

Shock-induced ignition with three-step chain-branching kinetics

Josue Melguizo-Gavilanes^{*†} and Luc Bauwens^{*}

^{*}Department of Mechanical and Manufacturing Engineering
University of Calgary 2500 University Dr. NW. Calgary T2N1N4, CANADA

[†]Corresponding address : jmelguiz@ucalgary.ca

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Abstract

Simulation of shock-ignition is a challenging problem because of the singular nature of the initial conditions due to the initial non-existence of the region of shocked reactive mixture. A combination of techniques has been developed, and results are obtained for a three-step chain-branching kinetic scheme. These techniques include replacing space as an independent variable by the ratio space over time, using initial conditions obtained from short time asymptotics. The essentially non-oscillatory numerical scheme used captures the entire ignition process : hot spot and shock formation, and appearance of a detonation wave. Numerical simulations were performed based upon the reactive Euler formulation, for a contact surface separating non-reactive hot fluid from cold reactive mixture. Chain-branching results were obtained for crossover temperatures corresponding to shocked fluid states from close to the limit to well inside the chain-branching zone. Since ignition occurs in a very thin zone compared with the length of the computational domain, the computations are relatively large, even though the problem is one-dimensional. Well inside the chain-branching region, results are consistent with the strong ignition model of Souloukhin and Oppenheim. Conversely, as we approach the limit, it seems that our model fails to capture the weak ignition regime.

Keywords : shock-ignition, detonation, chain-branching

1. Introduction

Ignition behind a shock moving through reactive mixture is likely a key element of the deflagration to detonation transition (DDT) process, for example, subsequent to shock reflections on obstacles. Chemistry being very temperature sensitive, the temperature jump across a shock leads to reaction rates orders of magnitude larger behind the shock. Shock ignition experiments by Voevodsky & Soloukhin¹⁾ and Meyer & Oppenheim²⁾ led to identification of distinct strong and mild ignition regimes, associated with competition between diffusive mechanisms (i.e. appearance of flames) and chain-branching chemistry. One dimensional simulations using complex kinetics for hydrogen-oxygen were performed by Oran & Boris³⁾. Analysis and one-dimensional simulation have been carried out for a shock ahead of a piston impulsively started^{4),5)}, or, as in the current scenario, for a shock moving across an interface such as a flame separating inert or burnt mixture from reactive

mixture⁶⁾⁻⁸⁾. Previous work was limited to single step Arrhenius kinetics, apart from Sharpe⁹⁾ and Blythe et al.¹⁰⁾. The former considered a two step chain-branching model in a piston problem whereas the latter considered the simplest three-step chain-branching mechanism¹¹⁾ at large activation energies. The current study also focuses upon the three-step scheme but for the flame or interface problem. Simulation is difficult because typically the region behind the shock, where ignition will take place, does not exist initially, potentially resulting in numerical artifacts in the region close to the wall or contact surface, which is precisely where a hot spot will appear. To avoid these, one can use grid refinement⁷⁾, or as in Melguizo-Gavilanes et al.¹⁰⁾ a transformation originally proposed by Short & Dold⁶⁾, in which the original problem formulation using space x and time t as the independent variables is converted using the variables $\eta = x/t$ and t , yielding a finite domain at $t = 0$. Initial conditions at a nonzero time can be obtained from short time perturbation, avoiding

difficulties brought about by the singular nature of the initial conditions that still remain in the transformed problem. Short & Dold⁶⁾ used the transformation to study only the early part of the process. In contrast, here a second order ENO algorithm is implemented, suitable for the entire ignition process, including rapid growth of the hot spot, shock formation, appearance of a detonation and shock merger. A validation study has been performed by Melguizo-Gavilanes *et al.*¹⁰⁾ for single step kinetics, yielding results in close agreement with Sharpe & Short⁷⁾. Results for cases from post-shock states close to the limit to well inside the explosion zone are discussed in light of experimental results²⁾.

