Critical initial conditions for spontaneous thermal ignition

R. O. Weber, E. Balakrishnan and G. C. Wake

a School of Mathematics and Statistics, University College of UNSW, ADFA Canberra 2600, Australia. E-mail: r-weber@adfa.oz.au
b Department of Mathematics and Statistics, Sultan Qaboos University, Sultanate of Oman
c Department of Mathematics, University of Canterbury, Christchurch, New Zealand

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Critical initial conditions for thermal ignition are calculated numerically for planar, cylindrical and spherical geometries. In each case a comparison is made on the bifurcation diagram with the intermediate steady state(s). It is clearly seen that the actual critical initial condition which depends upon the initial temperature profile, is partly below the intermediate steady state(s). The variation with ambient temperature, exothermcity and Biot number are all explicitly shown. The results are important for calculations relevant to the critical hot product assembly problem where a hot manufactured organic product is packed into containers or stockpiled at a subcritical ambient storage temperature. An example calculation for an interesting case study (milk powder) is given to illustrate the utility of our results.

1 Introduction

In the manufacturing industries, a product is often produced at an elevated temperature and subsequently packed into large containers before it can cool. If the product is an organic material, such as milk powder, there is the possibility that exothermic oxidation will further increase the temperature. At best, the higher temperatures cause degradation of the product, at worst, ignition may occur. Previous studies of this situation have only partially enabled the accurate prediction of critical initial temperature distributions which distinguish between the safe situation and the potentially dangerous one.

Mathematical investigations of the reaction–diffusion models used to describe this situation, began with Fujita, who worked with an exponential reaction function \( e^{-bT} \). This was thought to be a useful approximation to the Arrhenius temperature dependence of the reaction rate \( e^{-bRT} \), as follows from the use of the Frank-Kamenetski definition for non-dimensional temperature. Most subsequent work, such as Lacey and Wake and Weber and Barry, and the references therein, have used the same functional form for the reaction term. Gray and Scott performed analogous calculations to those described below, using the Frank-Kamenetski temperature variable and approximation. (Additionally, they only considered uniform initial conditions, rather than spatially distributed temperature profiles.) The exponential dependence on temperature meant that, above the initial condition threshold, the temperature–time behaviour inevitably led to infinite temperatures (often called blow-up). Furthermore, the exponential approximation is known to be accurate only at low temperatures and the complete bifurcation diagram is in fact qualitatively wrong as it contains no upper branch. This upper branch plays an important role for critical initial condition calculations, whenever there is any but the lowest temperature excess.

Gray and Wake and some of the references therein use the definition \( u = RT/E \) for non-dimensional temperature and retain the full temperature dependence in the Arrhenius function; namely \( e^{-1/T} \). There are two principal advantages to this definition and the retention of the full temperature dependence: (1) The ambient temperature retains its role as an independent control parameter. (2) The bifurcation behaviour at high temperatures, important to the evolution of initial conditions, is not compromised by any approximation.

However, Gray and Wake made the modelling approximation of replacing the non-dimensional temperature in the Arrhenius function with its average value \( u_0 \) (other spatially uniform approximations to the heat release rate could be used). That is, Gray and Wake approximated \( e^{-1/u_0} \approx e^{-1/\bar{T}} \); in order to derive some of their analytical results. As a consequence there remains some uncertainty about whether or not their results provide a close bound, preferably conservative, on the critical initial conditions.

In the present study we report on the results for critical initial conditions, making no approximations in the equations. By necessity, this means that our results are mostly obtained numerically (as we shall describe), but we endeavour to present them in a useful form, so that it is possible to garner critical initial conditions in dimensional variables for practical situations. Many fires have resulted from being assembled at too high an initial temperature even though the storage conditions were known to be sub-critical as far as the steady state theory is concerned, see for example Bowes (who refers to this as thermal explosion of the second kind), Rivers et al. and Smedley and Wake.

