

Properties of combustion waves in the model with competitive exo- and endothermic reactions

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Abstract In this paper we investigate the properties of the travelling combustion wave solutions in a diffusion-thermal model with a two-step competitive exo–endothermic reaction mechanism in one spatial dimension under adiabatic conditions. The model is analysed both numerically and analytically using asymptotic analysis. It is demonstrated that depending on the parameter values, the flame speed as a function of parameters is either a single-valued monotonic function or a double-valued c-shaped function with the turning point type of behaviour. For the case of single-valued flame speed, two flame regimes are identified: the regime with exo- and endothermic reaction domination. Two different routes to extinction are found as well as regions of the existence of combustion waves in the parameter space. Prospects of further work are also discussed.

Keywords Combustion waves · Competitive exo–endothermic reaction · Flame speed

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1 Introduction

This work concerns the existence and propagation of reaction fronts through reactive media where diffusive processes are present and where a main exothermic reaction is accompanied by an endothermic reaction as well. A number of authors have addressed problems where an independent endothermic process affects the progress of a combustion front [4, 8–10]; our concern in this paper is with the possibility of competitive endothermic and exothermic reactions, where the same reactive material provides the feed for both reactive steps [6]. Though most observed physico-chemical phenomena are a consequence of several, often numerous, concurrent or consecutive endothermic and exothermic reactive processes, useful understanding can often be gained by considering much simpler lumped models which reproduce the essential phenomenology. In some cases, notably when thermal effects are prominent in the process, the simplest useful model comprises a pair of reactions, one exothermic and one endothermic, characterised by different chemical kinetics. These reactions may feed on the same unique reactant material, so-called competitive reactions, or each reaction may independently consume a different reactant, so-called parallel reactions [2]. In the parallel case the coupling between the reactions is solely thermal, whereas in the competitive case there is a second coupling through the reactant consumption. In contrast to the case of parallel reactions, which has been widely studied, competitive reactions have received little attention. This situation is undesirable given the appropriateness of competitive schemes in modelling decomposition or pyrolysis processes [1, 12] and their applicability to ammonium nitrate based explosives [11]. An exception to this observation is the study by Calvin et al. [3] which established the existence of combustion wave multiplicity in the case of competing exothermic reactions. Whereas, in the parallel case, the net enthalpy production by complete consumption of both reactants is uniquely determined, this is not true for competitive reactions, where the net production depends on the full time history of the process; if the temperature is kept relatively low, by thermal diffusion or other extraneous effects e.g. Newtonian or radiative cooling, the net production may be, counterintuitively, actually increased. Hmaidi et al. [6] investigated the existence and stability of travelling one-dimensional reaction fronts propagating through a solid reactive slab (infinite Lewis number), effectively extending the work of Matkowsky and Sivashinsky [7] to the case where heat is lost through a competitive endothermic reaction term. The behaviour of the competitive system was modelled by regarding the endothermic reaction as a perturbation to an exothermic reaction. This necessitated some restrictions on the ordering of the kinetic parameters of the endothermic step. Specifically, the endothermic reaction was assumed to have twice the activation energy of the exothermic reaction and a pre-exponential frequency term much greater than that for the exothermic reaction. In the present paper we scrutinise the consequences of these assumptions, particularly with reference to the relative magnitudes of the activation energies. Moreover, we modify the ratio of activation energies so that the endothermic reaction plays a greater role in the overall reaction scheme and examine the behaviour of the resultant reaction fronts.

The work reported here therefore extends the work [6] and our previous work [13] to consider a more comprehensive range of parameter values. We again consider

propagation of a reaction front in which the driving exothermic reaction competes with an endothermic reaction that consumes both reactant and heat within the system. As mentioned, the parameter values we assume permit a stronger contribution from the endothermic reaction. Following [6], we assume adiabatic conditions, though unlike [6] we allow for the diffusion of reactant as well as heat [13]. We also relax the restriction of large activation energies and pre-exponential frequency of the endothermic reaction.

