

# Combustion waves in a model with chain branching reaction

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In this paper the steady planar travelling waves in the adiabatic model with two-step chain branching reaction mechanism are investigated numerically. The properties of these solutions are demonstrated to have similarities with the properties of non-adiabatic combustion waves that is, there is a residual amount of fuel left behind the travelling waves and the solutions can exhibit extinction. It is also shown that the model possesses a new type multiple travelling wave solutions (which we call wave trains) with complex structure of the profiles and varying speeds.

**KEY WORDS:** combustion waves, chain branching, wave trains

**AMS subject classification:** 35K57, 80A25

## 1. Introduction

Combustion waves have been studied for some time and are topic of a review [1]. They have been observed in numerous experiments [1] and play an important role in industrial processes, such as one of the current technologies for creating advanced materials: Self-propagating High-temperature Synthesis (SHS) [2].

To date only the simplest models of these phenomena, which use one-step chemistry, have been comprehensively analysed. In these models it is assumed

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that the reaction is well modelled 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:  $F \rightarrow P + \text{heat}$  or  $F + O_2 \rightarrow 2P + \text{heat}$ , where the temperature dependent rate of the reaction is given by Arrhenius kinetics  $k(T) = e^{-T_a/T}$  and  $T_a$  is the activation temperature.

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 [3,4]. Finally, 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 etc.

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 [3]. Moreover for many reactions, models with simple one-step kinetics may lead to erroneous conclusions as noted in [5]. In other words, if we want to obtain a realistic description of the flame kinetics several different steps, each with its own intermediate chemical species, have to be taken into account. As a result the analysis of models involving several reactions is far more difficult than that required for the one-step models which can often be comprehensively analysed using established methods.

Several models involving two chemical steps have been considered previously [3] and more recently [6,7]. These include several classes of models with two-step kinetics such as models with chain branching [3], fuel decomposition [3, 6], inhibition of flames by an endothermic reaction [7] etc. However these models have received very little attention from an analytical prospective since the well established methods (like asymptotic methods) allow the investigation only in very special cases when these models can be substantially simplified. Nevertheless the numerical analysis carried out in [7] (where the inhibition of flames by an endothermic reaction is studied) showed significant differences between the properties of the solutions to the models with one- and two-step chemistry such as, for example, existence of multiple (three) solutions for certain parameter regions. Since the behaviour of multi-step models can differ from their one-step prototypes we can expect a number of new phenomena such as bistability to be found as a result of such analysis.

The aim of the current paper is to investigate the properties of flames with complex kinetics which are described by the model with two-step chain branching reaction [3]. This model has practical applications in simulating the hydrogen oxidation [3,8] and is known to exhibit travelling front solutions [3]. However, to the best of our knowledge, the properties of the travelling front solutions have not been studied in detail. Moreover until now the existence of other types of travelling wave solutions distinct from the travelling fronts which are known to exist in the models with single step reaction was unclear. Here we show that

increasing complexity of the model to include the two-step reaction mechanism can result in a drastic change of the properties of the system: namely coexistence of multiple travelling wave solutions for some parameter values. In contrast to [7], where coexistence of pulse and front solutions have been observed, these new solutions are not pulses, but wave trains with several humps. Similar solutions were also observed in different reaction-diffusion systems [9]. However to the best of our knowledge they have never appeared in the combustion literature.

The rest of the paper is organized as follows. In the next section we introduce the governing partial differential equations for the model under investigation in dimensional and non-dimensional form. In Section 3 these equations are reduced to the system of ordinary differential equations for the travelling wave profiles. Section 4 is devoted to the investigation of the properties of the travelling front solution, which is common for both one- and two-step models. The travelling wave trains, which are specific for the model studied in this paper, are investigated in Section 5. Finally, in Section 6 the conclusions and final discussions are presented.

