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Behaviour of combustion waves in one-step and two-step models

Sharples, J.J. [1], V.V. Gubernov [2], H.S. Sidhu [1], I.N. Towers [1], A.V. Kolobov[2] and A.A. Polezhaev[2]

[1] School of Physical Environmental and Mathematical Sciences University of New South Wales at the Australian Defence Force Academy, Australian Capital Territory
[2] I.E. Tamm Theory Department, P.N. Lebedev Physical Institute of the Russian Academy of Sciences, 53 Leninsky Prospect, Moscow
Email: j.sharples@adfa.edu.au

Abstract: Premixed combustion waves in models with various reaction mechanisms have drawn the attention of researchers for a long period of time. Such models are typically described by parabolic reaction-diffusion systems of partial differential equations relating to energy conservation and the conservation of the chemical species involved in the various steps of the combustion reaction. Reaction-diffusion systems corresponding to combustion are distinguished by the strong nonlinear dependence of the reaction rates on temperature, which are often modelled by the Arrhenius law or some large-exponent power law.

Advances in computational power have allowed detailed numerical study of reaction mechanisms involving a variety of different steps. While these investigations have been useful in providing some quantitative predictions for observed phenomena, there is still uncertainty about the reliability of these complex models when applied to the prediction of the generic behaviour of flames. To avoid this uncertainty considerable effort has been put into the study of reduced reaction mechanisms that involve only one or two steps. One-step models have the advantage of being relatively simple, allowing analytical investigations into phenomena such as ignition and extinction of flames, which have been proved useful and qualitatively correct. However, in many reactions models assuming one-step reaction mechanisms can lead to misleading results. The logical next step is to consider two-step reaction mechanisms that have shown promise in capturing the essential behaviour of more complicated reaction schemes.

In this paper we discuss the generic properties of travelling combustion wave solutions to models involving one- and two-step reaction mechanisms assuming Arrhenius kinetics. In particular, for one-step models we discuss the dependence of the speed of the propagating flame front on the activation energy, Lewis number and the effects of heat loss, all of which can be controlled in a laboratory setting. In addition we present a summary discussion on the stability properties of one-step combustion waves. Stability of combustion waves is an important issue in applications such as self-propagating high-temperature synthesis (SHS) of advanced materials, where pulsating instabilities in combustion waves can lead to undesirable laminar irregularities in the product material. Understanding the dependence of flame speed and stability on the various prescribable parameters is therefore a subject of considerable practical interest.

The generic properties of solutions of the one-step adiabatic and nonadiabatic models are compared to those of an adiabatic model assuming a two-step chain branching and recombination mechanism. For the two-step reaction scheme it is found that the fuel Lewis number has a considerable effect on the qualitative dependence of the wave speed on the activation energy, while the Lewis number for radicals does not greatly alter the generic behaviour. In particular, when the fuel Lewis number is less than unity, travelling combustion wave solutions are stable and propagate with a speed uniquely defined by a monotonically decreasing function of the activation energy. Solutions exist for all values of the activation energy up to a finite value corresponding to a wave speed of zero (extinction). On the other hand, when the fuel Lewis number is greater than unity the wave speed is double-valued, with unstable 'slow' solutions and 'fast solutions that are either stable or that become unstable due to the onset of pulsations.

Keywords: *Combustion waves, multi-step kinetics, wave speed, stability, bifurcation*

1. INTRODUCTION

Understanding combustion is important for a variety of applications including industrial processes, environmental problems such as bushfires, and the synthesis of advanced materials. As a consequence, models that simulate combustion processes have been studied by researchers over a long period of time (Merzhanov and Rumanov, 1999). Of particular importance is the manner in which flames will propagate through a particular mixture. As such, there has been considerable interest in reaction-diffusion systems that admit travelling wave solutions, which in the context of combustion, are called combustion waves. A combustion wave describes a propagating flame front, which marks the transition from the initial mixture to the reactant product equilibrium phase. Reaction-diffusion systems describing combustion processes are distinguished by the extremely strong nonlinear dependence of the reaction rate on temperature, usually described by the Arrhenius law. The strong nonlinear nature of combustion models complicates their analysis; investigations typically rely on asymptotic methods or advanced numerical techniques as analytical solutions are rarely available.