2. Physical model

The problem is described by the reactive Euler's equations, in an infinite domain. The initial and boundary conditions are those of a Riemann problem in non-reactive fluid, separating a stream of low pressure unburnt fluid at uniform state and velocity coming from infinity on the left ($x < 0$) to steady burnt (or inert) mixture on the right ($x > 0$). The frame of reference is set such that, in the inert side, shocked fluid in the Riemann solution is at rest, and the problem is scaled such that pressure, density, and temperature are unity in this shocked fluid (i.e. the state variables are scaled by their post-shock values). The fluid is taken to be an ideal gas with constant specific heats. As a result the solution only departs from that of the Riemann problem because of the chemistry. Initially, that departure is small. In the current frame of reference, the Riemann (shock tube) solution includes a shock moving away at a constant negative speed, followed by reactive mixture at rest, a contact surface at $x = 0$ separating the shocked reactive fluid from a zone of burnt/inert fluid extending to an expansion wave that moves toward $x \rightarrow +\infty$. In a model including diffusion, the contact surface would become a premixed flame moving into the unburnt mixture; if the effect of diffusion is ignored then the burning velocity is negligible compared with the speed of sound, and the inviscid problem featuring the contact surface provides a consistent approximation. In effect, this formulation represents the problem of a shock that reflected from a wall behind a planar flame (i.e. on the burnt side), and then overtook the flame. The three-step chain-branching kinetic model¹¹⁾ includes initiation and chain-branching described by an Arrhenius rate, and a constant termination rate.

Using index I for initiation and B for branching, and introducing

$$\varepsilon = \exp\left(\frac{E_I}{T_I} - E_I\right), \alpha = \exp\left(\frac{E_B}{T_B} - E_B\right), \theta = E_B - \frac{E_B}{T}$$

chemistry can be written as

$$\frac{\partial \lambda_1}{\partial t} + u \frac{\partial \lambda_1}{\partial x} = -\varepsilon \lambda_1 \exp\left(\frac{E_1 \theta}{E_B}\right) - \alpha \rho \lambda_1 \lambda_2 \exp \theta$$

$$\frac{\partial \lambda_2}{\partial t} + u \frac{\partial \lambda_2}{\partial x} = \varepsilon \lambda_1 \exp\left(\frac{E_1 \theta}{E_B}\right) - \alpha \rho \lambda_1 \lambda_2 \exp \theta - \lambda_2$$

in which λ_1 is the reactant mass fraction, and λ_2 is the chain-branching specie. Time has been scaled such that the dimensionless constant termination rate is unity. Heat release (scaled by the ratio post-shock pressure divided by density) is associated with termination only. Values of ε are in the order of 10^{-6} or smaller. Neglecting the role of small initiation, the chain-branching region on the explosion pV diagram for the post-shock state corresponds to $\alpha > 1$ while for $\alpha < 1$, chain-branching does not take place¹²⁾.

3. Methodology

The current initial value problem does not lend itself to a straightforward numerical solution because (1) initially the region where chemistry takes place, made of reactive shocked fluid, does not exist, so that staircasing effects are possible, and (2) hot spot formation goes from very slow to very rapid. Dealing with these issues accurately and effectively requires a combination of techniques. To deal with the singular nature of the initial conditions, a transformed formulation is used⁶⁾ replacing space x as an independent variable by $\eta = x/t$ in which t is time. However in the transformed formulation, the CFL condition requires time steps approaching zero initially. Closed form results from short time asymptotics are used instead as initial conditions, at some small nonzero time, the derivation and resulting initial conditions are not shown here due to length limitations. Yet, see Melguizo-Gavilanes *et al.*¹⁰⁾ for a detailed derivation and validation for single step kinetics even if the resulting expressions are not the same.

4. Numerical simulation

To solve the problem at hand, a second order accurate ENO scheme was implemented. The well-validated code originated in Xu *et al.*¹³⁾; it has since been significantly modified and parallelized using MPI (Message Passing Interface). It has been used successfully in studies of multidimensional detonation waves¹⁴⁾, of realistic hydrogen accident scenarios¹⁵⁾, and of accelerating flames¹⁶⁾. The numerical domain goes from a negative value of η slightly smaller than η_s (the initial speed of the leading shock) to a positive value somewhat larger than the speed of sound behind the contact surface. This guarantees that the leading shock will never reach the left boundary. Likewise, the right boundary is placed at a value of η greater than the speed of sound behind the contact surface so that acoustic waves moving right will never reach this boundary and no reflection occurs.