## 2. Model

We consider an adiabatic model (in one spatial dimension) that includes two steps: autocatalytic chain branching  $A+B \rightarrow 2B$  and recombination  $B+M \rightarrow C+M$ . Following the work of [3] and [6] we assume 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. As noted in [3], in this scheme recombination stage serves both as an inhibitor which terminates the chain branching and an accelerant which produces heat. According to [6], equations governing this process can be written as

$$\begin{aligned} \rho c_p \frac{\partial T}{\partial t} &= k \frac{\partial^2 T}{\partial x^2} + \rho Q A_r Y_B, \\ \frac{\partial Y_A}{\partial t} &= D_A \frac{\partial^2 Y_A}{\partial x^2} - A_B Y_A Y_B e^{-E/RT}, \\ \frac{\partial Y_B}{\partial t} &= D_B \frac{\partial^2 Y_B}{\partial x^2} + A_B Y_A Y_B e^{-E/RT} - A_r Y_B \rho / M, \end{aligned} \quad (1)$$

where  $T$  is the temperature;  $Y_A$  and  $Y_B$  represent the concentrations of fuel and radicals respectively;  $\rho$  is the density;  $c_p$  is the specific heat;  $M$  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;  $R$  is the universal gas constant.

We introduce the non-dimensional variables

$$t' = \frac{\rho Q_{AB} R}{c_p M E} t, \quad x' = \sqrt{\frac{\rho^2 Q_{AB} R}{k M E}} x, \quad u = \frac{RT}{E}, \quad v = \frac{Y_A M}{\rho}, \quad w = \frac{Y_B M}{\rho}, \quad (2)$$

and we rewrite (1) as

$$\begin{aligned} u_t &= u_{xx} + r w, \\ v_t &= \tau_A v_{xx} - \beta v w e^{-1/u}, \\ w_t &= \tau_B w_{xx} + \beta v w e^{-1/u} - r \beta w, \end{aligned} \quad (3)$$

where primes have been dropped and the following non-dimensional parameters have been introduced:  $\beta = c_p E / Q R$ ,  $r = A_r / A_B$ ,  $\tau_{A,B} = \rho c_p D_{A,B} / k$ . Equations (3) are considered subject to the boundary conditions

$$\begin{aligned} u &= 0, \quad v = 1, \quad w = 0 && \text{for } x \rightarrow \infty, \\ u_x &= 0, \quad v_x = 0, \quad w = 0 && \text{for } x \rightarrow -\infty. \end{aligned} \quad (4)$$

On the right boundary we have cold ( $u=0$ ) and unburned state ( $v = 1$ ), where fuel has not been consumed yet and no radicals have been produced ( $w = 0$ ). We also take the ambient temperature to be equal to zero. As noted in [10] this is a convenient way to circumvent the so-called ‘‘cold-boundary problem’’ and it does not change the generic behaviour of the system. On the left boundary ( $x \rightarrow -\infty$ ) neither the temperature of the mixture nor the concentrations of fuel can be specified. We require that there is no reaction happening so the solution reaches a stationary point of (3). Therefore the derivatives of  $u$ ,  $v$  are zeros and  $w = 0$  for  $x \rightarrow -\infty$ .

### 3. Travelling wave solutions

We seek a solution to the problem (3), (4) 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 have introduced  $\xi = x - ct$ , a coordinate in the moving frame and  $c$ , the speed of the travelling wave. Substituting the travelling wave solution into the governing equations we obtain

$$\begin{aligned} u_{\xi\xi} + c u_{\xi} + r w &= 0, \\ \tau_A v_{\xi\xi} + c v_{\xi} - \beta v w e^{-1/u} &= 0, \\ \tau_B w_{\xi\xi} + c w_{\xi} + \beta v w e^{-1/u} - r \beta w &= 0. \end{aligned} \quad (5)$$

and boundary conditions

$$\begin{aligned} u &= 0, \quad v = 1, \quad w = 0 && \text{for } \xi \rightarrow \infty, \\ u_{\xi} &= 0, \quad v_{\xi} = 0, \quad w = 0 && \text{for } \xi \rightarrow -\infty. \end{aligned} \quad (6)$$

Following [3,6,7] we consider the case when Lewis numbers for the fuel and the radicals are equal to unity. Although this assumption simplifies the problem significantly, it still allows the investigation of the generic properties of the system (5) and (6).