Basic models used to describe combustion processes often assume one-step chemistry, where fuel (F) and oxidant (O_2) combine to produce products (P) and heat through the generic kinetic scheme $F + O_2 \rightarrow P + heat$. One-step models have the advantage of being amenable to analytical investigation through asymptotic methods, and have led to useful and qualitatively correct predictions of ignition, extinction and stability of diffusion flames and the propagation and stability of premixed flames, etc. (Merzhanov and Rumanov, 1999).

In the majority of cases, however, the chemical reactions in a flame front proceed according to a complicated mechanism involving a series of reaction steps, each with its own set of intermediate chemical species. Thus in many cases erroneous results can be produced by models that assume one-step chemistry (Westbrook and Dryer, 1981). In an effort to improve predictions of generic flame behaviour, without having to include all the effects of detailed multi-step kinetics, researchers have considered reduced kinetic mechanisms. These models are more amenable to analytical investigation than the detailed schemes and are still able to provide accurate predictions of the main flame characteristics for some reactions. An example of a reduced kinetic mechanism is the two-step scheme that involves autocatalytic chain branching $A + B \rightarrow 2B$ and recombination $B + M \rightarrow C + M + heat$, where A is the fuel, B is the radical, C the product and M a third species. In this scheme the recombination phase acts as both an inhibitor, which terminates the chain branching, and an accelerant that produces heat.

In this paper we report on investigations into the properties of combustion wave solutions to the model involving the two-step chain-branching reaction mechanism. Combustion models with chain-branching mechanisms usually involve the radical recombination reaction of second-order and higher (Zel'dovich et al., 1985). In this paper we study a model with the first-order recombination reaction introduced by Dold (2007), making comparisons with known results for one-step adiabatic and nonadiabatic models. In particular we discuss the dependence of the combustion wave speed on the various prescribable parameters, and compare the generic behaviour for the one- and two-step reaction mechanisms. In section 2 we outline the reaction-diffusion systems used to model flame propagation, in section 3 we discuss the travelling wave formulation of the systems, the solutions of which are discussed in section 4. In section 5 we give a brief account of some of the stability properties of combustion wave solutions

2. COMBUSTION WAVE PROPAGATION MODELS

In this section we briefly introduce the reaction-diffusion systems of partial differential equations used to model flame propagation with one- and two-step reaction mechanisms. The results pertaining to models with one-step reaction mechanisms are not new. However, due to the limited format, we are not able to give a comprehensive overview of the literature. Instead we refer the reader to the list of references, the papers cited within them and other sources.

2.1. One-step model

(a) Adiabatic case: We consider flame propagation through a premixed fuel in one spatial dimension with combustion described by the one-step kinetic scheme $F + O_2 \rightarrow P + heat$. Assuming that no heat is lost to the surrounding media and that the rate of the exothermic combustion is given by the Arrhenius law, the equations describing conservation of energy and mass lead to the following parabolic system of reaction-diffusion equations (Weber et al., 1997):

$$u_t = u_{xx} + v e^{-1/u}, \quad v_t = \tau v_{xx} - \beta v e^{-1/u} \tag{1}$$

The system is written in non-dimensional form. The quantities u and v are the non-dimensional temperature and concentration of fuel, respectively, τ is the inverse of the Lewis number (ratio of diffusion rates of mass and heat) and β is the ratio of activation energy to heat release. The variables t and x are non-dimensional time and space coordinates.

We consider (1) subject to the boundary conditions

$$\begin{aligned} &u(x,t) = u_a + \beta^{-1}, \quad && v(x,t) = 0 \quad \text{as} \quad x \to -\infty, \\ &u(x,t) = u_a, && v(x,t) = 1 \quad \text{as} \quad x \to +\infty. \end{aligned} \tag{2}$$

On the right boundary ($x \to +\infty$) we have a cold ($u = u_a$) and unburned ($v = 1$) state, whereas on the left boundary ($x \to -\infty$) we have a hot ($u = u_a + \beta^{-1}$) and burned ($v = 0$) state. We denote the (dimensionless) ambient temperature by u_a. Typically u_a will be small compared to the reaction temperature and in many treatments it is assumed that $u_a = 0$ (e.g. Weber et al., 1997). This assumption does not change the qualitative behaviour of the combustion wave solutions and circumvents the so-called 'cold boundary problem' that arises when using the Arrhenius law to describe the reaction rates. The 'hot' boundary condition $u = u_a + \beta^{-1}$ arises from an integral relation for the travelling wave solution (see section 3).