A key idea for the determination of critical initial conditions is to determine whether the critical situation distinguishes between initial conditions that evolve to a high-temperature steady state and those that evolve to a low-temperature steady state. Implicit in this is the assumption that such high- and low-temperature steady states actually exist. For the model considered here there is a large body of literature concerned with the steady states, most of which can be accessed from ref. 1–10. We can summarise the findings as follows. For \( \lambda \) greater than a transitional value and a range of \( u_0 \) dependent upon \( \lambda \) (always between \( u_0 = 0 \) and \( u_0 = 0.25 \) with maximum value

decreasing as $\lambda$ increases) there are three steady states (or more for certain geometries such as the sphere). These include a low-temperature steady state and a high-temperature steady state, both of which are stable to small perturbations in temperature. Above the stable low-temperature steady state there is an unstable intermediate steady state which acts as a 'watershed' for initial conditions. This watershed is a shape-dependent function.

2 Mathematical model

We shall assume that well established mathematical models$^2$ can be used to describe the self-heating phenomena which occur in many organic materials. Specifically we ignore reactant consumption, so we need only consider the conservation of energy:

$$\rho c_p \frac{\partial T}{\partial t} = k \nabla^2 T + \rho Q A e^{-E/RT}$$

This equation applies to the interior of the combustible material for positive time. In this paper we shall be concerned to complete the description of a well posed problem that can only occur in many organic materials. Specifically we ignore reaction throughout the material must also be prescribed in order to complete the description of a well posed problem that can only occur in many organic materials. Specifically we ignore reaction

$$L \frac{\partial T}{\partial n} = -\frac{h S}{\Omega} (T - T_s)$$

where $\partial / \partial n$ represents the derivative in the direction normal to the surface of the material. The initial temperature distribution throughout the material must also be prescribed in order to complete the description of a well posed problem that can be solved,

$$T(x, 0) = T_0 + C g(x)$$

In the following description of results, we shall consider several families of initial temperature distribution, each given by a different form for $g(x)$. For comparison purposes we shall always scale $g(x)$ so that $g_{max} = 1$.

To non-dimensionalise the model we first substitute $u = RT/E$ and then rescale the space and time coordinates. This results in

$$\frac{\partial u}{\partial t} = V^2 u + \lambda e^{-1/u}, \ x \in \Omega, \ t > 0$$

(1a)

$$\frac{\partial u}{\partial n} + Bi (u - u_s) = 0, \ x \in \partial \Omega, \ t > 0$$

(1b)

$$u(x, 0) = u_s + C g(x), \ x \in \Omega$$

(1c)

where the region occupied by the combustible material is labelled $\Omega$ and its boundary is $\partial \Omega$. Note that although the space and time coordinates have been scaled, they are still called $x$ and $t$. At the conclusion we shall demonstrate how to return to dimensional coordinates and provide an example calculation to illustrate the application. Meanwhile, the non-dimensional coordinates will be used to minimise the number of repetitive computations. The Biot number is a non-dimensional measure of the heat transfer from the material to the surroundings. Infinite Biot number is when the boundary temperature is prescribed as being equal to $u_s$ (Dirichlet condition). The constant $C$ in eqn. (1c) has been non-dimensionalised with the same factor as the temperature.

3 Mathematical observations

In this section we consider the general behaviour of the solutions to the mathematical model of eqns. (1a)–(1c). In particular, we shall describe the expected dependence of the critical value of the initial condition, $u_s + C g(x)$ upon the three parameters $u_s$, $\lambda$, $Bi$, all of which can vary independently to accommodate different physical scenarios. The numerical results in the following sections bear witness to our expectations here. Nevertheless, we should note that some of our statements below are yet to be formally proven.

We begin by noting that if the initial condition has a single maximum interior to the region $\Omega$, then the solution $u(x, t)$ also has a single maximum interior to $\Omega$. This follows from noticing that if the Laplacian, $\nabla^2 u$, is initially negative, it remains so.

Next we state that the steady problem (with $u_s = 0$) has at least three steady states provided $\lambda > \lambda_s$ and for some $u_s$ (less than 0.25). The lower and upper steady states, $u(x)$ and $\bar{u}(x)$ respectively, are locally asymptotically stable. There is at least one intermediate steady state $u_m(x)$ which is unstable. It has not been possible to establish this observation rigorously. Suffice to say that all computations (e.g. ref. 5–9) demonstrate that this holds. Indeed for many simply connected $\Omega$ regions there are exactly three such steady solutions.