In the case  $\tau_A = \tau_B = 1$  equations (5) possess an integral  $S = \beta u + v + w$ . Using  $S$  and first boundary condition in (6) equations (5) can be reduced to a system of two second order equations for temperature and fuel concentration

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

On the right boundary we require that  $u = 0$  and  $v = 1$ , whereas on the left boundary ( $\xi \rightarrow -\infty$ ) we modify the boundary conditions as follows

$$u = \beta^{-1}(1 - \sigma), \quad v = \sigma, \quad (8)$$

where  $\sigma$  denotes the residual amount of fuel left behind the wave and is yet unknown. We note here that at first glance system (7) looks very similar to equations describing the dynamics of the one-step adiabatic reaction model [10]. However, in contrast to the one-step case, equations (7) do not have integral, which allows further simplification [10]. Moreover boundary conditions (8) suggest that there can be some fuel left behind the reaction zone, which is impossible in the case of a one-step adiabatic reaction model.

#### 4. Classical travelling front solution

We solve equations (7) numerically using shooting and relaxation methods as described in [4]. As a first step we solve equations (7) employing the fifth-order Runge–Kutta method on the interval  $\xi \in [-L_1, L_2]$ , where  $L_{1,2} > 0$  are taken to be sufficiently large. Our numerical integrator allows the estimation of the local relative error, which has been set to be less than  $10^{-5}$  in our calculations.

As a second step the solution obtained with the shooting method is used as the guess solution for a more accurate method; namely relaxation (see [4] and references within for the detailed description of the method). The relaxation routine allows the control the average local correction made on each iteration step. The solution is considered to be resolved if the correction is less than  $10^{-15}$ .

Combination of the shooting and relaxation methods allows us to obtain the solution to (7) with high degree of accuracy. This was tested by changing the step of the grid and comparing the resulting variations in the values of the internal parameters of the problem such as the speed. For example, a fourfold mesh refinement changes the value of  $c$  only in the ninth significant digit.

The numerical methods described above allow us to obtain the dependence of the speed of the travelling front on the parameters of the problem  $r$  and

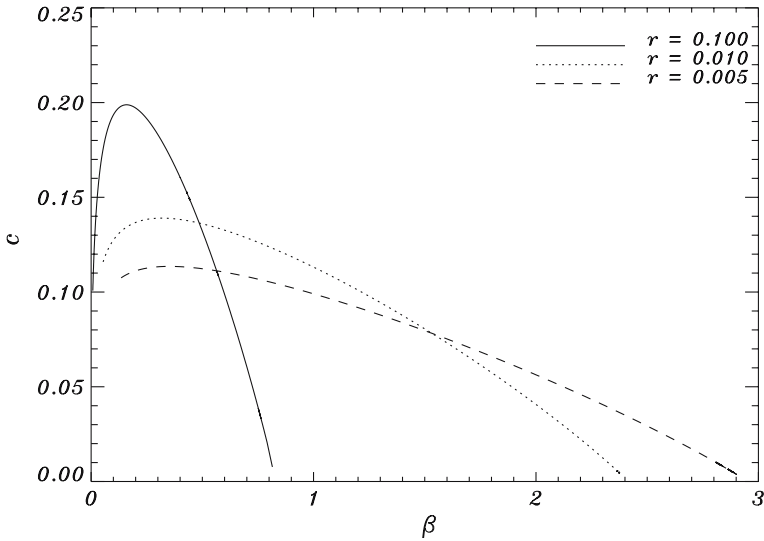


Figure 1. Speed of the front as a function of  $\beta$  for various values of  $r$ .