(b) Nonadiabatic case: The adiabatic scheme described above is of restricted validity in real world applications, where consideration of the effects of heat loss to the surrounding media is essential. Assuming that heat is lost to the surroundings through Newtonian cooling, the equations (1) are amended as follows (Weber et al., 1997):

$$u_t = u_{xx} + v e^{-1/u} - l(u - u_a), \quad v_t = \tau v_{xx} - \beta v e^{-1/u} \tag{3}$$

where now l is the heat loss coefficient. The heat loss term refers to heat fluxes orthogonal to the spatial dimension denoted by x, e.g. if the one-dimensional model is taken to describe flame propagation along a cylinder of infinite length (with a priori averaging over the radial dimension) the heat loss term refers to radial heat loss. In order for solutions to exist, l must be sufficiently small.

We consider (3) subject to the boundary conditions

$$\begin{aligned} &u(x,t) = u_a, \quad v(x,t) = \sigma \quad \text{as} \quad x \to -\infty, \\ &u(x,t) = u_a, \quad v(x,t) = 1 \quad \text{as} \quad x \to +\infty. \end{aligned} \tag{4}$$

The conditions on the right boundary ($x \to +\infty$) correspond to ambient temperature ($u = u_a$) and unburned fuel ($v = 1$), while on the left boundary ($x \to -\infty$) the inclusion of heat loss means that temperature will return to the ambient value ($u = u_a$) and that there will be a residual amount of fuel ($v = \sigma$) left behind the propagating flame front.

2.2. Two-step model

We consider an adiabatic model for premixed flame propagation in one spatial dimension with combustion described by the two-step kinetic scheme involving autocatalytic chain branching $A + B \to 2B$ and recombination $B + M \to C + M + heat$. The energy and mass conservation equations governing this process can be written in non-dimensional form as (Gubernov et al., 2008)

$$u_t = u_{xx} + r w, \quad v_t = \tau_A v_{xx} - \beta v w e^{-1/u}, \quad w_t = \tau_B w_{xx} + \beta v w e^{-1/u} - r \beta w \tag{5}$$

where u, v and w are the non-dimensional temperature, concentration of fuel (A) and concentration of radicals (B), respectively; x and t are dimensionless space and time coordinates, τ_A and τ_B are the inverse of the Lewis numbers for fuel and radicals, β is the ratio of activation energy to heat release and r is the ratio of the reaction constants for recombination and chain branching.

The equations (5) are considered subject to the boundary conditions

$$\begin{aligned} &u(x,t) = u_a, \quad v(x,t) = 1, \quad w(x,t) = 0 \quad \text{as} \quad x \to +\infty, \\ &u_x(x,t) = 0, \quad v_x(x,t) = 0, \quad w(x,t) = 0 \quad \text{as} \quad x \to -\infty. \end{aligned} \tag{6}$$

On the right boundary ($x \to +\infty$) we have a cold ($u = u_a$) and unburned ($v = 1$) state where fuel has not yet been consumed 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 the fuel can be specified, but requiring that no reaction is taking place implies that u and v are constant and that $w = 0$.

3. TRAVELLING WAVE SOLUTIONS

3.1. One-step model

(a) Adiabatic case: To obtain travelling wave solutions of the system (1) we introduce the ansatz

$$u(x,t)=u(\xi),\quad v(x,t)=v(\xi) \tag{7}$$

where $\xi = x - ct$ is a coordinate frame that moves with constant speed c. Substitution of (7) into (1) reduces the reaction-diffusion system (1) to a system of ordinary differential equations

$$u_{\xi\xi}+cu_{\xi}+ve^{-1/u}=0,\quad \tau v_{\xi\xi}+cv_{\xi}-\beta ve^{-1/u}=0 \tag{8}$$

with boundary conditions

$$\begin{aligned} &u=u_a+\beta^{-1}, && v=0 \quad \text{as} \quad \xi\to-\infty,\\ &u=u_a, && v=1 \quad \text{as} \quad \xi\to+\infty. \end{aligned} \tag{9}$$

Adding the second equation in (8) to the first multiplied by β and integrating from $-\infty$ to $+\infty$ yields the condition $u = u_a + \beta^{-1}$. Thus, without heat loss, the temperature on the 'hot' boundary is the maximum temperature achieved by the flame.