5. Results

The scheme above was used for a series of cases, all for a shock moving away at a Mach number of 0.7 from the contact surface, a dimensionless heat release of 4 and a ratio of specific heats, $\gamma = 1.4$. A range of values of α was simulated, from far outside of the chain-branching region to far inside. Specific values were $\alpha = 0.5, 0.7, 0.85, 0.99, 1.0, 1.01, 1.2, 1.9, 2.16$. These were obtained varying the chain-branching cross-over temperature T_B . The other

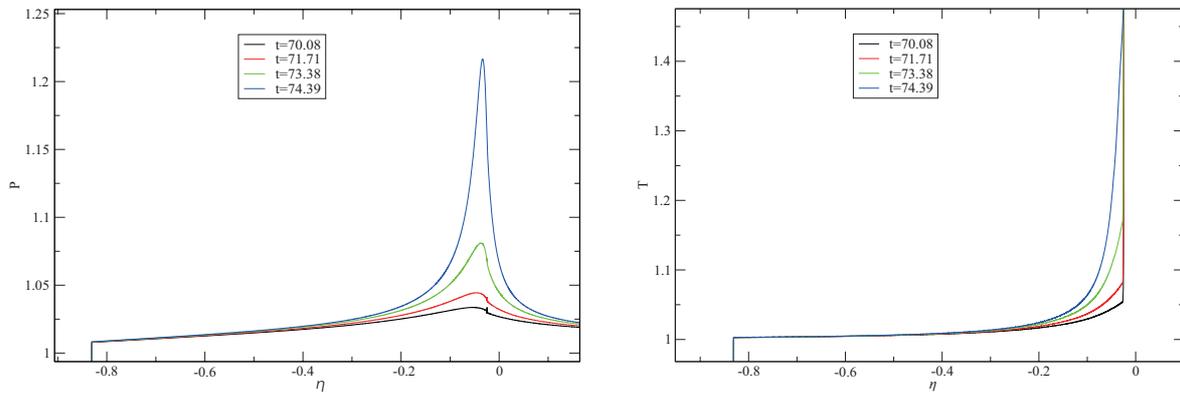


Fig. 1 $\alpha = 1.01$: Pressure and temperature during hot spot formation

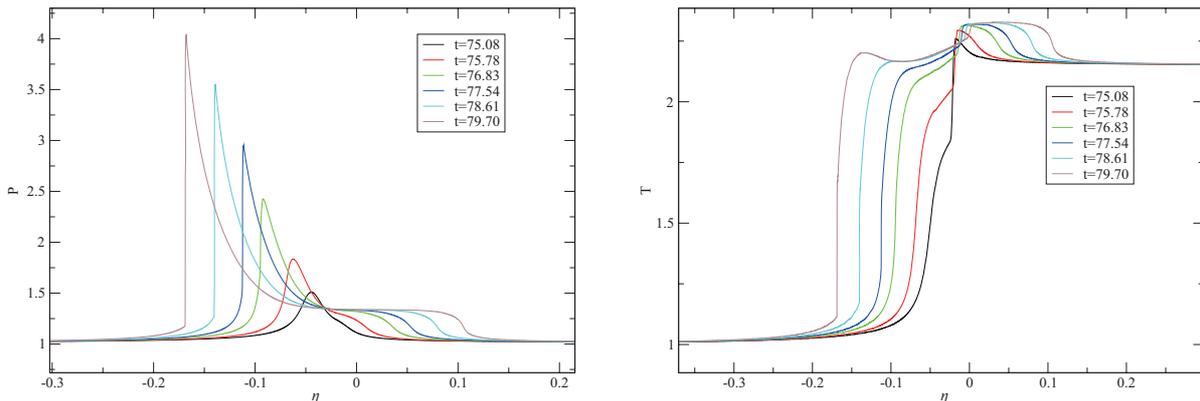


Fig. 2 $\alpha = 1.01$: Pressure and temperature during shock formation phase

parameters were kept constant, with $E_B = 8.0$, $\epsilon = 1.6196 \times 10^{-6}$, $T_I = 3.0$, and $E_I = 20$. The resolution used corresponded to 102,400 grid points for η going from -2.5 to $+2.5$. According to the validation study for single step kinetics¹⁰, this resolution is adequate at least for moderate activation energies. In the chain-branching scheme, stiffness becomes an issue only when far outside the limit. Detailed results are shown for post-shock states inside the explosion zone, specifically for $\alpha = 1.01, 1.91$, and 2.16 . All of which are well-converged. Results for $\alpha = 0.99$ and $\alpha = 1.01$ are very close to those for $\alpha = 1.0$. Figure 1 shows the early part of the hot spot formation process for $\alpha = 1.01$. The later part of the process, namely, shock formation is shown in Figs. 2 and 3. Next, Figs. 4 and 5 show similar results for $\alpha = 1.91$. Finally, Figs. 6 and 7 correspond to $\alpha = 2.16$. All results are shown using η on the abscissa. The time to ignition is orders of