$\beta$ . In figure 1 we plot  $c$  as function of  $\beta$  for  $r = 0.1, 0.01, \text{ and } 0.005$ . At first glance the dependence of  $c$  on  $\beta$  resembles the behaviour of the speed of the front for the model with one-step reaction scheme, which was studied in [4]. Namely,  $c$  reaches maximum for some value of  $\beta$  and decays monotonically as we increase (or decrease)  $\beta$  from the value corresponding to the maximum. However more detailed investigation shows that there is a substantial difference between the prediction of one- and two-step models. In particular for the model with one-step reaction mechanism the travelling front solution exists for any value of  $\beta$  and decays exponentially to zero as we increase  $\beta$ . This is not the case for the model with two-step reaction mechanism. In figure 2 we plot the logarithm of the speed as a function of parameter  $\beta$  for  $r = 0.001$ . It is clearly seen that the speed of the front does not follow the exponential law for large values of  $\beta$  as is the case for the one-step model. Moreover  $c(\beta)$  exhibits a critical type of behaviour: as we increase  $\beta$  up to some critical value  $\beta_c = 4.2\dots$  the speed of the front decays rapidly and it appears that the travelling front solution ceases to exist for  $\beta > \beta_c$ . On the other hand the difference between the one-step and the two-step models is strengthened by the fact that there is residual amount of fuel left behind the front in the case of the two-step model, which is identically equal to zero in the case of the one-step adiabatic model. In figure 3 we plot  $\sigma$  as a function of  $\beta$  for  $r = 0.01, 0.005, \text{ and } 0.001$ . The residual amount of fuel increases monotonically with  $\beta$ . It is interesting to note that  $\sigma$  becomes significant as we approach the critical value  $\beta_c$  for fixed  $r$ .

To some extent the properties of the two-step adiabatic model, which is studied here, resemble the properties of the non-adiabatic one-step model,

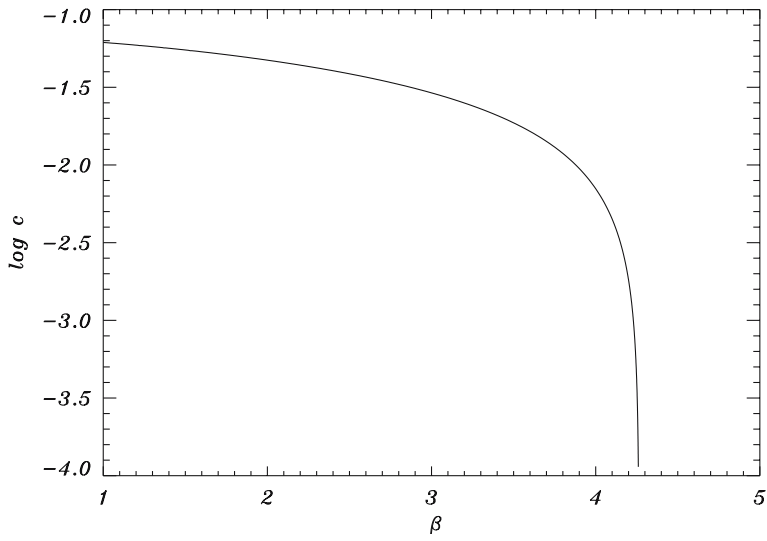


Figure 2. Logarithm of speed of the front as a function of  $\beta$  for  $r = 0.001$ .

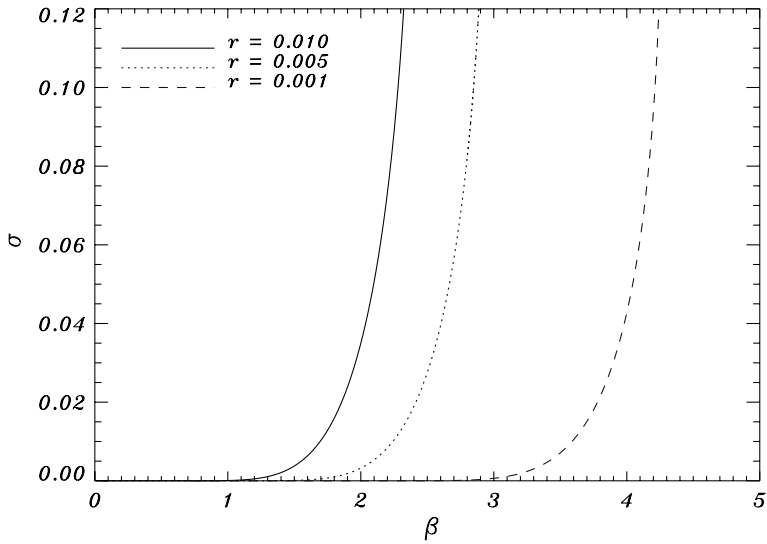


Figure 3. Dependence of  $\sigma$  on  $\beta$  for various values of  $r$ .