(b) Nonadiabatic case: Similarly for the nonadiabatic case, substitution of (7) into (3) results in the system of ordinary differential equations

$$u_{\xi\xi}+cu_{\xi}+ve^{-1/u}=l(u-u_a),\quad \tau v_{\xi\xi}+cv_{\xi}-\beta ve^{-1/u}=0 \tag{10}$$

with boundary conditions

$$\begin{aligned} &u=u_a, && v=\sigma \quad \text{as} \quad \xi\to-\infty,\\ &u=u_a, && v=1 \quad \text{as} \quad \xi\to+\infty. \end{aligned} \tag{11}$$

Adding the second equation in (10) to the first equation multiplied by β, integrating and imposing the boundary conditions (11) yields $c(1-\sigma) = \beta l u^*$, where u^* denotes the integral of u from $\xi = -\infty$ to $\xi = +\infty$. The residual amount of fuel σ is therefore a function of the mean temperature, the prescribed parameters β and l, and the internal wave speed parameter c.

3.2. Two-step model

Imposing the ansatz $u(x, t) = u(\xi)$, $v(x, t) = v(\xi)$ and $w(x, t) = w(\xi)$, where $\xi = x - ct$ is a coordinate in the moving frame and c is the speed of the travelling wave, the reaction-diffusion system (5, 6) reduces to the system of ordinary differential equations

$$u_{\xi\xi}+cu_{\xi}+r\,w=0,\quad \tau_A v_{\xi\xi}+cv_{\xi}-\beta vwe^{-1/u}=0,\quad \tau_B\, w_{\xi\xi}+cw_{\xi}+\beta vwe^{-1/u}-r\beta w=0, \tag{12}$$

subject to the boundary conditions

$$\begin{aligned} &u=u_a, && v=1, \quad w=0 \quad \text{as} \quad \xi\to+\infty,\\ &u_{\xi}=0, && v_{\xi}=1, \quad w=0 \quad \text{as} \quad \xi\to-\infty. \end{aligned} \tag{13}$$

The boundary conditions (13) can be modified by multiplying the first equation in (12) by β, and adding the second and third equations. Integrating the resultant sum from $\xi = -\infty$ to $\xi = +\infty$ and using (13) yields the following condition

$$\int_{-\infty}^{\infty}\left(\beta u+v+w\right)_{\xi}d\xi=0,\quad \text{or}\quad \beta\lim_{\xi\to-\infty}u=1-\lim_{\xi\to-\infty}v.$$

Hence the boundary conditions (13) may be modified as

$$\begin{aligned} &u=u_a, && v=1, \quad w=0 \quad \text{as} \quad \xi\to+\infty,\\ &u=\beta^{-1}(1-\sigma), && v=\sigma, \quad w=0 \quad \text{as} \quad \xi\to-\infty. \end{aligned} \tag{14}$$

where σ denotes the residual amount of fuel left behind the propagating flame, which is a function of the prescribed parameters τ_A, τ_B, β and r.

4. PROPERTIES OF TRAVELLING COMBUSTION WAVES

Combustion wave solutions to the systems (8, 9), (10, 11) and (12, 14) are determined numerically using shooting and relaxation methods. The shooting methods employ fourth-order and fifth-order Runge-Kutta algorithms to obtain approximate solutions of the single-step and two-step systems, respectively, over the interval $[-L_1, L_2]$, where L_1, $L_2 > 0$ are sufficiently large. The approximate solutions are then refined using relaxation methods, which deliver solutions with an associated error less than 10^{-15}. See Gubernov et al. (2003) for more details and references.

The numerical methods described above allow properties of the combustion waves, such as their speed, to be determined. For the single-step systems, the dependence of the speed c of the flame front on the prescribed parameters $\{\tau, \beta\}$ and $\{\tau, \beta, l\}$ is obtained, while for the two-step system the speed of the flame front depends on the prescribed parameters $\{\tau_A, \tau_B, \beta, r\}$.