2 Model

We consider a diffusional-thermal model with two-step kinetics for premixed combustion wave propagation in one spatial dimension under adiabatic conditions. It is assumed that the reactant undergoes two competitive reactions: one exothermic and one endothermic, and that the reaction products are chemically inert and have no effect on physical properties such as the diffusivity of the reaction surroundings. Arrhenius kinetics are assumed for both reactions. The nondimensional equations governing this process can be found in [13] and can be written as

$$\begin{aligned} u_t &= u_{xx} + v(e^{-1/u} - qre^{-f/u}), \\ v_t &= L^{-1}v_{xx} - v\beta(e^{-1/u} + re^{-f/u}), \end{aligned} \quad (1)$$

where t and x are non-dimensional time and space coordinates; u and v are the dimensionless temperature and fuel concentration; β is the dimensionless activation energy of the exothermic reaction; q is the ratio of the enthalpies of the endo- to exothermic reaction; r is the ratio of pre-exponential factors of the endothermic to exothermic reaction; f is the ratio of the activation energy of endothermic to exothermic reaction; L is the Lewis number for the fuel.

Equation (1) are considered subject to the boundary conditions

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

On the right boundary we have cold ($u = 0$) and unburned state ($v = 1$). The nondimensionalized ambient temperature is taken to be equal to zero. On the left boundary ($x \rightarrow -\infty$) 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 for $x \rightarrow -\infty$.

We seek the solution to (1–2) in the form of a travelling wave solution, which propagates without changing its speed and form and in the co-moving coordinate frame, $\xi = x - ct$, satisfy

$$\begin{aligned} u_{\xi\xi} + cu_{\xi} + v(e^{-1/u} - qre^{-f/u}) &= 0, \\ L^{-1}v_{\xi\xi} + cv_{\xi} - \beta v(e^{-1/u} + re^{-f/u}) &= 0, \end{aligned} \quad (3)$$

where c is the flame speed.

3 Asymptotic analysis

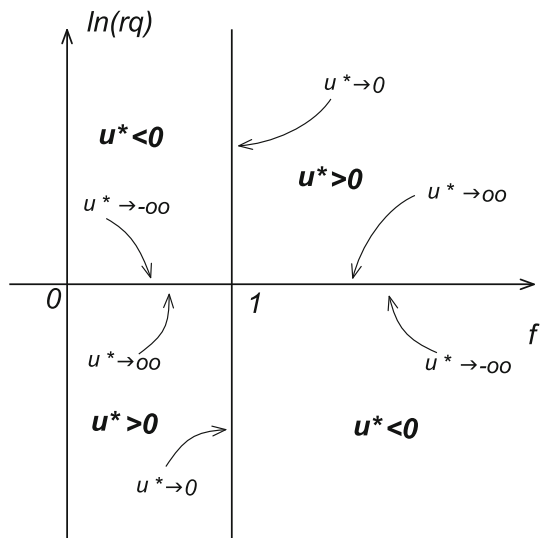
We define crossover temperature as $u = u^*$, such that the rate of heat release from the exothermic reaction is equal to the rate of heat consumption by the endothermic reaction, i.e. $u^* = (f - 1)/\ln(rq)$. Characteristic values of the crossover temperature are shown in Fig. 1 depending on the parameter values. There are two regions with $u^* > 0$: (i) $f > 1, rq > 1$, (ii) $f < 1, rq < 1$; and two regions with $u^* < 0$: (iii) $f < 1, rq > 1$ and (iv) $f > 1, rq < 1$. For regions of parameters where u^* is negative, for any physically feasible flame temperature the rates of heat release and consumption cannot be equal i.e. one always dominates the other. For the case $f > 1, rq < 1$ the exothermic reaction always releases much more heat than can be consumed by the endothermic reaction. Therefore the solution can exist in this parameter region. On the other hand, if $f < 1, rq > 1$ the heat balance is in favour of heat consumption and the rate of heat reduction by the endothermic reaction is always greater. Therefore this implies flame extinction. In this paper we are mainly focused on the analysis of flame properties in the region (i) where $f > 1, rq > 1$.