investigated in [11]. This is expected since the recombination step works as an inhibitor of the chain branching reaction and to certain extent plays the same role as the heat exchange with the surrounding in the one-step non-adiabatic model. In both cases there is a non-zero residual amount of fuel left behind the reaction zone. The similarity between these two cases is also strengthened by likeness of the behaviour of the speed of the front as a function of the parameter  $\beta$ . Namely, in both one-step non-adiabatic and two-step adiabatic models the travelling front solution cease to exist as we approach some critical value of  $\beta_c$  (in combustion literature this event is usually called extinction [11]). However, the route to extinction in these models appears to be different. In the case of the one-step non-adiabatic 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 meet each other (this event is also known as a turning point or a saddle node 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 as shown in figure 2.

In figure 4 the speed of the front is plotted as a function of parameter  $r$  for  $\beta = 0.5$ . Qualitatively the behaviour of  $c(r)$  is described in [3]. For  $r = 0$  there is no heat release, the temperature does not grow and the speed of the front is negligible. When  $r$  is small the recombination step works as an accelerator of the chain branching and the speed of the front grows. For large values of  $r$  the recombination reaction starts to work as an inhibitor and therefore  $c(r)$  decreases.

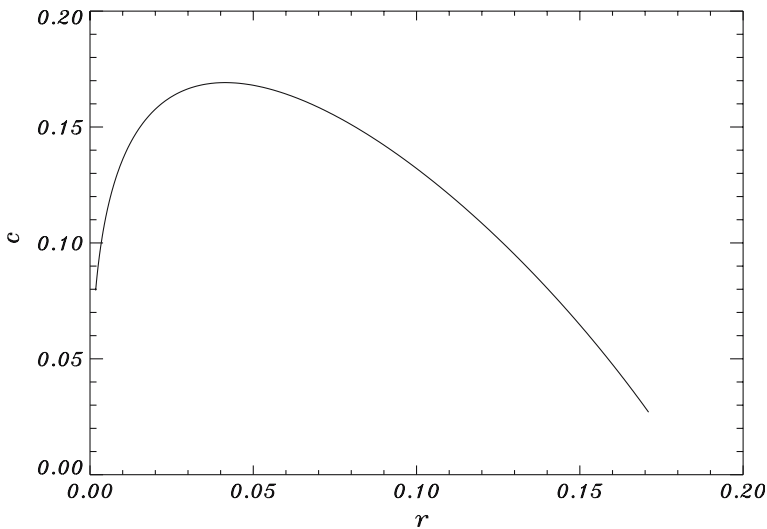


Figure 4. Speed of the front as a function of  $r$  for  $\beta = 0.5$ .



## 5. Travelling wave train solutions

In the previous section we have discussed the differences between the predicted flame behaviour obtained with the one- and two-step models. In both models the travelling combustion fronts have one common property: temperature and fuel concentration are monotonic functions of coordinate  $\xi$ . Perhaps the most significant property of the two-step model studied in this paper which distinguishes it from both adiabatic and non-adiabatic one-step combustion schemes is the coexistence of several (more than two) travelling wave solutions such that  $u(\xi)$  and  $v(\xi)$  are no longer monotonic functions. Travelling wave solutions in a form of a pulse (which are described by non-monotonic functions of coordinate) are investigated in [7] for the model with two-step reaction mechanism (one of the steps is exothermic and the other step is endothermic reaction). However in this paper we demonstrate the existence of travelling wave solutions such that  $u(\xi)$  and  $v(\xi)$  are not monotonic functions and these solutions are not pulses. In what follows we refer to them as wave trains. We can find no evidence that such solution structures have ever been reported in the combustion literature. Schematically a wave train can be imagined as a combination of the front and one, two or more pulse solutions. The whole wave structure travels with a certain speed  $c_i$  which is in general different from the speed of the combustion front. Here we use the index 1 to refer to the wave train solutions consisting of a front and a single pulse, index 2 to denote the wave train solutions consisting of a front and a two pulses and so on. In figure 5 we demonstrate solution profiles with different structures: (a) and (b) correspond to type 1 solutions; (c) and (d) show type 2 solutions. These solutions are plotted for various values of  $\beta$ : figures (a) and (c) for  $\beta=1.28$  and (b) and (d) for  $\beta=2.37$ . The solution profiles shown in figure 5 do not seem to satisfy the right boundary conditions, since we plot solutions on only a part of the integration interval in order to demonstrate the inner structure of the solution profiles. Over the full interval of integration used in our numerical scheme  $u(\xi)$  and  $v(\xi)$  approach the limiting values to within the error tolerance  $10^{-15}$ . These solutions have been computed with different grid spacing, and domain size ( $L_1$  and  $L_2$ ) and are entirely consistent hence we do not believe that they are numerical artifacts.