Given that there are just three steady states $u(x)$, $u_m(x)$ and $\bar{u}(x)$, one can establish that all initial conditions will evolve to either the lower $u(x)$ or the upper $\bar{u}(x)$ steady state. Furthermore, the intermediate steady state acts as a 'watershed'. Namely, initial conditions which are entirely below or above $u_m(x)$ will not cross $u_m(x)$ at any $x$ and will evolve to either $u(x)$ or $\bar{u}(x)$ according to where they begin. If there are more than three steady states (as can occur in three dimensions) then the intermediate steady states are no longer exact 'watersheds' and, indeed, the long term behaviour of initial conditions lying entirely between unstable steady states is less certain.

We are then led to make our main hypothesis. Namely, that for each family of initial conditions, in every geometrical situation, $u(x, 0) = u_s + C g(x)$, there exists a critical value, $C_{cr}$, such that any initial conditions with $C < C_{cr}$ evolve to the lower (cool) steady state $u(x)$ and any conditions with $C > C_{cr}$ evolve to the upper (hot) steady state $\bar{u}(x)$. That is, for each family of initial conditions, $C_{cr}$ plays the role of a watershed.

It then follows that, for each geometrical situation, the value of $C_{cr}$ is dependent upon the three parameters $u_s$, $\lambda$, $Bi$. As $u_s$ is the ambient temperature, $\lambda$ is proportional to the exothermicity, and $Bi$ (the Biot number) is the proportionality constant for heat flux exchange with the surroundings we expect that: (i) $C_{cr}$ will increase as $u_s$ decreases; (ii) $C_{cr}$ will increase as $\lambda$ decreases; (iii) $C_{cr}$ will increase as $Bi$ increases.

The following results from our computations show that these physically intuitive statements hold, at least for reasonable parameter ranges, which indicates the existence of a family of critical initial conditions i.e. one for each generic shape.

4 Uniform initial condition/planar geometry

To illustrate the key features, we begin with the simplest case which is where the initial condition is a constant $C$ added to the ambient temperature $[g(x) = 1$ in eqn. (1c)].

Furthermore, we consider only one space coordinate corresponding to the infinite slab, planar geometry and we take the Biot number to be infinite. In this case the symmetry about
\( x - 0 \) allows us to state the complete, well posed problem as

\[
\begin{align*}
\frac{\partial u}{\partial t} &- \frac{\partial^2 u}{\partial x^2} + \lambda e^{-1/x} \\
\frac{\partial u}{\partial x} (0, t) & = 0 \\
u(1, t) & = u_a \\
u(x, 0) & = u_x + C
\end{align*}
\]

for \( x \in [0,1] \) and \( t \in [0,\infty) \). We are then principally interested in determining the critical value of \( C \), which we denote \( C_{cr} \), as a function of \( \lambda \) and \( u_a \), which distinguishes between initial conditions that do lead to ignition [i.e. converge to the high-temperature steady state of eqn. (2)] and those which do not. An important related question is the time it takes for the initial condition to evolve to the high-temperature steady state; it is expected to increase markedly when \( C \) is just above \( C_{cr} \).

The approximate method of Gray and Wake\(^6\) can be used to estimate \( C_{cr} \); for example, when \( \lambda = 10^6 \) and \( u_a = 0.05 \) the estimate is \( C_{cr} \approx 0.0043 \). In addition to being a good estimate, this also assists in locating the exact value which needs to be numerically determined. Our numerical method is a second-order central differencing of the second derivative and Crank–Nicholson for the time iterations. This method is reasonably efficient and can be implemented in mathematical software programs such as MATLAB. For the present example it gave \( C_{cr} \approx 0.0043 \). In Fig. 1 we present full results for \( \lambda = 10^6 \) and \( Bi \to \infty \). It contains the stable lower and the unstable intermediate steady-state branches displayed as the maximum value of \( u(x) \) [namely \( u(0) \) because of the symmetry] as a function of the ambient temperature \( u_a \). The stable upper steady state is not shown as it is orders of magnitude above the other two and, in fact, is of no physical interest [in spite of the fact that it is an attractor, in practical situations ignition occurs long before \( u(x, t) \) is close to it]. Also displayed is the curve of \( u_a + C_{cr} \) vs. \( u_a \) (this curve is labelled with \( g(x) = 1 \) in Fig. 1) which is seen to lie below the unstable intermediate steady state by ca. 5–10%. This is a significant feature as it means that there is a range of initial conditions, which lie between the \( u_a + C_{cr} \) curve and the unstable intermediate steady state, which are not stable initial temperature configurations. We wish to stress that these are the sort of initial conditions which may be thought to be safe after only a casual inspection when, in fact, they are unsafe. We have also included the curve of \( u_a + C_{cr} \) when \( g(x) = 1 - x^2 \). This parabolic function would be expected to be a reasonable first approximation to the unstable intermediate steady state. Indeed the results [shown as the maximum critical temperature in this case, also \( u_a + C_{cr} \), which occurs at \( x = 0 \) where \( g(x) = 1 \) vs. \( u_a \) corroborate this expectation.