For parameter values with $u^* > 0$, there is finite value of the crossover temperature, unless $qr \rightarrow 1$ and $u^* \rightarrow \infty$ as shown in Fig. 1 with arrows. The limit $f \rightarrow 1$ is also distinguished, since u^* tends to zero in this case and changes sign as we cross the line $f = 1$. In fact, for $f = 1$ the problem (3) becomes a single-step model

$$\begin{aligned} u_{\xi\xi\xi} + cu_{\xi} + v(1 - rq)e^{-1/u} &= 0, \\ L^{-1}v_{\xi\xi\xi} + cv_{\xi} - \beta v(1 + r)e^{-1/u} &= 0. \end{aligned} \tag{4}$$

Obviously, if $rq > 1$ then heat release is negative in (4) and no solutions can exist. So, this supports the assumption that $f = 1$ and $rq > 1$ is the boundary of the region, $f < 1$ and $rq > 1$, where the solution does not exist. If $rq < 1$, then changing the variables to $z = \xi\sqrt{1 - rq}$, yields

Fig. 1 Plane of parameters $\ln(rq)$ versus f



$$\begin{aligned} u_{zz} + \tilde{c}u_z + ve^{-1/u} &= 0, \\ L^{-1}v_{zz} + \tilde{c}v_z - \tilde{\beta}ve^{-f/u} &= 0, \end{aligned} \quad (5)$$

where $\tilde{\beta} = \beta(1+r)/(1-rq)$ and $\tilde{c} = c/\sqrt{1-rq}$. In the limit of large $\tilde{\beta}$ this gives a well known result for the flame speed, $\tilde{c} = \sqrt{2L\tilde{\beta}^{-1}e^{-\tilde{\beta}/2}}$, and flame temperature, $u_b = \tilde{\beta}^{-1}$. Note that as $rq \rightarrow 1$, the value of $\tilde{\beta}$ becomes unlimited and these expressions for speed and temperature should be accurate. In the original variables we have

$$c = \sqrt{\frac{2L(1-rq)^2}{(1+r)\beta}} \exp\left[-\frac{\beta(1+r)}{2(1-rq)}\right], \quad u_b = \frac{1-rq}{\beta(1+r)}. \quad (6)$$

It is seen that according to (6) as rq tends to unity the flame speed approaches zero in a very abrupt manner, also the flame temperature vanishes. This is another argument towards extinction at $f = 1$ and $rq > 1$. It is noted that $rq = f = 1$ is a unique situation where the governing equations degenerate to

$$\begin{aligned} u_{\xi\xi} + cu_{\xi} &= 0, \\ L^{-1}v_{\xi\xi} + cv_{\xi} - \beta(1+r)ve^{-1/u} &= 0, \end{aligned} \quad (7)$$

which again has only the trivial solution $u = 0$ and $v = const.$

3.1 Endothermic reaction dominated regime

In the case $f - 1 \rightarrow 0$ and $rq > 1$ the crossover temperature vanishes while being positive and it can be treated as a small parameter for the asymptotic analysis i.e. $u^* \equiv \epsilon$, where ϵ is an asymptotically small number. We introduce a new coordinate $\eta = c\xi$ and rewrite (3) as

$$\begin{aligned} u_{\eta\eta} + u_{\eta} + vc^{-2}(e^{-1/u} - qre^{-f/u}) &= 0, \\ L^{-1}v_{\eta\eta} + v_{\eta} - vc^{-2}\beta(e^{-1/u} + re^{-f/u}) &= 0, \end{aligned} \quad (8)$$

which is solved subject to the following boundary conditions $u \rightarrow \epsilon(1 - u_0)$, $v \rightarrow 0$ for $\eta \rightarrow -\infty$ and $u \rightarrow 0$, $v \rightarrow 1$ for $\eta \rightarrow +\infty$. The boundary condition for u in the burned region implies that it is smaller than the crossover temperature. This is physically feasible since this condition implies that the heat released in the course of the exothermic step exceeds the heat consumption in the endothermic step and this excess of enthalpy is used to preheat the cold fresh mixture in front of the reaction region.