The structure of wave train solutions consists of several regions. Similar to the travelling front solution there is a preheat zone in front of the combustion wave where the fuel has not been consumed yet and the temperature slowly increases; behind the combustion wave there is a product region where the temperature is reaching a maximum value and fuel is not consumed. As shown in figure 5, in contrast to the travelling front, the region between the preheat and the product zone is constituted of several alternating regions where fuel consumption becomes more or less intensive. This process is accompanied by variation of the temperature along the complex structured inner region. The temperature reaches a local maximum in the regions where fuel is burned more

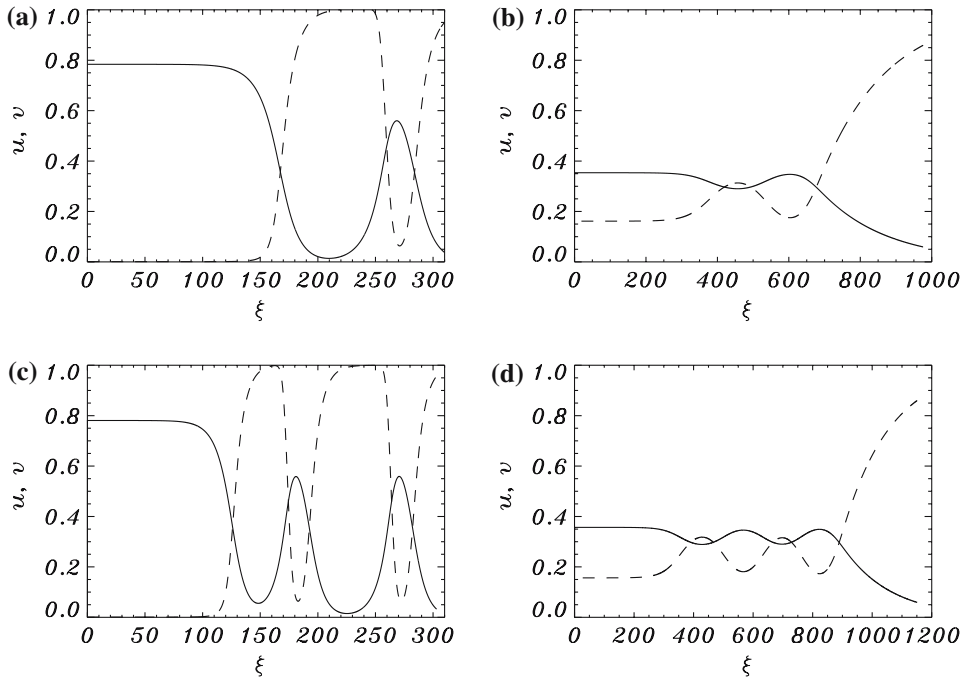


Figure 5. Temperature (solid lines) and fuel (dashed lines) profiles for  $r = 0.01$  and various values of parameter  $\beta$ :  $\beta = 1.28$  in figures (a) and (c);  $\beta = 2.37$  in figures (b) and (d).

effectively so that the fuel concentration drops down approaching a local minimum. The regions where the fuel consumption is more effective (with maximum of the temperature and minimum of the fuel concentration) are intermitted with the zones where the fuel consumption is less effective so that  $u$  and  $v$  are reaching local minimum and maximum respectively.