4.1. One-step model

The shooting and relaxation methods were used to obtain values of the speed of the combustion wave for a variety of values of the prescribed parameters $\{\tau, \beta\}$ and $\{\tau, \beta, l\}$. The speed of the flame front exhibits fundamentally different behaviour for the adiabatic and nonadiabatic cases.

(a) Adiabatic case: In the adiabatic model the speed of the combustion wave corresponds to the surface $c(\beta, \tau)$. Plots of the wave speed as a function of the activation energy β and for a number of different values of τ can be seen in figure 1a. The speed c decreases monotonically with β in all cases. For large values of β the speed decays according to $\tau\beta c^2 \sim 2e^{-\beta}$. Typical profiles of temperature and fuel are also shown in the inset panel of figure 1a.

(b) Nonadiabatic case: For the nonadiabatic model the speed of the combustion wave depends on the three parameters β, τ and l. Several cross-sections of the wave speed $c(\beta, \tau, l)$, taken parallel to the β-axis for $\tau = 0.5$ and a number of different values of l can be seen in figure 1b. In contrast to the adiabatic case the wave speed in the nonadiabatic case is double-valued with respect to β. That is, for each value of β the solution possesses a 'fast' and 'slow' branch. The 'slow' branches correspond to the bottom part of the curves in figure 1b, while the 'fast' branches correspond to the top part. Both solution branches exhibit monotonic behaviour with respect to β (for constant τ and l) and l (for constant β and τ). Also in contrast to the adiabatic case there is a distinct value of β beyond which no travelling wave solutions exist (the turning point on the curves in figure 1b). This is referred to as the extinction point. The value of β corresponding to extinction is denoted β_e. Thus there are two propagating fronts for $\beta < \beta_e$ and no solutions for $\beta > \beta_e$. Similarly there is a critical value of the heat loss parameter corresponding to extinction; if l is below this critical value two solutions exist, while if l is above the critical value there are no solutions. Typical profiles of temperature and fuel for the nonadiabatic case are shown in the inset panel of figure 1b.

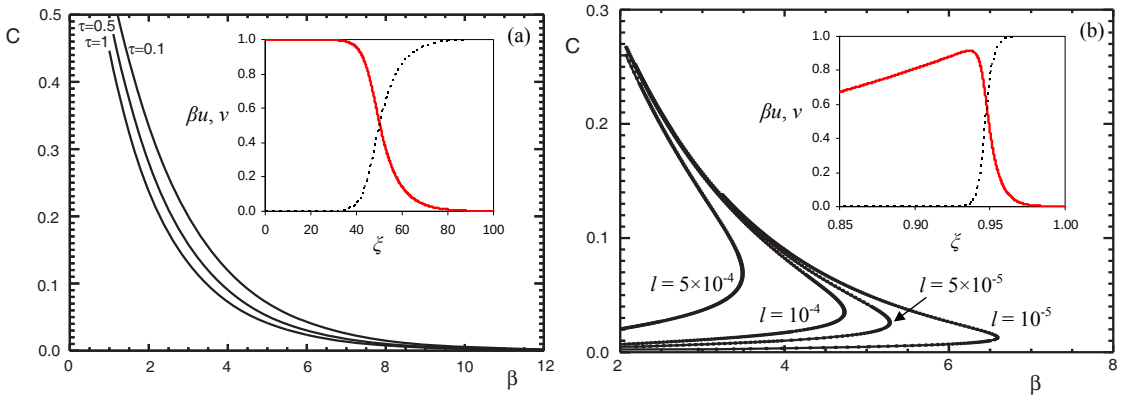

Figure 1. (a) Wave speed c as a function of β for the single step adiabatic model with different values of the parameter $\tau = 0.1, 0.5, 1.0$. The inset shows the temperature βu (red solid) and fuel v (black dashed) profiles for $\beta = 3$ and $\tau = 1.0$ with $c = 0.13$ (b) Wave speed c as a function of β for the single step nonadiabatic model with $\tau = 0.5$ and four different values of $l = 1\times10^{-5}, 5\times10^{-5}, 1\times10^{-4}, 5\times10^{-4}$. The inset shows the temperature βu (red solid) and fuel v (black dashed) profiles for $\beta = 3$, $\tau = 0.5$ and $l = 5\times10^{-4}$, with $c = 0.14$.