magnitude larger when outside the chain-branching zone. Times are approximately 240 times longer for $\alpha = 0.5$ than for $\alpha = 2.16$. However, for α close to 1, they are only approximately 7.5 times larger. Practically, an extremely long tube would be required for a detonation to appear in shocked mixture outside the chain-branching region.

Figure 1 shows that for $\alpha = 1.01$, temperature is monotonically increasing as we move from the shock to the contact surface, and that the latter initially located at $\eta = 0$, is slowly moving to the right. This is due to thermal expansion induced by the chemistry which acts as a source of volume, venting on both directions. The reactant mass fraction during the early part of the process is consumed at a higher rate as time advances and as we

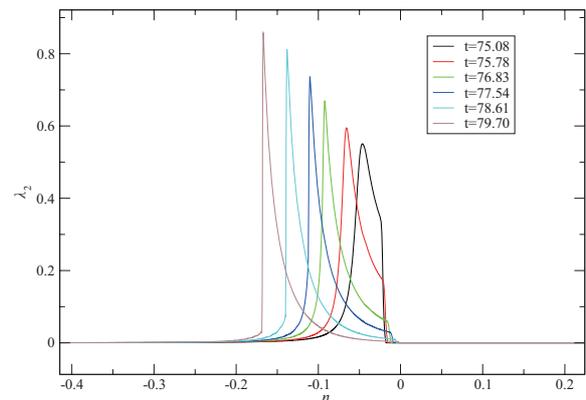


Fig. 3 $\alpha = 1.01$: Mass fraction of the chain-branching specie during shock formation phase

approach the contact surface. The consumption of reactant is due to the combined action of initiation and branching steps. In Figs. 2 and 3 the pressure, temperature and mass fraction of chain-branching specie profiles during shock formation phase are shown, Fig. 2 illustrates how the hot spot that was initially growing slowly accelerates as a result of the temperature increase in its vicinity producing higher reaction rates, and stronger chemistry. This spontaneous wave moves left and rapidly steepens to form a shock which eventually turns into a detonation wave that approaches a steady ZND structure propagating in shocked mixture. Also, the temperature behind the contact surface during the shock formation phase increases as the fluid is being compressed due to pressure waves emanating from the reaction zone.