As we decrease  $\beta$  the local temperature peaks become higher and the fuel concentration troughs become lower as shown in figure 5(a, c). The wave train starts to look like well separated front and one or more pulses travelling ahead of the front solution. In the opposite limit, as we increase  $\beta$  up to the values close to the extinction value  $\beta_c$  the solution profiles flatten and approach the shape of the travelling front. The overall change of temperature and fuel concentration in the region with alternating behaviour gets smaller in comparison with the maximum values of  $u$  and  $v$  as shown in figure 5(b, d).

In figure 6 we plot the difference between the speed  $c_1$  of the travelling wave train of type 1 and the speed of the classical travelling front solution  $c$  as a function of  $\beta$  for  $r = 0.001$ . Figures 4 and 6 indicate that the solution branches  $c_1(\beta)$  and  $c(\beta)$  are located close to each other on the  $c$  versus  $\beta$  plane. For some value  $\beta_1 \approx 3.1 \dots$  the difference  $c - c_1$  equals zero. So in this case there exist two solu-

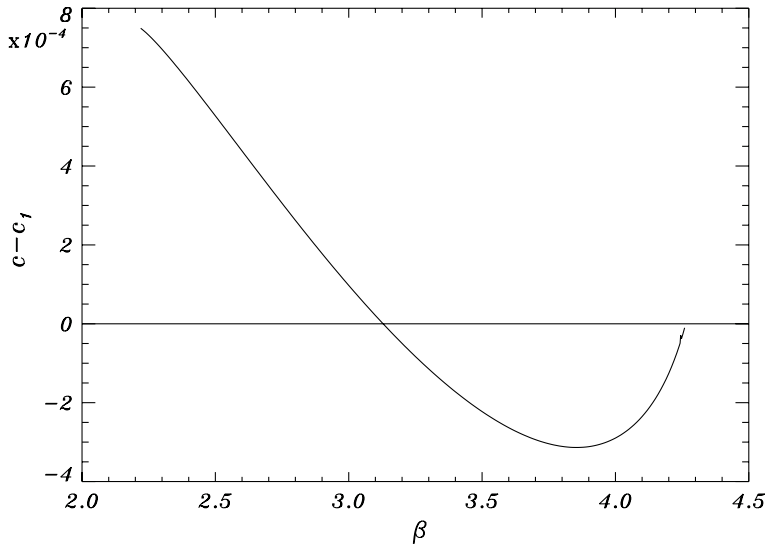


Figure 6. Dependence of  $c - c_1$  on  $\beta$  for  $r = 0.001$  showing the difference between the classical travelling wave solution and the type 1 solution.

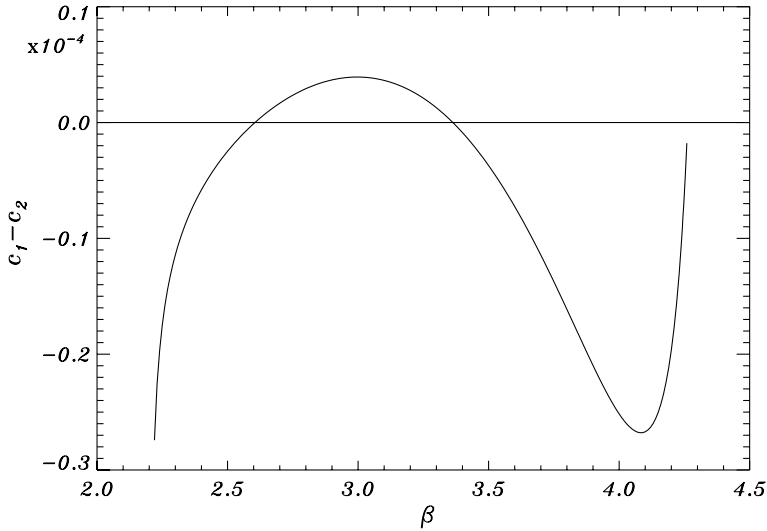


Figure 7. Dependence of  $c_1 - c_2$  on  $\beta$  for  $r = 0.001$ .

tions travelling with the same speed (although the residual amount of fuel left behind the reaction is different  $\sigma_1 \neq \sigma$ ).