4.2. Two-step model

The behaviour of the adiabatic model assuming a two-step reaction mechanism differs substantially from that assuming single-step chemistry. In particular, for the two-step model there exists a critical value $\beta = \beta_e$ beyond which the solution ceases to exist. This is in contrast to the adiabatic model with single-step reaction mechanism for which solutions existed for all values of β.

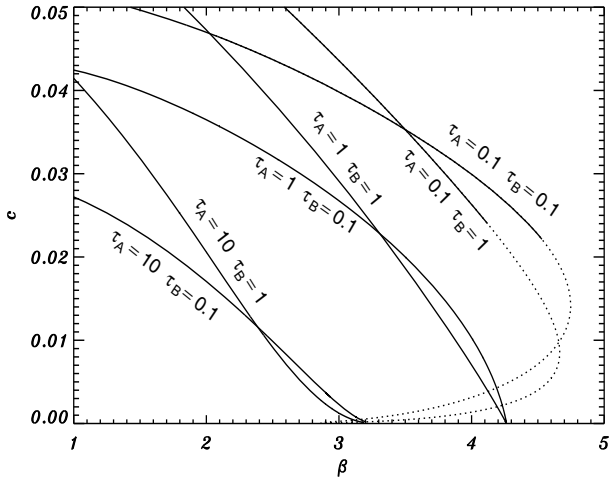

Figure 2. Wave speed c as a function of β for the two-step adiabatic model for $r = 0.001$ and various values of the parameters τ_A and τ_B.

The Lewis number for the fuel (τ_A^{-1}) has a significant effect on the wave speed profile, whereas variation of the Lewis number for the radicals (τ_B^{-1}) produces some quantitative differences but does not affect the overall behaviour of the solutions. In the case $\tau_A > 1$, the wave speed decays monotonically with β, and approaches zero as $c \sim (\beta - \beta_e)^2$, while for $\tau_A = 1$, the speed c is still a monotonically decreasing function of β, but now approaches zero in a linear fashion. The dependence of flame speed c on β changes significantly when $\tau_A < 1$. In this case $c(\beta)$ becomes double-valued: there are either two solutions with different flame speeds or the solutions cease to exist due to extinction above some threshold value β_e where the fast and slow solution branches meet. The dependence of the flame speed on β for various values of the parameters τ_A, τ_B with $r = 0.001$ can be seen in figure 2.

5. STABILITY OF TRAVELLING WAVE SOLUTIONS AND PULSATING WAVES

5.1. One-step model

(a) Adiabatic case: The travelling wave solutions are either stable or exhibit pulsating instabilities as β reaches a critical value $\beta_h < \beta_e$, corresponding to a Hopf bifurcation. For $\tau^{-1} >> 1$ the value of β_h tends to a finite limit value, whereas if $\tau \to 1$, then $\beta_h \to \infty$.

(b) Nonadiabatic case: The 'slow' solution branch is always unstable due to uniform instability. The 'fast' solution branch is either stable or exhibits pulsating instability due to the Hopf bifurcation. There is a critical value τ_{BT} (corresponding to a Bogdanov-Takens bifurcation) such that for $\tau > \tau_{BT}$ the 'fast' branch is stable for all β up to $\beta = \beta_e$ (turning point); for $\tau < \tau_{BT}$ the solution is stable for $\beta < \beta_h$ and unstable for $\beta > \beta_h$. β_h originates from the extinction locus at $\tau = \tau_{BT}$ and approaches a finite asymptotic value as $\tau^{-1} \to \infty$.

5.2. Two-step model

We investigate the stability of the travelling wave solutions using the Evans function (Gubernov et al., 2003) and direct integration of the partial differential equations using finite difference methods. Travelling wave solutions exist above the extinction curve in figure 4. The 'slow' solution branch, which exists for $\tau_A < 1$, is always unstable, while the 'fast' branch ($\tau_A > 1$)

