained from the integration of (3) up to the fourth significant digit. In order to illustrate the convergence of the solution of (1) to a travelling wave, the dependence of the maximum value of radical concentration on time is plotted in figure 4(a) with the solid line. The squares represent the prediction obtained from the Evans function method which gives $Re(\lambda) = -1.3502... \times 10^{-4}$ and $Im(\lambda) = 6.5786... \times 10^{-4}$, which results in $w_{max}(t) - w^{\infty}_{max} \sim \sin(\Im\lambda t + \varphi)\exp(\Re\lambda t)$, where $w^{\infty}_{max} = \lim_{t\to\infty} w_{max}(t)$ and φ is a phase parameter which can determined from the linear analysis. However here φ is taken to be a free parameter which is chosen to best fit the numerical data obtained from the integration of (1). We have also plotted the envelope of $w_{max}(t)$ with the dotted lines in the same figure.

The initial perturbation of the solution profile is sufficiently large in the case $\beta = 4.0$. As a result the maximum value of the concentration of radicals begins to oscillate with large amplitude.

However, as shown in figure 4(a), these oscillations converge exponentially fast to a constant value corresponding to the $\max[w(\xi)] = 0.1271...$ for the autowave solution of (3). The behaviour of the function $w_{max}(t)$ is similar to the prediction of the linear analysis. The difference can be observed only at the initial stages of the flame propagation when the perturbation is large and the nonlinearity in the system plays a considerable role. After the first period of oscillations the amplitude decreases by almost twice the amount compared to the initial period of oscillations and $w_{max}(t)$ obtained from the integration of (1) becomes identical to the prediction of the linear analysis. The frequency of oscillations and the rate of the perturbation decay found with the Evans function method agree with the results of the numerical analysis of (1).

For $\beta = 4.22$ the travelling wave solution is predicted to become unstable (see figure 3 curve 2). The rate of the exponential growth of the linear perturbation is predicted by the Evans function method as $Re(\lambda) = 1.0490... \times 10^{-4}$ and the frequency of oscillations as $Im(\lambda) = 4.6530... \times 10^{-4}$. The initial solution profile is taken in the form of the autowave and the perturbations are imposed by the truncation errors of the numerical scheme. The solid line represents the results from the numerical integration of (1), the prediction of the linear analysis is shown with the squares connected with the dashed lines, and the dotted lines mark the envelope of the linear perturbation growth. The maximum value of the radical concentration grows exponentially in time in an oscillatory manner. After several periods of oscillations the amplitude of pulsations becomes large. At $t = 4 \times 10^4$ the discrepancy between the results obtained with the linear analysis and direct numerical integration of the governing partial differential equations becomes visible indicating that the nonlinearity is beginning to affect the evolution of perturbations. At $t \approx 5 \times 10^4$ the amplitude of oscillations reaches a critical value whereby the distortion of the initial autowave profile of temperature and concentration of species with growing perturbations becomes too large, and the travelling wave cannot propagate further and the flame extinguishes. The agreement between the prediction of the linear perturbation analysis and the results of the numerical integration of (1) is good in estimating both the rate of exponential growth and the frequency of oscillations. It should be noted that we have not observed the formation of steady propagating pulsating waves as was for the case of the one-step model [16]. Same qualitative results were obtained for other values of $\beta > 4.1225$.

4 Conclusions

In this paper we investigated the properties and the linear stability of the travelling premixed combustion waves in a model with two-step chain-branching reaction mechanism in the adiabatic limit. The model was introduced by J. Dold and co-authors in [10]. The current paper naturally extends our previous analysis of the model with equidiffusional approximation [11, 13] by relaxing the constrains on the Lewis numbers for fuel and radicals. The Lewis numbers in this study are chosen in such a way as to cover all possible cases i.e. $L_{A,B} \ll 1$, $L_{A,B} \gg 1$, and $L_{A,B} \sim 1$. The aim of such a selection of parameters is to understand how they affect the properties and the stability of the combustion waves in the two-step model studied here.

The investigation of the traveling combustion waves revealed that the Lewis number for fuel L_A has a substantial affect on the properties and the stability of the premixed flames in our model, whereas L_B only has quantitative effects on the behavior of combustion waves. It was demonstrated that depending on the Lewis number for fuel, the flame speed has either subcritical or supercritical behaviour as function of the dimensionless activation energy. The transition from sub- to supercritical type of dependence occurs when $L_A = 1$.

When the Lewis number for fuel is less than unity, the flame speed is a single-valued, monotonically decreasing function of β. The flame extinction occurs as the speed of combustion wave decays to zero quadratically as a function of β when the critical values of the activation energy is approached i.e. $c \sim (\beta - \beta_e)^2$. The travelling combustion wave was found to be stable for all parameter values in this case.

