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Analysing combustion waves in a model with chain branching

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Abstract

We analyse the travelling wave solutions in an adiabatic model with two-step chain branching reaction mechanism. We show that the behaviour of the combustion waves are similar to the case of the corresponding nonadiabatic one-step reaction, namely there is residual amount of fuel left behind the travelling waves and the solutions can exhibit extinction. We also analyse how the speed of the travelling wave solutions and the residual amount of fuel left behind the fuel change, as control parameters are varied.

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1 Introduction

Any scientist who has gazed into a campfire will appreciate the complexity of combustion and the difficulty in constructing a theoretical model of the process. To date only the simplest models have been comprehensively analysed. Combustion waves have been studied for some time and are the topic of a relatively recent review by Merzhanov and Rumanov [9]. Combustion waves have been observed in numerous experimental situations [9], and play important roles in industrial processes, such as the production of exotic materials using Self propagating, High temperature Synthesis (SHS) [8] .

One-step irreversible reaction models have contributed greatly to our understanding of combustion phenomena. In these models it is assumed that the reaction is well modeled by a single step of fuel (F) and oxidant (O_2) combining to produce products (P) and heat. The generic kinetic schemes of models with one-step chemistry are

$$F \xrightarrow{k(T)} P + \text{heat} \quad \text{or} \quad F + O_2 \xrightarrow{k(T)} 2P + \text{heat},$$

where the temperature dependent rate constant $k(T)$ is given by Arrhenius kinetics $k(T) = e^{-T_a/T}$, T_a is the activation temperature, and T is the temperature of the reaction. These models have proven their usefulness since they are relatively simple and allow analytical investigation using asymptotic methods in the limit of infinitely large activation temperature [4, 18].

The most important feature of one-step models is that they have led to many useful and qualitatively correct predictions for phenomena such as: ignition, extinction and stability of diffusion flames; propagation and stability of premixed flames; flame balls and their stability; structure and propagation of flame edges etcetera.

However, in the overwhelming majority of cases, the chemical reactions in flames proceed according to a complex mechanism, that involves a variety of different steps [18]. Moreover, for many reactions, models with simple one-step kinetics may lead to erroneous conclusions as noted by Westbrook and Dryer [17]. In other words, if we want to obtain a realistic description of the flame kinetics, several different reaction steps, each with its own intermediate chemical species, have to be taken into account. Recent advances in computational power make it possible to study the flame behaviour using full numerical solutions of the equations of energy and mass transfer for all of the species involved with detailed chemistry. Several numerical codes have been developed in order to carry out these calculations, such as Sandia's CHEMKIN code [15] or the Flame Master code [10]. These numerical algorithms have been successfully applied to analyze the properties of both diffusion [11] and premixed flames [12]. Although such investigations are useful in providing some quantitative results for observed phenomena, there is still a great deal of uncertainty about the reliability of these complex models when applied to the prediction of generic behaviour of flames such as stable combustion regimes, limits of the flame extinction and particularly the onset of pulsating and cellular combustion regimes, when the dynamics of reactions begin to change rapidly in space and/or time.

A logical compromise between the models with singe step and detailed multi-step kinetics has been found recently by using reduced kinetic mechanisms. In a many articles [1, 11, 12, 13, e.g.] the detailed schemes of the hydrogen and methane oxidation, which include dozens of intermediate reactions, were successfully reduced to several steps. The remarkable feature of these type of models is that they allow on one hand, analytical investigation [1, 13] to be successfully undertaken, while on the other hand they are able to produce excellent quantitative results [11, 12] to predict flame characteristics accurately for some specific reactions such as flame propagation velocities, flame structures including the profiles of temperature and reactants etc. These studies are very important for applied problems, in which the properties of flames for specific reaction and flame configuration are studied.