As it is usually done in combustion theory, the governing equations are considered separately in two regions: the inner zone where the reaction terms are maximal, and the outer regions, where the reaction is frozen and the transport processes are dominating. The outer regions are located in front and behind the reaction zone, which has an asymptotically small thickness and for definiteness can be considered to be

located at $\eta = 0$. The choice of the location of the reaction zone is arbitrary due to the translational symmetry of the problem (3). The outer problem can therefore be written as

$$\begin{aligned} u_{\eta\eta} + u_\eta &= 0, \\ L^{-1}v_{\eta\eta} + v_\eta &= 0. \end{aligned} \tag{9}$$

Taking into account the boundary conditions, the solution for $\eta < 0$ can be written as $u^- = \epsilon(1 - u_0)$, $v = 0$, and for $\eta > 0$ as $u^+ = u^+(0) \exp(\eta)$, $v^+ = 1 - v(0)^+ \exp(-L\eta)$. The values $u^+(0)$ and $v^+(0)$ should be obtained from matching with the inner region solution.

We introduce the an inner coordinate as $z = \eta/\epsilon$ and seek the solution to the inner problem in a form of the series $u(z) = \epsilon(1 - u_0 - \epsilon s + \dots)$, $v(z) = \epsilon p + \dots$, which after substitution into (3), yields in the leading order

$$\begin{aligned} s_{zz} - Qpe^{-s} (s_1 + s) &= 0, \\ L^{-1}p_{zz} - \beta Qpe^{-s} (1 + q^{-1}) &= 0, \end{aligned} \tag{10}$$

where the reaction terms are expanded into a series in ϵ . The definition of the crossover temperature is recalled, and the following notations have been introduced: $s_1 = u_0/\epsilon$ and $Q = \epsilon^{-2}c^2e^{-\epsilon(1-u_0)}$. It is convenient to define $p = L\beta(1 + q^{-1})\epsilon(f - 1)^{-1}y$ and rewrite (10) as

$$\begin{aligned} s_{zz} - \Omega ye^{-s} (s_1 + s) &= 0, \\ y_{zz} - \Omega ye^{-s} &= 0, \end{aligned} \tag{11}$$

where $\Omega = L\beta Q(1 + q^{-1})$.

Comparing the solutions in the outer and inner regions we find that $u^+(0) = \epsilon(1 + u_0)$, $v^+(0) = 1$, and s and y have to satisfy the following boundary conditions

$$\begin{aligned} s = 0, \quad y = 0 & \text{ for } z \rightarrow -\infty, \\ s_z = 1 - u_0, \quad y_z = \beta \ln(rq) (1 + q^{-1}) & \text{ for } z \rightarrow \infty. \end{aligned} \tag{12}$$

Equations (11–12) constitute a two-point boundary value problem with Ω and s_1 being the eigenvalues. We solve it numerically by using the shooting and relaxation methods. This approach gives us the numerical values for Ω and s_1 , which are both of the order of $O(1)$.

For other parameters fixed and being $O(1)$, the following expression for the flame speed and temperature u_f in the product zone can be expressed as

$$u_f \rightarrow u^*, \quad c = \sqrt{\frac{\beta L(1 + q^{-1})}{\Omega}} u^* \exp\left(-\frac{1 + s_1 u^*}{2u^*}\right), \tag{13}$$

where Ω and s_1 are constants of the order of $O(1)$ and do not depend of f .

