

Critical Ignition in Rapidly Expanding Flows Described by a Power Law

Maxwell, B. M. and Radulescu, M. I.

Department of Mechanical Engineering, University of Ottawa
Ottawa, Canada

1 Introduction

Quenching of chemical reactions in the presence of strong gasdynamic expansions has been identified as an important mechanism controlling ignition behind decaying shocks, as applied to the detonation wave reaction zone structure [1-3], detonation wave initiation by Taylor-Sedov blast waves [4], detonation diffraction at an abrupt area change [5, 6] and shock induced diffusion-ignition of transient under-expanded jets [7]. To appreciate the generic mechanism controlling this process, consider the exothermicity following a mass element of reacting inviscid fluid that has just been compressed by a shock wave, or suddenly heated by other means. The subsequent rate of change of sensible energy e of the fluid particle (by definition a closed thermodynamic system) is given by the first law of thermodynamics, which relates it to the rate of energy addition, in this case by chemical reactions, and the rate of work done on the particle by the surrounding fluid.

$$\frac{De}{Dt} = \dot{q} + \frac{p}{\rho^2} \frac{D\rho}{Dt} \quad (1)$$

Here the derivatives denote the usual material derivative (or Lagrangian derivative) in fluid dynamics following a fluid element of fixed mass. The heat release term, \dot{q} , is usually positive and tends to increase the temperature, potentially leading to a thermal explosion [12]. For the flow behind a decaying shock wave, the second term, $(p/\rho^2)(D\rho/Dt)$, is negative due to the volumetric expansion of the fluid particles responsible for the shock weakening. This expansion term thus acts as an energy sink. Hence there are two competing terms that affect the ignition process, whose balance dictates the rate of temperature change and the outcome of the thermal explosion process. The problem thus bears close similarity to the well-studied problem of homogeneous ignition in the presence of heat loss to the container walls (e.g., [8]).

Lundstrom and Oppenheim [1], henceforth denoted as LO, first suggested that the reactions behind decaying shocks can be altogether quenched. They employed an extension of a quasi-steady ignition model for the thermally neutral induction zone coupled with the flowfield dictated by a decaying self-similar blast wave. The ignition model is however not realistic because no energy is liberated during the induction period, which would otherwise influence the ignition time history of the thermal explosion. At present, it is not clear whether such criticality is a consequence of the assumed thermally neutral reaction

zone in their model, or whether more realistic reaction models will eventually ignite, albeit after a very long time.

In a study of initiation of detonations, Eckett, Quirk and Shepherd[4], henceforth denoted as EQS, suggested that such criticality exists for reactions behind decaying shocks. They derive an approximate asymptotic model based on one step kinetics by assuming that the losses can be estimated at the state immediately behind the shock, which they then apply to complex chemistry calculations for validation. The complexity of the set-up, however, could not easily substantiate the conclusions reached. Maxwell and Radulescu[7] also studied this criticality of ignition using complex chemistry for self-similar reactive jet start-up. Likewise, the use of complex chemistry prevented them to clearly identify what causes extinction.

The present work thus attempts to clarify the role of the competition between reactivity and expansion on the ignitability of the gases exposed to volumetric expansion in a simple well-posed generic problem amenable to analysis. To simplify the analysis, following LO, it is *assumed a priori* that the expansion term follows a power law in time, i.e.,

$$\rho \sim t^{-n} \quad (2)$$

In many cases, the constant n can be obtained analytically under different approximations for self-similar blast wave problems of the type considered by LO and EQS, for self-similar jet start-up [9] and also for steady problems, where the expansion can follow a power law in time along a streamline due to flow divergence. The equivalence between steady hypersonic flows and self-similar unsteady blast waves [10] permits us to interchangeably apply the same results to both steady and unsteady flows. Evidently, the same problem can be also posed as a gas contained in a piston-cylinder assembly, where the piston is recessed at a prescribed rate such that the gas density decays as a power law in time. Furthermore, a local self-similar solution can always serve as an intermediate asymptotic regime in more complex problems [11]. It is thus this generic form of loss term that we are assuming in the present study, in order to clarify the dynamic evolution of the ignition process and the inherent ignition criticality.