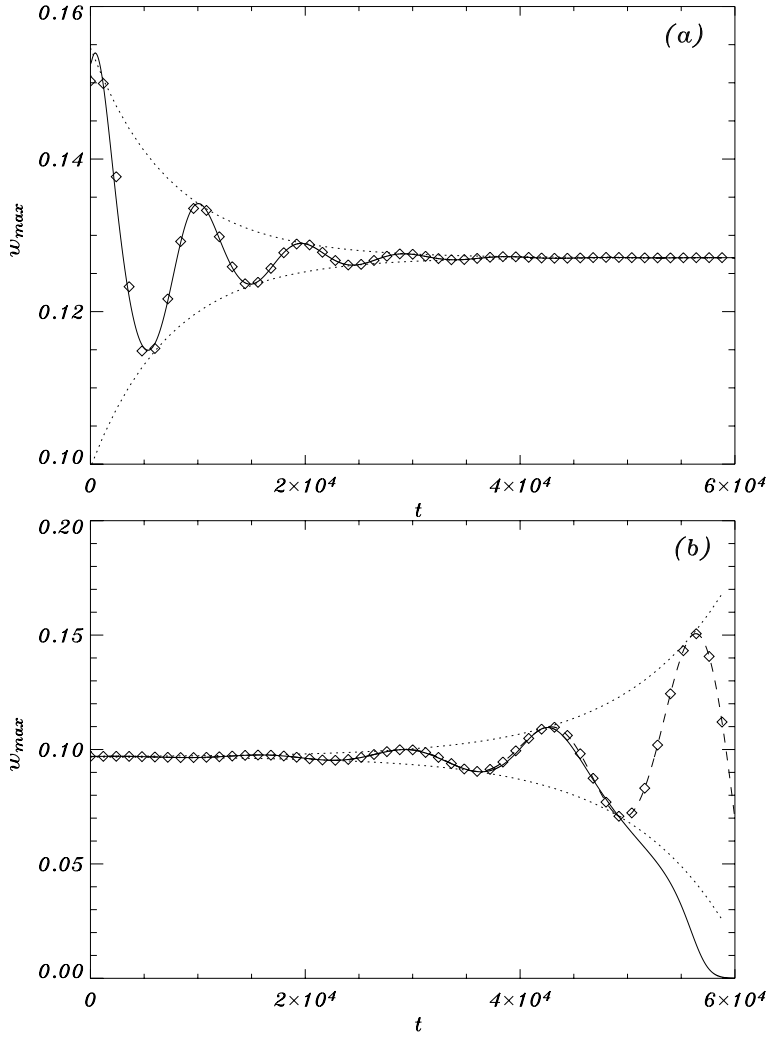

Figure 4: The dependence of the maximum value of the concentration of radicals on time for $L_A = 10$, $L_B = 1.0$, (a) $\beta = 4.0$ and (b) $\beta = 4.22$. The solid line shows the results from the numerical integration of the governing equations (1), the squares represent the prediction of the linear stability analysis (in (b) squares are connected with a dashed line). The dotted lines represent the envelope of oscillations of the linear perturbations.

For Lewis number for fuel greater than unity, the dependence of flame speed on β is double valued and C-shaped. The travelling wave solutions either do not exist or there are two solutions travelling with different speed corresponding to fast and slow solution branches in the parameter space. Flame extinction occurs when the slow and fast solution branches merge as a result of the turning point bifurcation. The slow solution branch is a monotonically increasing function of β. For larger values of β, it ceases to exist at the turning point and for smaller β, the flame speed decrease to zero as $c \sim (\beta - \beta_e)^2$ as the critical parameter value of the activation energy, β_e, is reached. For the fast solution branch, $c(\beta)$ is a monotonically decreasing function. The travelling combustion wave is stable for small values of the dimensionless activation energy and it losses stability via the Hopf bifurcation as β is increased towards the turning point.

The transition from single- to double-valued nature of the dependence of the flame speed occurs for the critical value of the Lewis number for fuel, $L_A = 1$. This case have been investigated earlier in [11, 13] for $L_B = 1$ both numerically and analytically. It was shown that the speed of combustion wave is a unique, monotonically decreasing function of β. The flame speed vanishes at the extinction point for a finite value of activation energy β_e approaching zero according to linear law i.e. $c \sim (\beta - \beta_e)$. Our current study reveals that the same type of behaviour is alos true for L_B ranging between 0.1 and 10. The Evans function stability analysis shows that the travelling combustion wave is stable for $L_A = 1$. This study clearly shows that the case $L_A = 1$ is a distinguished limit, where the transition from one pattern in the flame dynamics to the other occurs.