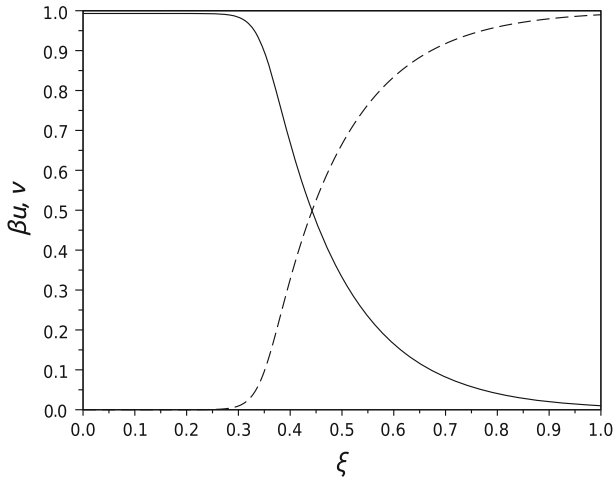


Fig. 2 Combustion wave solution profiles, $u(\xi)$ and $v(\xi)$, for $L = 1$, $\beta = 10$, $f = 2$, $q = 2$ and $r = 5$

3.2 Exothermic reaction dominated regime

For the case $f \gg 1$ the second endothermic reaction is deactivated and the one-step exothermic reaction model can be considered. In the case of asymptotically large β , this gives the well know formulas for the flame speed and burned temperature

$$u_f \rightarrow u_b = \beta^{-1}, \quad c = \sqrt{\frac{2L}{\beta}} \exp\left(-\frac{\beta}{2}\right). \quad (14)$$

Equation (14) are valid regardless of the values of other parameters r and q as long as they are finite.

4 Travelling wave solutions

The two-point boundary value problem (2–3) was solved numerically by using the standard shooting and relaxation methods described elsewhere (see [5] and references therein). In Fig. 2 a typical travelling wave solution profile is plotted in a co-moving coordinate frame. The dependence of u on ξ is shown with the solid line and the concentration of fuel, $v(\xi)$, with the dashed line. The interval of integration is scaled so that ξ changes in the range from 0 to 1 in Fig. 2. In the original variables the length of the interval of integration is approximately 1300.

In Fig. 3, the maximum flame temperature is plotted as a function of f for $L = 1$, $q = 5$, $r = 2$ and two values of β as shown in figure caption. It is clearly seen in figure that there are two distinct flame regimes. For $f > 2$ the endothermic reaction is almost completely frozen, the flame temperature is equal to the adiabatic flame temperature in the one-step model (14), which is shown with the dashed lines and is equal to the inverse of the activation energy of the exothermic reaction. We can call this an exothermic reaction dominated regime. As f is decreased there is an intermediate

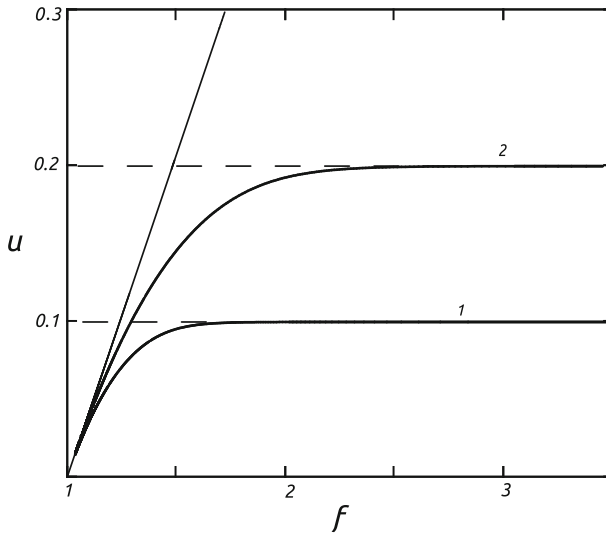


Fig. 3 Dependence of the flame temperature, u_f , on f for $\beta = 5$ (curve 1) and $\beta = 10$ (curve 2) while other parameters are $L = 1, q = 5, r = 2$. The thick solid lines correspond to results of the numerical integration. The dashed lines represent the asymptotic results (14) for the one-step model. The thin solid curve is plotted according to (13)