2 The gas model

Starting with the exact energy equation (1) for an inviscid non-heat conducting reactive gas, similar to LO, we start with the *ansatz* that density can be assumed to decay as a power law in time, i.e., $D \ln \rho / D \ln t = -n$, where n is a constant to model the self-similar expansion. For simplicity, we will assume an ideal gas with constant specific heats, where the specific internal energy, pressure and specific gas constant are given respectively by $e = c_v T$, $p = \rho R T$, $c_v = R / (\gamma - 1)$ where R is the ideal gas constant, T is the temperature and γ is the isentropic exponent. Both the reactant and product are assumed to have the same constant specific heat and molar mass, again for simplicity. The conservation of energy can thus be written as:

$$c_v \frac{DT}{Dt} = -Q \frac{D\alpha}{Dt} - n R \frac{T}{t} \quad (3)$$

where Q is the chemical energy available, $D\alpha/Dt$ represents the rate of reactant depletion, and α is a progress variable representing the molar fraction of reactant, ranging from 1 to 0. For simplicity, the gas is assumed to undergo an irreversible first order Arrhenius reaction such that the rate of depletion of the reactant is given by:

$$\frac{D\alpha}{Dt} = -k\alpha e^{-E_a/RT} \quad (4)$$

where E_a is the activation energy and k is the reaction rate constant.

The initial conditions can be formulated when the unreacted mass particle ($\alpha = 1$) first crosses the shock at some reference time $t = t_1$, or is when the mass particle is injected in an expanding flow, and reaches the initial temperature $T = T_1$. This way, the variable t_1 can serve as a Lagrangian coordinate for that particular particle path. It is also convenient to introduce a new time coordinate t' which denotes the time elapsed after the particle has been shocked, i.e., $t' = t - t_1$. Time and temperature are non-dimensionalized by $\tilde{t} \equiv t'/t_{\text{ig}}$; $\tilde{T} \equiv T/T_1$. For analytical convenience we have used the constant density ignition delay as the characteristic time scale (see below), which is given by $t_{\text{ig}} = k^{-1}\varepsilon q^{-1}e^{1/\varepsilon}$ where ε and q are respectively the non-dimensional inverse activation energy and heat release given by $\varepsilon = (E_a/RT_1)^{-1}$ and $q = Q/(c_v T_1)$. Therefore, the governing equations become

$$\frac{D\tilde{T}}{D\tilde{t}} = \varepsilon\alpha e^{1/\varepsilon} e^{-1/\varepsilon\tilde{T}} - \frac{n(\gamma-1)\tilde{T}}{\tilde{t} + nD_a}; \quad \frac{D\alpha}{D\tilde{t}} = -\frac{\varepsilon\alpha}{q} e^{1/\varepsilon} e^{-1/\varepsilon\tilde{T}}; \quad \tilde{T}(\tilde{t}=0) = \alpha(\tilde{t}=0) = 1 \quad (5)$$

Clearly, the two parameters entering the temperature evolution equation controlling the expansion is the exponent n and a Damkohler number D_a , which is given by the ratio of the expansion time scale $t_{\text{exp}} = (D\ln\rho/Dt)^{-1} = t_1/n$ evaluated at the initial time t_1 and the characteristic ignition delay in the absence of expansion t_{ig} , yielding

$$D_a \equiv \frac{t_{\text{exp}}}{t_{\text{ig}}} = \frac{t_1/n}{t_{\text{ig}}} \quad (6)$$

3 Numerical solution

The evolution of the temperature was first determined numerically by integrating the system of first order ordinary differential equations (5). An example showing the character of the solution obtained is shown below for parameters $\varepsilon = 0.1$, $\gamma = 1.2$, $q = 20$, $n = 1$. The power law exponent for density decay corresponds approximately to spherical Taylor-Sedov blast waves.

It was found that below a critical value of Damkohler number, the ignition is quenched. Figure 1 shows an example of the temperature evolution obtained near the ignition criticality for values of the Damkohler number bracketing the critical condition for ignition. In this case, the critical limit is between $D_a = 0.99454922$ and $D_a = 0.99454923$. For values above this threshold, ignition was observed at earlier time. An ignition event is characterized by a sudden increase in temperature and complete consumption of the reactant. For sub-critical values of Damkohler number, a more rapid extinction was observed as the Damkohler number was lowered. This can be seen in Figure 1. Also shown is the non-reactive solution (obtained for $q = 0$) of equation (5) given by $T_{\text{NR}} = (t/nD_a + 1)^{-n(\gamma-1)}$ and the solution obtained without the loss term displaying the classical thermal explosion character [12]. A few observations are noteworthy. First note that there is a clearly defined separatrix between the solutions that lead to ignition from the ones that do not. As the critical Damkohler number is sought with further precision, the time for ignition shifts to larger times, which can be orders of magnitude longer than in the homogeneous case. Another interesting observation pertains to the change in solution behaviour in the subcritical case of ignition, below the separatrix. After the failed ignition event, it can be seen that the rate of decay of temperature follows the non-reactive solution, suggesting that the reactions are fully quenched.