Cases $\alpha = 1.91$ and $\alpha = 2.16$ are not very different from

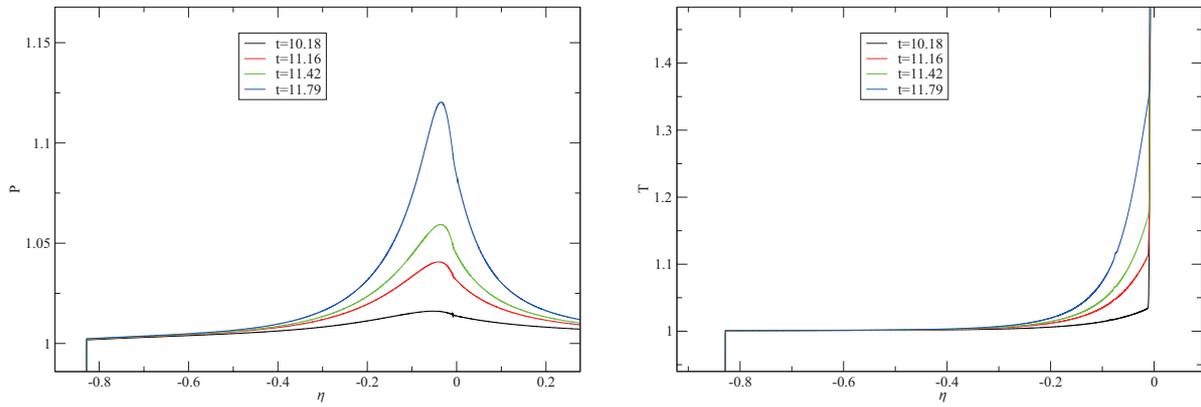


Fig. 4 $\alpha = 1.91$: Pressure and temperature during hot spot formation

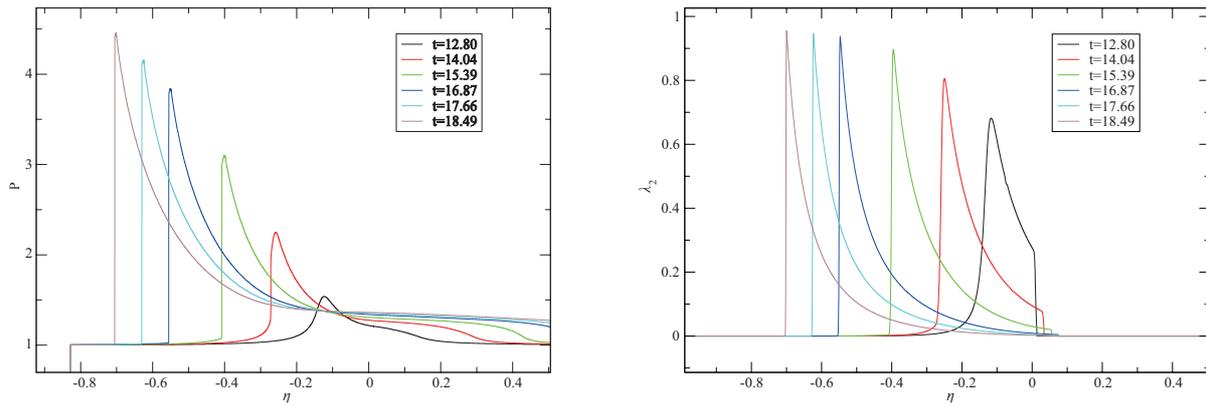


Fig. 5 $\alpha = 1.91$: Pressure and chain-branching specie during shock formation phase

the previously described case. The latter is faster and on η plots, the hot spot appears wider, due to the same η range at an earlier time corresponding to a smaller distance. That the behavior is similar appears to conflict with experimental evidence²⁾ whereby strong ignition occurs if deep inside the chain-branching zone while weak ignition occurs close to the limit. However, according to Meyer & Oppenheim²⁾, in the weak case, flames are observed, which the current non-diffusive model does not capture (except due to numerical diffusion), so that these flames and/or propagation of the interface between burnt or hot mixture and cold reactive mixture are not properly modeled in the current physical model. Likewise, any multidimensional effects or instability associated with the contact surface or flame is not simulated. These should play a bigger role closer to the limit because it takes longer for a hot spot to appear. As a consequence the current model may not capture the weak ignition case.