5 ‘Cylindrical’ and ‘spherical’ geometry

The complete well posed problem in these cases is

\[
\begin{align*}
\frac{\partial u}{\partial t} & - \frac{\partial^2 u}{\partial x^2} + \frac{1}{x} \frac{\partial u}{\partial x} + \lambda e^{-1/x} \quad \text{(cylindrical)} \\
\frac{\partial^2 u}{\partial x^2} & + \frac{2 \partial u}{\partial x} + \lambda e^{-1/x} \quad \text{(spherical)}
\end{align*}
\]

for \( x \in [0,1] \) and \( t \in [0,\infty) \). As for the planar case, we can use the approximate method of Gray and Wake\(^6\) to estimate \( C_{cr} \) for any choice for \( g(x) \). For example, for \( \lambda = 10^6 \), \( u_a = 0.05 \) and \( g(x) = 1 \), we find \( C_{cr} \approx 0.01397 \) (cylindrical) and 0.0195 (spherical). In Fig. 2 and 3, we present the full results for \( \lambda = 10^6 \) and \( Bi \to \infty \); showing the steady states, \( u(0) \), and \( u_a \).

Fig. 1 Comparison of \( u(0) \) vs. \( u_a \) from the solution of the steady problem, with the critical initial conditions obtained numerically when \( u(x, 0) = u_a + C \) [lower (---)] and \( u(x, 0) = u_a + C (1 - x^2) \) [upper (--)]. Only the maximal, central temperatures (at \( x = 0 \)) are shown. All computations are for the ‘planar’ geometry with \( \lambda = 10^6 \) and \( Bi \to \infty \).

Fig. 2 As Fig. 1 but for cylinder geometry.

Fig. 3 As Fig. 1 but for spherical geometry."
Fig. 4 Variation of critical initial condition with \( \lambda \). Planar geometry with \( u_0 = 0.03 \) and \( Bi \rightarrow \infty \).  

\[ g(x) = 1 \]

\[ g(x) = 1 - x^2 \]

\[ u_0 + C_\alpha g(0) \]

\[ g(x) = (1 - x^2)^2 \]

\[ u_0 + C_\alpha g(0) \]

Fig. 5 Variation of critical initial condition with \( Bi \). Planar geometry with \( u_0 = 0.03 \) and \( \lambda = 10^6 \).  

\[ g(x) = 1 \]

\[ g(x) = (1 - x^2) \]

\[ u_0 + C_\alpha g(0) \]

\[ g(x) = (1 - x^2)^2 \]

\[ u_0 + C_\alpha g(0) \]

\[ g(x) = 1 \]

\[ g(x) = (1 - x^2)^2 \]

\[ u_0 + C_\alpha g(0) \]

\[ g(x) = 1 \]

\[ g(x) = (1 - x^2)^2 \]

\[ u_0 + C_\alpha g(0) \]

\[ g(x) = 1 \]

\[ g(x) = (1 - x^2)^2 \]

\[ u_0 + C_\alpha g(0) \]
thermal conductivity
normal direction to surface
time variable, scaled by \((k/pc_pL^2)\)
non-dimensional temperature
average value of \(u\)
non-dimensional ambient temperature
adiabatic burnt temperature, average thereof
upper solution to the steady-state problem
lower solution to the steady-state problem
space variable, scaled by \(L\)
fuel consumption parameter = \(c_pEQR\)
Frank-Kamenetskii\(^2\) non-dimensional temperature
density
non-dimensional parameter

References