4 Critical ignition limit

In order to get further insight into the dependence of the critical Damkohler number on the parameters of the system, we have conducted an analysis based on large activation energy asymptotics. Since in practice the activation energy is usually quite large, i.e., ε is small, we seek an asymptotic expansion of the form:

$$\tilde{T} = \tilde{T}_1 + \varepsilon \tilde{T}_2 + O(\varepsilon^2); \alpha = \alpha_1 + \varepsilon \alpha_2 + O(\varepsilon^2) \quad (7)$$

The exponential can thus be approximated as usual

$$e^{-1/\varepsilon \tilde{T}} = e^{-1/\varepsilon (\tilde{T}_1 + \varepsilon \tilde{T}_2 + O(\varepsilon^2))} = e^{(-1/\varepsilon)(\tilde{T}_1 - \varepsilon \tilde{T}_2 + O(\varepsilon^2))} = e^{-\tilde{T}_1/\varepsilon} e^{\tilde{T}_2 + O(\varepsilon)} \quad (8)$$

The governing equations (5) become

$$\begin{aligned} \frac{D}{D\tilde{t}} \left(\tilde{T}_1 + \varepsilon \tilde{T}_2 + O(\varepsilon^2) \right) &= \varepsilon \alpha e^{(1-\tilde{T}_1)/\varepsilon} e^{\tilde{T}_2 + O(\varepsilon)} - n(\gamma - 1) \frac{\tilde{T}_1 + \varepsilon \tilde{T}_2 + O(\varepsilon^2)}{\tilde{t} + nD_a} \\ \frac{D}{D\tilde{t}} \left(\alpha_1 + \varepsilon \alpha_2 + O(\varepsilon^2) \right) &= -\frac{\varepsilon(\alpha_1 + \varepsilon \alpha_2 + O(\varepsilon^2))}{q} e^{(1-\tilde{T}_1)/\varepsilon} e^{\tilde{T}_2 + O(\varepsilon)} \\ \tilde{T}_1(0) + \varepsilon \tilde{T}_2(0) + O(\varepsilon^2) &= \alpha_1(0) + \varepsilon \alpha_2(0) + O(\varepsilon^2) = 1 \end{aligned} \quad (9)$$

We seek the solution to the system above in the asymptotic limit $\xi \equiv n(\gamma - 1)\varepsilon^{-1} = O(1)$. This last requirement essentially puts bounds on the expansion strength and specific heat ratio. Three cases can be considered

$$n = O(\varepsilon) \text{ and } (\gamma - 1) = O(1); n = O(1) \text{ and } (\gamma - 1) = O(\varepsilon); n = O(\varepsilon^\nu) \text{ and } (\gamma - 1) = O(\varepsilon^{1-\nu}) \quad (10)$$

The first case in (10) corresponds to a weak expansion. The second case corresponds to a strong expansion in the Newtonian limit $\gamma \rightarrow 1$, while the latter is the general intermediate case. Imposing (10) in (9), we get to leading order

$$\frac{D\tilde{T}}{D\tilde{t}} = \frac{D\alpha_1}{D\tilde{t}} = 0, \tilde{T}_1(0) = \alpha_1(0) = 1 \quad (11)$$

From which we deduce that

$$\tilde{T}_1(t) = \alpha_1(t) = 1 \quad (12)$$

At order ε , we get

$$\frac{D\tilde{T}_2}{D\tilde{t}} = e^{\tilde{T}_2} - \frac{\xi}{\tilde{t} + nD_a}, \frac{D\alpha_2}{D\tilde{t}} = -\frac{e^{\tilde{T}_2}}{q}, \tilde{T}_2(0) = \alpha_2(0) = 1 \quad (13)$$