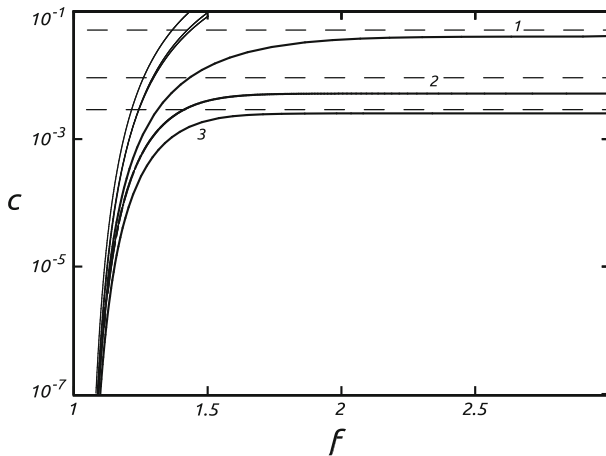


Fig. 4 Dependence of the logarithm of the speed of combustion wave, c , on f for $q = 5, r = 2$ and $L = 1, \beta = 5$ (curve 1), $L = 10, \beta = 10$ (curve 2) and $L = 1, \beta = 10$ (curve 3). The thick solid lines correspond to results of the numerical integration. The dashed lines represent the asymptotic results (14) for the one-step model. The thin solid curves are plotted according to (13)

flame regime around $f = 1.5$, where the endothermic reaction becomes important and the temperature diverges from u_b . As f tends to unity the endothermic reaction dominated regime emerges. In this case the flame temperature converges to u^* and vanishes as f approaches 1 according to Eq. (13).

In Fig. 4, the dependence of the flame speed on f is plotted. The other parameter values are given in the figure caption. It is seen to exhibit the same type of behaviour as

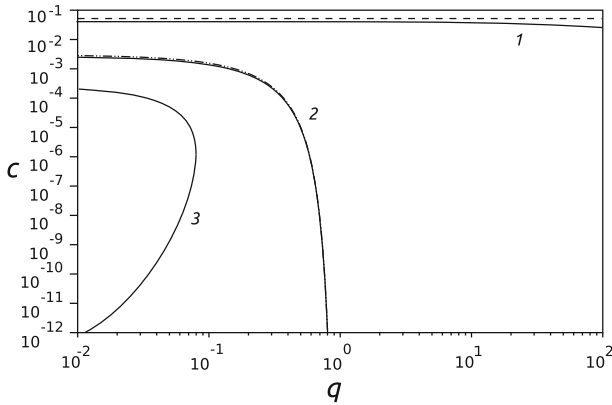


Fig. 5 Dependence of the speed of combustion wave, c , on q in a log–log scale for $L = r = 1$, $\beta = 5$ and $f = 2$ (curve 1), $f = 1$ (curve 2), and $f = 0.96$ (curve 3). The solid lines correspond to results of the numerical integration. The dashed lines represent the asymptotic results (14) for the one-step model. The dash-dotted line are plotted according to (6)

the flame temperature. For large f the speed is constant and is close to the asymptotic flame speed for the one-step model (14) shown with the dashed lines. The discrepancy between the numerical and asymptotic results is larger for $\beta = 5$ (curve 1) and reduces for $\beta = 10$ (curve 3). This is expected since the asymptotic results (14) are valid for $\beta \gg 1$. Changing L from 1 (curve 3) to 10 (curve 2) also affects the flame speed as it is qualitatively predicted by formulas (14). The quantitative difference between numerics and analytics is significant here, which is also expected, since in the derivation of (14) it was assumed that $L \sim O(1)$.

To summarize, we see that two flame regimes of exo- and endothermic reaction domination are clearly demonstrated both numerically and asymptotically. The two approaches are shown to agree well. It is also seen in Figs. 3 and 4 that as f approaches 1 for finite $rq > 1$, flame extinction occurs. The extinction is characterized by the linear decrease of the flame temperature as a function of $f - 1$ and occurs very sharply for the flame speed.