In this article we focus on the investigation of premixed combustion waves in a model with non-competing two-step chain branching reaction. As noted by Dold et al. [2] the chain branching reaction model is more realistic in describing real flames such as hydrocarbon flames in comparison to other two-step reaction models. This model was initially considered by Liñán [7] and Zeldovich et al. [18] and then generalized by Joulin et al. [6]. It is usually referred to as Zeldovich–Liñán model. We believe that this preliminary investigation is a constructive step towards an understanding of the travelling wave solution in chain branching reactions.

2 Mathematical formulation

We consider an adiabatic model in one spatial dimension that includes two steps:

$$\begin{aligned}\text{Autocatalytic chain branching} \quad & A + B \to 2B\,; \\ \text{Recombination} \quad & B + M \to C + M\,. \end{aligned} \tag{1}$$

Here A is the fuel, B is the radical, C is the product, and M represents any molecule that is required to start the reaction but remains unchanged by the reaction. Following Dold et al. [2, 3] and Zeldovich et al. [18] we assume that all heat of the reaction is released during the recombination stage and the chain branching stage does not produce or consume any heat. As noted by Zeldovich et al. [18], in this scheme the recombination stage serves both as an inhibitor which terminates the chain branching and an accelerant which produces heat. According to Dold and Weber [3], equations governing this process can be written as

$$\begin{aligned}
\rho c_p \frac{\partial T}{\partial t} &= k\frac{\partial^2 T}{\partial x^2} + \rho Q A_r Y_B \,,\\
\frac{\partial Y_A}{\partial t} &= D_A \frac{\partial^2 Y_A}{\partial x^2} - A_B Y_A Y_B e^{-E/RT} \,,\\
\frac{\partial Y_B}{\partial t} &= D_B \frac{\partial^2 Y_B}{\partial x^2} + A_B Y_A Y_B e^{-E/RT} - A_r Y_B \rho / M_w \,,
\end{aligned} \tag{2}$$

where T is the temperature, Y_A and Y_B represent the concentrations of fuel and radicals respectively, ρ is the density, c_p is the specific heat, k is the thermal conductivity, M_w is the mean molecular weight, D_A and D_B represent the diffusivities of fuel and radicals respectively, A_r and A_B are constants of recombination and chain branching reactions respectively, Q is the heat of the recombination reaction, E is the activation energy for chain branching reaction, and R is the universal gas constant.

We introduce the nondimensional variables

$$\begin{aligned}
t' &= \frac{\rho Q A_B R}{c_p M_w E} t \,, \quad x' = \sqrt{\frac{\rho^2 Q A_B R}{k M_w E}} x \,,\\
u &= \frac{RT}{E} \,, \quad v = \frac{Y_A M_w}{\rho} \,, \quad w = \frac{Y_B M_w}{\rho} \,,
\end{aligned} \tag{3}$$

and rewrite (2) as

$$\begin{aligned}
u_t &= u_{xx} + rw \,,\\
v_t &= \tau_A v_{xx} - \beta v w e^{-1/u} \,,\\
w_t &= \tau_B w_{xx} + \beta v w e^{-1/u} - r\beta w \,,
\end{aligned} \tag{4}$$

where we drop the primes and the following nondimensional parameters have been introduced: $\beta = c_p E/QR$, $r = A_r/A_B$, $\tau_{A,B} = \rho c_p D_{A,B}/k$. Here parameter β is the dimensionless activation energy of the chain branching step which coincides with the corresponding definition for the one step model given by Gubernov et al. [4]. Parameter r is the ratio of the characteristic time of the recombination and branching steps and cannot be reproduced in one step approximation of the flame kinetics. The parameters $\tau_{A,B}$ represent the inverse Lewis numbers for the fuel and radicals respectively.

Equations (4) are considered subject to the boundary conditions

$$\begin{aligned} &u = 0\,, \quad v = 1\,, \quad w = 0 \quad \text{for } x \to \infty\,, \\ &u_x = 0\,, \quad v_x = 0\,, \quad w = 0 \quad \text{for } x \to -\infty\,. \end{aligned} \tag{5}$$

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 ($w = 0$). 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 (4). Therefore the derivatives of u and v are zeros and $w = 0$ for $x \to -\infty$.