These expressions can be integrated exactly to find the dependence of the perturbation on time given the appropriate Damkohler number as initial condition. Before doing so, it is interesting to investigate

the importance of the parameter ξ in front of the loss term by integrating (13) with either the energy source term or the loss term individually. The chemical source term allows the exponential of the perturbation temperature to grow like $(1 - \tilde{t})^{-1}$ and become infinite as $\tilde{t} \rightarrow 1$ (hence our choice for the characteristic ignition time). On the other hand, the loss term causes the exponential of temperature decay like $(\tilde{t}/nD_a + 1)^{-\xi}$. Clearly, for $\xi < 1$, the production term will always dominate the loss term, hence ensuring that ignition is always achieved. The more interesting situation, however, is when $\xi > 1$. In this case, the loss term can overcome the production term if the Damkohler number D_a is sufficiently small. Formally integrating (13) yields

$$\tilde{T}_2(\tilde{t}) = -\xi(\xi - 1)^{-1} \ln(\Phi) - \ln \left((\xi - 1)^{-1} \Phi^{\xi/(\xi-1)} (nD_a + \tilde{t}) \left(1 - \Phi^{1/\xi} (nD_a + \tilde{t})^{\xi-1} \right) \right) \quad (14)$$

$$\Phi = \frac{1 - \xi + nD_a}{(nD_a)^\xi} \quad (15)$$

When $\xi > 1$, \tilde{T}_2 grows to large values in finite time only if $\Phi > 0$. We interpret this condition as the condition for ignition. The resulting critical value for the Damkohler number becomes

$$D_a^* = \frac{\gamma - 1}{\varepsilon} - \frac{1}{n} \quad (16)$$

or in dimensional terms,

$$\frac{t_1}{t_{\text{ig}}} = n(\gamma - 1) \frac{E_a}{RT_1} - 1 \quad (17)$$

Figure 2 shows the comparison between the analytical prediction for the critical Damkohler number as a function of inverse activation energy for $\xi = 2$ and $n = 1$ and letting the inverse activation energy ε and isentropic exponent γ vary. For large activation energies ($1/\varepsilon$), the agreement is very good. At a small activation energy of approximately 3, which is usually smaller than most typical values for combustible gases, the prediction overestimates the numerically determined limit by approximately 5 percent, which can be considered quite good.

5 Conclusions

In conclusion, we demonstrated numerically and analytically using a generic simple model that the presence of a power-law expansion can suppress ignition provided the expansion is strong enough or the chemical reactions sufficiently sensitive to changes in temperature. The present study thus confirms the earlier findings of LO, EQS, (Maxwell and Radulescu)[7] and [13] for the existence of a critical expansion rate that can suppress ignition under different reacting fluid dynamic problems.

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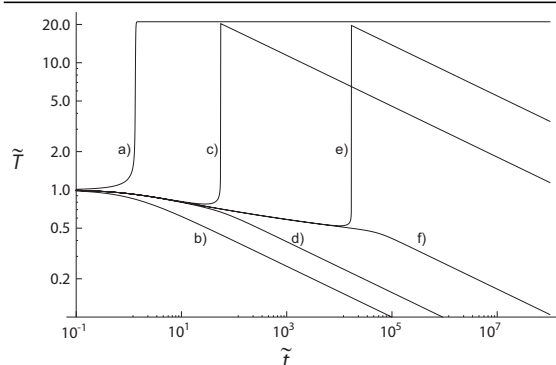


Figure 1: Temperature evolution near the critical ignition bifurcation; a) homogeneous thermal explosion, b) inert self-similar expansion, c) $D_a = 1$, d) $D_a = 0.99$, e) $D_a = 0.99454923$, and f) $D_a = 0.99454922$.

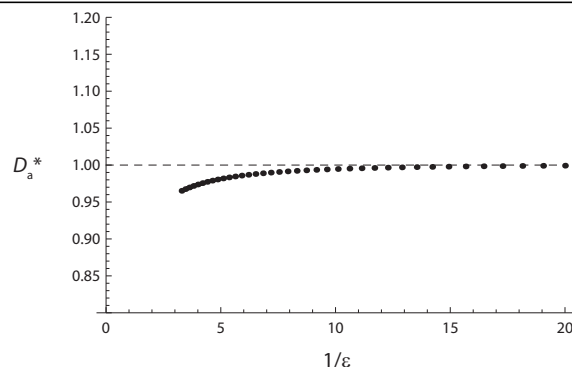


Figure 2: The critical Damkohler number obtained numerically (points) compared to the closed form solution for $\xi = 2$, $n = 1$ and varying the inverse activation energy ε and γ .

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