The above analysis corresponds to the region (i) of the parameter plane, where both rq and f are greater than one (see Fig. 1). Next we investigate the properties of the combustion wave solutions in other regions of parametric plane 1. The results of these studies are represented in Fig. 5, where the dependence of the flame speed is plotted versus q in a log–log scale. Parameter r is taken to be equal to one, therefore the sign of $\ln(rq)$ is solely determined by the value of q . The values of L and β are fixed to 1 and 5 respectively. The properties of the travelling waves are studied along several lines $f = \text{const}$ in the plane $\ln(rq)$ versus f , so as to cover all characteristic regions of the plane in Fig. 1. The curve marked with the label ‘1’ corresponds to $f = 2$. This is a regime where the exothermic reaction dominates the endothermic reaction which is not activated. As a result, the flame speed is almost independent of the ratio of heats of the reactions, q , over four orders of magnitude variation of q . The dashed line also shows the asymptotic value of c for large activation energy calculated according to (14). There is significant discrepancy between the numerical and asymptotic results,

which is expected since the value of the activation energy, $\beta = 5$, is not very large here. On the right boundary of the graph the numerically obtained flame speed starts to decrease and the dashed and solid lines begin to diverge. In this case q is becoming large and the crossover temperature, u^* , becomes comparable to u_b from (14). Therefore, the assumption of deactivated endothermic reaction breaks for $q \gg 1$. As f is decreased from 2 to 1 the $c(q)$ dependence bends for smaller values of q since the crossover temperature decreases as f tends to 1 and the endothermic reaction becomes activated for smaller values of q . It should be noted that for $f > 1$ the travelling wave solution exists for all values of q considered here. However, for $f \rightarrow 1$ the limiting behaviour is approached which is governed by (6). The numerical results for the case $f = 1$ are shown with the solid line marked '2'. The dash-dotted line also represents the asymptotic data plotted according to formula (6), which agrees very well with the numerical results. The case $f = 1$ is distinguished from the case $f > 1$ since as rq tends to 1 the flame speed, c , tends to 0 as is clearly seen in Fig. 5 and Eq. (6). For values of q greater than 1 the travelling combustion waves cease to exist. Curve 2 in Fig. 5 marks the switching of the monotonic dependence of c on q to c-shaped type of $c(q)$ behaviour. This is illustrated in Fig. 5 with the curve '3' plotted for $f = 0.96$, where the turning point type of flame extinction is clearly observed: for q less than the turning point value, q^* , there are two combustion wave solutions travelling with different speed, whereas for $q > q^*$ the travelling wave solutions cease to exist. The turning point value q^* strongly depends on f and rapidly decreases as f is becoming smaller than one.

5 Conclusions

In this paper we have investigated the properties of the travelling combustion wave solutions in a diffusion-thermal model with two-step competitive exo–endothermic reaction mechanism in one spatial dimension under adiabatic conditions. The model is investigated both numerically by solving the two-point boundary value problem and analytically using asymptotic analysis. The results of these approaches agree well, thus supporting their validity.

It is found that the flame speed as a function of parameters is either a single-valued monotonic function or a double-valued c-shaped function with the turning point type of behaviour. The switching between these two types of behaviour occurs at $f = 1$ and $rq < 1$. For $f < 1$ the speed of combustion wave as a function of q exhibits a turning point at a certain value of $q = q^*$ such that $rq^* < 1$. For $q < q^*$ there are two combustion wave solution branches, fast and slow, travelling with different velocities and the solutions cease to exist for $q > q^*$. In the case $f > 1$, the flame speed is a single-valued monotonic function with respect to the parameters. It is found that for values of f sufficiently larger than one, the endothermic reaction is not activated and the combustion wave solution behaves similarly to the one-step exothermic reaction model. We call this regime of flame propagation the exothermic reaction dominated regime. In the case when $rq > 1$ as f tends to one, the endothermic reaction is activated and begins to dominate the exothermic reaction. This is accompanied by a decrease of the flame temperature, which

follows the crossover temperature, u^* , and reduces the combustion wave velocity. As a result, at $f = 1$, the flame speed vanishes and the extinction of combustion wave occurs.

To summarize, the model shows rich dynamical behaviour depending on the choice of the parameter values. Interesting instability characteristics are also expected to occur under different combustion regimes as discussed in this paper. We have succeeded with the stability analysis in the exothermic reaction dominated regime [13] and the aim of our future investigation is to carry out a systematic stability analysis of combustion waves in the current model.

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