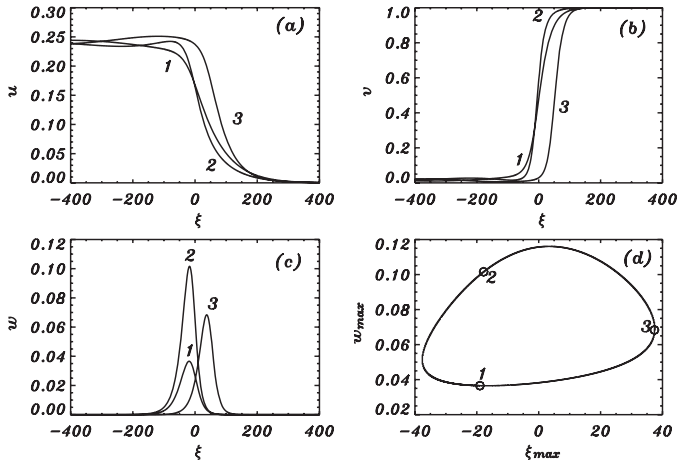

Figure 3. (a) Temperature u, (b) concentration of fuel v, (c) concentration of radicals w as functions of the moving coordinate ξ for the pulsating combustion wave with $\tau_A^{-1} = 3$, $\tau_B^{-1} = 1$ and $\beta = 4.08$ ($\beta_h \approx 4.0703$). Solution profiles are sampled at $t_1 = 0$, $t_2 = 8750$ and $t_3 = 17500$ and are marked as 1, 2 and 3, respectively. Panel (d) shows values of the maximum value of w plotted against the location of the maximum value of w.

is either stable or exhibits pulsating instabilities due to the Hopf bifurcation once the Hopf locus is crossed. An example of a pulsating solution can be seen in figure 3. The sections of the $c(\beta)$ profile corresponding to unstable solutions are represented by dotted lines in figure 2. The Bogdanov-Takens bifurcation is now anchored at $\tau_A = 1$, therefore in contrast to the one-step nonadiabatic case, the pulsating instability exists for all $\tau_A < 1$. For $\tau_A > 1$ (for which $c(\beta)$ is monotonic) the solutions are stable over the whole range of existence. These results are summarised in the bifurcation diagram, figure 4.

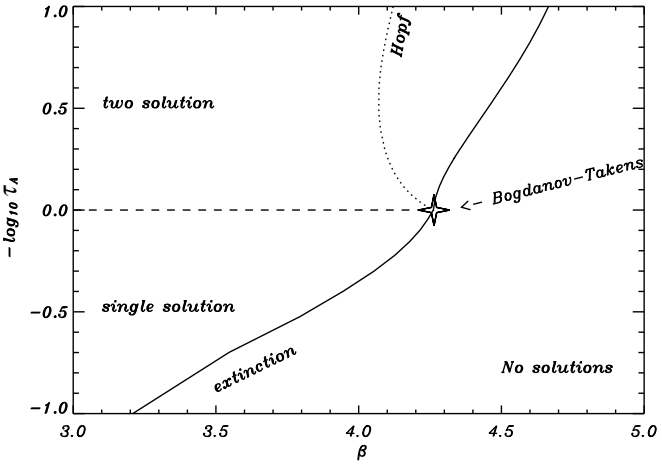

Figure 4. Bifurcation diagram for τ_B =1. The solid and dotted curves are the loci of critical parameter values for extinction and Hopf bifurcation, respectively.

6. DISCUSSION AND CONCLUSIONS

The generic features of one-dimensional one-step adiabatic and nonadiabatic premixed combustion wave solutions were summarised and compared with the results of investigations into a one-dimensional adiabatic model with a two-step chain branching reaction mechanism. In the two-step model the parameter τ_A had a substantial effect on the qualitative properties of combustion waves, while τ_B had only a quantitative effect.

For $\tau_A > 1$ the travelling wave was stable with the speed defined as a monotonically decreasing function of β. Solutions were found to exist for values of β up to a finite value β_e, corresponding to extinction. The speed approached zero as β approached β_e. For $\tau_A < 1$, on the other hand, $c(\beta)$ was double-valued, with the travelling wave solution exhibiting extinction for finite values of β, in contrast to the one-step adiabatic case. Extinction in the two-step $\tau_A < 1$ case also corresponds to $c \neq 0$ due to a turning point bifurcation, in contrast to the $\tau_A > 1$ case. The 'slow' solution branch was always unstable, while the 'fast' solution branch could be either stable or could lose stability with respect to pulsating instabilities due to a Hopf bifurcation. For $\tau_A \to 1$ the Hopf and extinction loci coincide due to the Bogdanov-Takens bifurcation. In these types of models it appears that the Bogdanov-Takens bifurcation point is the key to predicting instabilities.

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