2.1 Travelling wave solution

We seek a solution to the problem (4)–(5) in the form of a travelling wave $u(x,t) = u(\xi)$, $v(x,t) = v(\xi)$, and $w(x,t) = w(\xi)$, where we introduce $\xi = x - ct$, a coordinate in the moving frame and c is the speed of the travelling wave. Substituting the travelling wave solution into the governing equations we obtain

$$\begin{aligned} &u_{\xi\xi} + cu_\xi + rw = 0\,, \\ &\tau_A v_{\xi\xi} + cv_\xi - \beta vwe^{-1/u} = 0\,, \\ &\tau_B w_{\xi\xi} + cw_\xi + \beta vwe^{-1/u} - r\beta w = 0\,, \end{aligned} \tag{6}$$

and boundary conditions

$$u = 0\,, \quad v = 1\,, \quad w = 0 \quad \text{for } \xi \to \infty\,,$$

$$u_\xi = 0\,, \quad v_\xi = 0\,, \quad w = 0 \quad \text{for } \xi \to -\infty\,. \tag{7}$$

Following Simon et al. [14] and Zeldovich et al. [18] we consider the case when Lewis numbers for the fuel and the radicals are equal to unity (that is, we assume equal diffusivity of the reactant, the radical and heat). Although this assumption simplifies the problem significantly, it still allows the investigation of the generic properties of the system (6) and (7).

In the case $\tau_A = \tau_B = 1$ equations (6) possess an integral $S = \beta u + v + w$. Using S and the first boundary condition in (7), equations (6) can be reduced to a system of two second order equations for the temperature and the fuel concentration

$$\begin{aligned} &u_{\xi\xi} + cu_\xi + r(1 - \beta u - v) = 0\,, \\ &v_{\xi\xi} + cv_\xi - \beta v(1 - \beta u - v)e^{-1/u} = 0\,, \end{aligned} \tag{8}$$

where $w = 1 - \beta u - v$. On the right boundary ($\xi \to \infty$) we require that $u = 0$ and $v = 1$, whereas on the left boundary ($\xi \to -\infty$) we modify the boundary conditions to

$$u = \beta^{-1}(1 - \sigma)\,, \quad v = \sigma\,, \tag{9}$$

where σ denotes the residual amount of fuel left behind the wave and is unknown until a solution is obtained. We note here that at first glance, system (8) looks very similar to the equations describing the dynamics of the one-step adiabatic reaction model considered by Weber et al. [16]. However, in contrast to the one-step adiabatic case, equations (8) do not have an integral which enabled further simplification as given by Weber et al. [16]. Moreover, boundary conditions (9) suggest that there can be some fuel left behind the reaction zone, which is impossible in the one-step adiabatic reaction model case.

3 Travelling front solutions

The solutions to equations (8) subject to the boundary condition (7) exhibit travelling front solutions similar to those found for the one-step reaction models, such that $u(\xi)$ and $v(\xi)$ are monotonic and $u_\xi(\xi)$, $v_\xi(\xi)$ and $w(\xi)$ are bell shaped functions of the spatial coordinate. Typical solution profiles $u(\xi)$, $v(\xi)$, and $w(\xi)$ are shown in Figures 1 and 2 for $r = 0.001$, $\beta = 1.0$ and $\beta = 4.0$ respectively. Note that numerically we determined the extinction value of β to be $\beta \approx 4.26\cdots$. Hence in Figure 1 the nondimensional activation energy, β, is well below the extinction value, and the concentration of radicals, which is shown with the dotted line, can reach values of the order $\mathcal{O}(1)$. Parameter σ becomes negligible and no fuel leakage from the reaction zone is observed. However, in Figure 2 (when the value of β is close to the extinction value) the residual amount of fuel, σ, becomes significant indicating that the conversion of fuel is incomplete and a fuel leakage occurs. The fuel decomposition reaction region has the same width as the recombination reaction zone indicating that close to the extinction limit the flame structure changes to the regime similar to the fast recombination regime of the Zeldovich–Liñán model.