Pulsating instabilities emerge due to the occurrence of the Hopf bifurcation on the solution branch. The rate of exponential growth of the unstable modes and the frequency of oscillations were estimated by using the Evans function method. It is known from the analysis of the one-step models that the evolution of pulsating instabilities can lead to the formation of the pulsating waves travelling with speed oscillating in time and characterized by periodic dependence of the solution profiles on time. We have studied the behaviour of the fast travelling wave solutions by direct integration of the governing partial differential equations for $L_A = 10$ and $L_B = 1$. The results of this numerical analysis are in excellent agreement with the investigation of the travelling wave solutions obtained from the integration of the ordinary differential equations and the stability analysis carried out with the use of the Evans function. The behaviour of the travelling wave solutions, once steady propagating wave solutions lose stability, was investigated. It was shown that the pulsating instabilities grow exponentially in time exactly as predicted by the Evans function analysis. However as the amplitude of oscillations becomes large the travelling wave solution exhibits extinction rather than pulsating behavior. Consequently, the formation of stable pulsating solutions have not been observed in our investigation.

It has to be noted that the process of the emergence of pulsating combustion waves from steady propagating flames has not been fully understood even for the one-step models. Therefore this phenomenon has to be further investigated for both types of models. The other important issue that needs clarification is the exact scenario for the onset of instabilities in the current model. In [12] it was demonstrated that the onset of pulsating instability for the travelling combustion waves in one-step models is due to the occurrence of Hopf bifurcation which originates in the parameter space from the codimesion 2 point known as the Bogdanov-Takens bifurcation. This bifurcation is shown to be responsible for the switching nature of instability from pulsating to the uniform type. It is important to understand whether a similar scenario exists for the two-step model considered here. Our current analysis leads us to belive this is likely, however this point needs to be thoroughly examined and is a subject of our current investigation.

References

[1] Zeldovich Y B 1948 Teorii rasprostranenia plameni. *Zh. Phys. Khim.* **22**, 27-49, English translation in NACA TM 1282 (1951)

[2] Liñán, A 1971 A theoretical analysis of premixed flame propagation with an isothermal chain-branching reaction. Insituto Nacional de Technica Aerospacial "Esteban Terradas" (Madrid), USAFOSR Contract No. E00AR68-0031, Technical Report No. 1.

[3] Joulin, G, Liñán, A., Ludford, G S S, Peters, N and Schmidt-Lainé, C 1985 Flames with chain-branching/chain-breaking kinetics, *SIAM J. Appl. Math.* **45**, 420-434.

[4] Chao, B H and Law, C K 1994 Laminar flame propagation with volumetric heat loss and chain branching-termination reactions, *Int. J. Heat Mass Transfer* **37**, 673-680.

[5] Joulin, G and Clavin, P. 1979 Linear stability analysis of nonadiabatic flames: a thermal-diffusional model, *Combust. Flame* **1979**, 139-153.

[6] Seshardi, K and Peters, N 1983 The influence of stretch on a premixed flame with two-step kinetics, *Combust. Sci. and Tech.* **33**, 35-63.

[7] Mikolaitis, D W 1986 Adiabatic flame Speeds and the Zeldovich-Liñán model, *Combust. Sci. and Tech.* **49**, 277-288.

[8] Tam, R Y 1988 Stretch response and large heat release in the Zeldovich-Liñán model, *Combust. Sci. and Tech.* **60**, 125-142.

[9] Tam, R Y 1988 Damköler-number ratio asymptotics of the Zeldovich-Liñán model, *Combust. Sci. and Tech.* **62**, 297-309.

[10] Dold, J. W., 2007, Premixed flames modelled with thermally sensitive intermediate branching kinetics. *Combust. Theory Mod.*, published online.

[11] Gubernov, V V, Sidhu, H S and Mercer, G N 2006 Combustion waves in a model with chain branching reaction. *J. Math. Chem.*, **39**, 1–14.

[12] Gubernov V V, Mercer G N, Sidhu H S and Weber R O 2004 Evans function stability of nonadiabatic combustion waves, *Proc. R. Soc. Lond. A*, **460**, 1259-1275.

[13] Gubernov, V V, Sidhu, H S and Mercer, G N 2007 Combustion waves in a model with chain branching reaction and their stability, *submitted to Combust. Theory Mod. 2007*

[14] Evans, J W, 1972 Nerve axon equations: III Stability of the nerve impulses, *Indiana Univ. Math. J.* **22**, 577-593.

[15] Gubernov V V, Mercer G N, Sidhu H S 2006 Generalized compound matrix method, *Appl. Math. Lett.* **19**, 458-463.

[16] Weber, R O, Mercer, G N, Sidhu, H S, and Gray, B F, 1997 Combustion waves for gases (Le = 1) and solids (Le $\to \infty$), *Proc. R. Soc. Lond. A*, **453**, 1105-1118.