For α close to unity, the behavior is similar to that within the explosion region, even for α slightly below the limit, confirming an observation in Bédard-Tremblay *et al.*¹²⁾ whereby there is no sharp difference in behavior at the limit, but rather, a progressive transition between two extreme behaviors. In all cases, temperature initially peaks at the contact surface, but eventually the temperature maximum moves up and approaches a position close behind the shock that forms, similar to that in a steady ZND wave. In contrast with the cases closer to the limit, for $\alpha = 2.16$, the structure is still very transient as the new secondary shock approaches the original leading

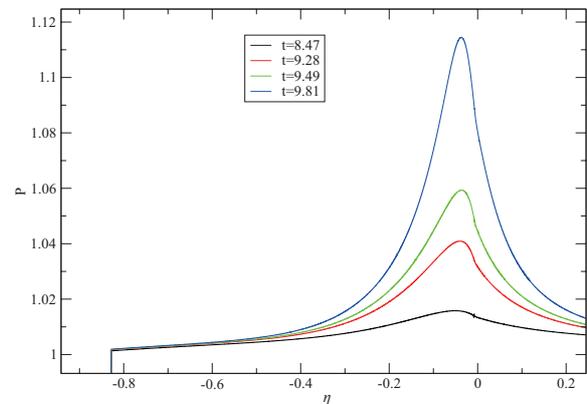


Fig. 6 $\alpha = 2.16$: Pressure during hot spot formation

shock. This is consistent with the relatively short distance between the contact surface and the shock. Fig. 7 (left) is a continuation of Fig. 6. Initially, the hot spot grows in one place, and then as adjacent parcels of gas ignite in turn a pressure wave moves away from the contact surface. The steepening of the pressure waves can be observed in the plots between times 10.65 to 11.68. It is evident that between times 11.68 and 12.80, a secondary shock is born, different from the leading shock located at approximately located at $\eta = -0.83$. Indeed, in all cases shown the birth of a secondary shock was captured by our model.

6. Conclusion

The approach proposed, based upon replacing space as an independent variable by the ratio space/time, was

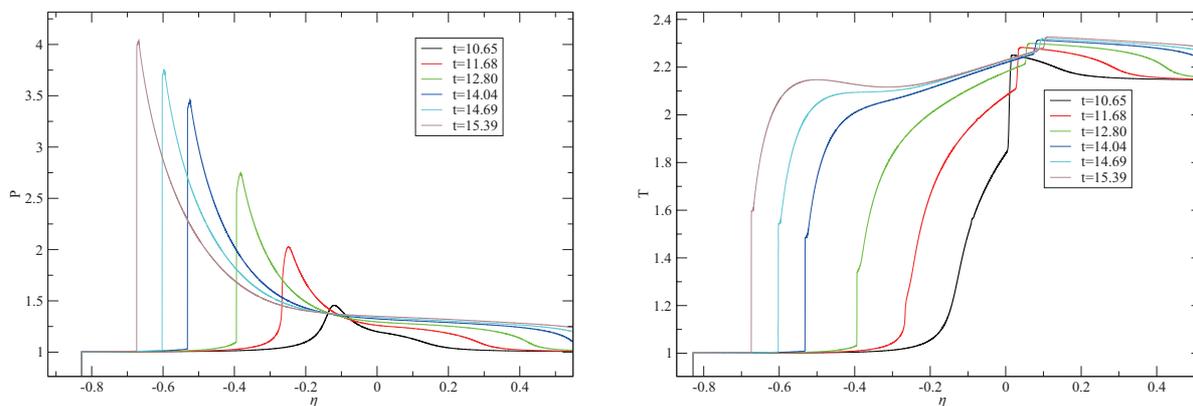


Fig. 7 $\alpha = 2.16$: Pressure and temperature during shock formation phase

shown to be reliable for this difficult problem, providing enough resolution and resolving the uncertainties due to the singular nature of the initial conditions in the shock-initiated ignition problem. It also allowed for a reasonably sized computation, although well resolved both in space and in time. Results show the difference in behavior when the post-shock state is close to the limit or inside the chain-branching explosion zone, and the corresponding difference in induction times. Results far inside the explosion region are consistent with experimental evidence²⁾ in the case of strong ignition; however it appears that the Euler physical model is not able to reproduce the weak ignition model, when closer to the limit.

Acknowledgments

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