The properties of the travelling front solutions are summarized in Figure 3 where the speed of the front, c, is shown as functions of β for several values of r. At first glance, the dependence of c on β in Figure 3 resembles the behaviour of the speed of the front for the model with one-step reaction scheme, which was studied by Gubernov et al. [4]. Namely, c reaches the global maximum for some value of β and decays monotonically as we increase (or decrease) β from the value corresponding to the maximum. However, more detailed investigation shows that there is a significant difference between the prediction of the one- and two-step models. In particular, for the model with one-step reaction mechanism the travelling front solution exists for any value of β and its speed decays exponentially to zero as we increase β. This is not the case for the model with two-step reaction mechanism studied here. As we increase β (for a fixed r) up to some critical value β_c, the

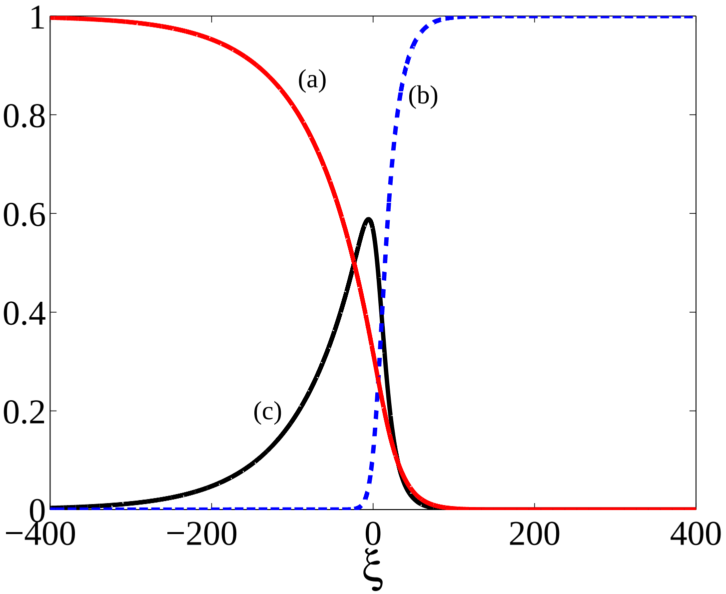

Figure 1: Travelling front solution profiles for: (a) temperature $\beta u(\xi)$; (b) fuel $v(\xi)$; and (c) radical $w(\xi)$; for $\beta = 1.0$, $r = 0.001$.

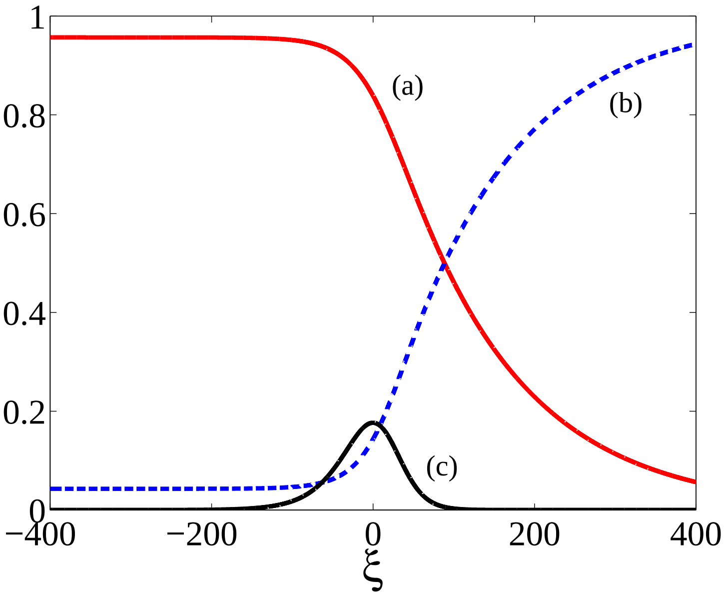

Figure 2: Travelling front solution profiles for: (a) temperature $\beta u(\xi)$; (b) fuel $v(\xi)$; and (c) radical $10w(\xi)$; for $\beta = 4.0$, $r = 0.001$.