In figure 7 we plot variation of  $c_1 - c_2$  with  $\beta$  for  $r = 0.001$ , where  $c_2$  is the speed of the travelling wave train of type 2. Comparing figures 4, 6 and 7 we conclude that the distance between the solution branches  $c_i(\beta)$  and  $c_{i+1}(\beta)$

is getting smaller with increasing index  $i$ , which we use to distinguish the wave train solutions as described previously. Although not shown here the same pattern was observed for the wave trains of type 3 (i.e.  $i = 3$ ) and 4 (i.e.  $i = 4$ ) which we are able to obtain numerically. Functions  $c(\beta)$  and  $c_2(\beta)$  intersect at some value  $\beta_2$  close to  $\beta_1$  for which  $c = c_1$ . In this case there exist two solutions with the same speed ( $c = c_2$ ) and different values of the residual amount of fuel ( $\sigma \neq \sigma_2$ ). It also follows from figure 7 that curves  $c_1(\beta)$  and  $c_2(\beta)$  intersect for two different values of  $\beta_1^1 \approx 2.6\dots$  and  $\beta_2^1 \approx 3.4\dots$ . This implies again that for these values of  $\beta$  there exist two travelling wave trains with the same speed and different values of residual amount of fuel ( $\sigma_1 \neq \sigma_2$ ).

As parameter  $\beta$  is increased up to the extinction value  $\beta_c$  the velocities of the wave trains of various types approach zero. It appears that the wave trains cease to exist at the same value  $\beta_c$  for which there is extinction of the travelling front solution.

## 6. Conclusions

In this paper we study the properties of the steady planar travelling waves in the model with two-step chain branching reaction mechanism, which was introduced in [6]. The main focus of our work is to investigate what new features does this model of flame propagation predict, that cannot be observed by using the one-step reaction scheme.

For the one-step adiabatic model there exists a single travelling wave solution in a form of the combustion front for any parameter values. All fuel is consumed during the reaction and the temperature behind the reaction zone reaches its maximal value. It has been known [3] that the two-step counterpart of this model with chain branching exhibits the analogous travelling front solution as well. Numerical analysis carried out in this paper has shown that the properties of the combustion front for the one- and two-step adiabatic models are different from each other. In the latter case the presence of the intermediate species such as radicals, which are produced during the recombination step, affects the flame propagation in two similar ways to the influence of the heat loss on the combustion waves in the model with one-step reaction mechanism: (i) there is a residual amount of fuel left behind the reaction; (ii) the travelling front solution exists only in a certain parameter region.

The most striking difference between the models with one- and two-step chain branching reaction mechanism is the coexistence of multiple solutions in the latter case. In this paper we have found that besides the conventional travelling combustion front which can be observed in both models, the model with chain branching possesses a set of solutions of more complex structure, which we call travelling wave trains. These new solutions profiles are not pulses and they differ from each other by the number of local temperature maxima reached

in the process of wave propagation. We have studied the properties of the wave trains numerically. It appears that the wave train solutions exist only in some parameter regions. On the boundary of this region the solutions cease to exist due to extinction. The investigation of speed of the wave train solutions has revealed that for certain parameter values there can be two solutions travelling with the same speed (however these solutions have different residual amount of fuel left behind the reaction).

The existence of wave trains raises a number of questions that we hope to address in the future. It is not clear how many solutions can the model possess for a given set of parameter values (we were able to obtain five). It would be extremely interesting to investigate the stability of these solutions and to determine which of them can be physically observed etc.

To summarize, we believe that the current paper clearly demonstrates that by taking into account more complex reaction schemes than just a simple one-step model results in dramatic changes to the properties and behaviour of the propagating flames. We hope that the present work will further boost the interest of the combustion community to the investigation of the models with multiple-step reaction mechanisms such as chain branching, fuel decomposition etc.

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