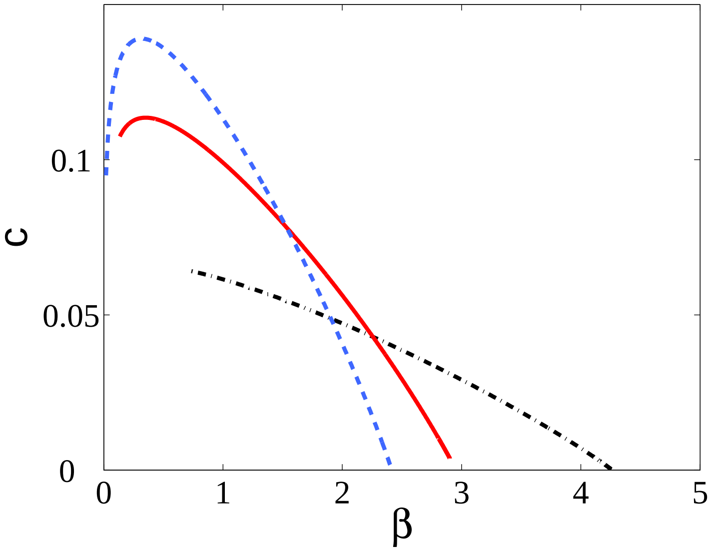

Figure 3: Speed of the front as a function of β for various values of r. The solutions for $r = 0.001$, 0.005 and 0.01 are denoted by the black dashed-dot, red bold and blue dashed curves respectively.

speed of the front decays rapidly and the travelling front solution ceases to exist for $\beta > \beta_c$. Furthermore, there is some residual amount of fuel left behind the front in case of the two-step model unlike the one-step adiabatic model, which has zero residual fuel. To some extent, the properties of the two-step adiabatic model studied here resemble the properties of the nonadiabatic one-step model investigated by Gubernov et al. [5], more than the adiabatic one-step studied by Gubernov et al. [4]. This is expected since the recombination step, which is an inhibitor of the chain branching reaction, plays a similar role as the heat exchange with the surrounding medium in the one-step nonadiabatic model. In both cases there is a nonzero residual amount of fuel left behind the reaction zone. The similarity between these two cases is also strengthened by the likeness of the behaviour of the speed of the front as a function of the parameter β. Namely, in both the one-step nonadiabatic and the two-step adiabatic models, the travelling front solution ceases to exist as we approach some critical value of β_c (in the combustion literature this event is usually called extinction as stated by Gubernov et al. [5]). However, the route to extinction in these models is different. In the case of the one-step nonadiabatic model, for given parameter values, there are either two solution branches with different speeds or no solutions. The extinction occurs when the two solution branches coalesce (this event is also known as the turning point or the fold bifurcation). For the two-step reaction mechanism the extinction occurs when the speed of the front drops down to zero as we approach the critical parameter values.

4 Conclusions

We undertook a preliminary investigation into the combustion wave propagation in an adiabatic model with two-step chain branching reaction mechanism. In contrast to Dold and Weber [3] we do not use the activation energy asymptotic approach and consider the model for general values of activation energy. We also used different nondimensional parameters which enabled more convenient comparison between the properties of one- and two-step models. The model exhibits travelling combustion front solutions. The properties of these solutions differ from the properties of one-step models. Combustion waves exist in certain regions of the parameter space and are characterized by non-zero residual amount of reactant, σ, left behind the travelling wave. This is not possible in one-step adiabatic flame models as all the fuel is used up in such models. To some extent this characteristic of the two-step adiabatic model is similar to the properties of premixed combustion waves in nonadiabatic one-step models, which can also exist in a certain region of parameter space and exhibit extinction as the boundaries of this region are reached. We are presently investigating the stability of these travelling wave solutions, and extending our work to cases when the Lewis numbers for the fuels and the radicals are *not* equal to unity as we have done in this investigation.

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