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Using the Method of Lines to determine critical conditions for thermal ignition

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Abstract There are many situations in which the critical conditions for thermal ignition cannot be determined analytically. These include cases where the chemistry needs to be properly considered, where the geometry is not just the simplest and where other processes must be included. In these circumstances, numerical (or at least semi-analytical) means are used to determine critical conditions for thermal ignition. Once confronted with a numerical approach to solving a problem, it is necessary to be a little circumspect about the results and seek independent means to corroborate them. For this reason, the present paper reports on the Method of Lines to investigate a recent reactive hotspot problem which has previously been shown to display unexpected behaviour and demonstrates the use of sensitivity analysis to rigourously determine criticality in such a dissipative system.

Keywords Critical conditions · Method of Lines · Thermal ignition

1 Introduction

The study of thermal ignition initiated by a hot spot has a long history in the combustion literature [1]. For most industrial purposes such an event is undesirable as numerous factory fires have been attributed to such phenomena; these are often due to the accumulation of reactive material close to hot surfaces. On the other hand, hot-spot ignition via lasers is currently being developed for safe and controlled detonation of explosives such as cluster bombs [2]. Determining the conditions for thermal ignition is therefore extremely useful for many practical applications. However, these conditions can only be determined by numerical (or at least semi-analytical) means.

In this paper we revisit the problem investigated by Brindley et al. [3] and McIntosh et al. [4]. Their study deals with a constant-power heat source (hot spot) embedded in a concentric spherical volume of

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reactive material (with relatively low exothermicity). The former study was based mainly on numerical investigation of the model using the NAG library routine `D03PBF`, whereas the latter study examined an approximate model for such a system. Brindley et al. [3] showed "unexpected behaviour for small-sized power sources, in which, over a range of source radii, there are two distinct critical values for the power, with an apparently "safe" region occupying some of the range between them." Obviously such a result has serious safety implications for chemical-plant operations. In this paper we report on our use of the Method of Lines [5] to corroborate this unusual result and, more importantly, we also describe in detail our use of sensitivity analysis [6, 7] to allow a precise and rigorous determination of criticality. This last point is particularly noteworthy as the techniques of sensitivity analysis have not been all that widely used in the combustion literature and one major purpose of the current work is to address this shortcoming.

For completeness, we begin with a brief description of the Method of Lines (MoL) and the hot-spot ignition problem. We next discuss accuracy considerations, present the numerical results and are then able to explain in some detail the use and significance of sensitivity analysis.

2 Method of Lines

The MoL technique is a versatile approach to obtain numerical solutions to partial differential equations (in principle, all the major classes of elliptic, hyperbolic and parabolic, linear and non-linear, in one, two and three spatial dimensions). The method has been made popular in science and engineering largely through the work of W.E. Schiesser and coworkers.

The concept behind the MoL is that the partial derivatives in spatial directions are replaced by finite-difference approximations, as is normally done in the FTCS approach. This results in only time derivatives and often leads to a system containing only ordinary differential equations. Subsequently, all that is required to solve these equations is a good numerical ODE solver such as those found in MATLAB. However, application of the MoL can sometimes give rise to a mixed system of differential and algebraic equations (DAEs). These can also be solved utilizing slightly more complicated MATLAB functions, such as `ODE15s`. Other DAE solvers include `BESIRK` which is available in MAPLE [7].

3 Governing equations and the implementation of the numerical scheme

According to [3], the governing equations for the ignition of reactive material (including reactant consumption) by a localised constant-power heat source (hot spots) can be given as

$$\rho c_p \frac{\partial T}{\partial t} = QAX\mathrm{e}^{-E/RT} + k\left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r}\right), \tag{1}$$

$$\frac{\partial X}{\partial t} = -XA\mathrm{e}^{-E/RT}. \tag{2}$$

Here $T = T(r,t)$ is the temperature within the sphere whereas $X = X(r,t)$ is the reactant density. The condition $X = 0$ corresponds to a completely burnt, non-reacting volume of material, which will be assumed to have the same physical properties of the original reactant. As in most combustion-wave-propagation problems, the rate of heat release is assumed to be proportional to the reactant density.

The boundary conditions are

$$-k\frac{\partial T}{\partial r} = \frac{P}{4\pi r_0^2} \quad \text{at } r = r_0, \tag{3}$$

$$T(r_1,t) = T_\mathrm{a}, \tag{4}$$

which basically represent a constant heat flux over the spherical surface of the hot spot associated with a constant total power P (r_0 is the source radius), and the temperature at the radius of the reacting sphere is

at ambient temperature at all times (infinite Biot number). We shall assume that initially the reactants are all at ambient temperature and possess uniform reactant density, that is,

$$T(r,0) = T_a \quad \text{and} \quad X(t,0) = \rho. \tag{5}$$

We now proceed to discretise the governing equations and the boundary conditions to a form that can be used by the MoL. If we consider the variation of T with respect to r to take place along a grid defined in r (where a particular value of r will be specified in terms of an integer index i as is normally done in explicit finite-differences schemes), the partial derivative $\frac{\partial^2 T}{\partial r^2}$ can be replaced with an algebraic approximation that evaluates it at point i. The derivative $\frac{\partial T}{\partial t}$ is kept, and this leads to a system of differential equations with only one independent variable t; i.e., a system of ODEs. To replace the derivative $\frac{\partial T}{\partial r}$ and $\frac{\partial^2 T}{\partial r^2}$, the same second-order expression as used in the finite-difference scheme (central space) is substituted. In other words,

$$\frac{\partial T_i}{\partial r} \approx \frac{T_{i+1} - T_{i-1}}{2h}$$

$$\frac{\partial^2 T_i}{\partial r^2} \approx \frac{T_{i+1} - 2T_i + T_{i-1}}{h^2}$$

where $h = \dfrac{r_1}{n-1}$ is the grid spacing. Substituting these in Eq. 1 yields:

$$\frac{\partial T_i}{\partial t} = \frac{QXA\mathrm{e}^{-E/RT_i}}{\rho c_p} + \frac{k}{\rho c_p}\left(\frac{T_{i+1} - 2T_i + T_{i-1}}{h^2} + \frac{T_{i+1} - T_{i-1}}{r_i h}\right)$$

where r_i indicates the radial position of node i being evaluated. Rearranging gives

$$\frac{\partial T_i}{\partial t} = \alpha X\mathrm{e}^{-E/RT_i} + \frac{\beta}{r_i}\left((r_i + h)T_{i+1} - 2r_i T_i + (r_i - h)T_{i-1}\right) \tag{6}$$

where $\alpha = \dfrac{QA}{\rho c_p}$, $\beta = \dfrac{k}{\rho c_p h^2}$.

The reactant-depletion equation is kept as the dimensionalised form of Eq. 2, since the dependence on T_i is already accounted for in Eq. 6. Thus

$$\frac{\partial X_i}{\partial t} = -X_i A\mathrm{e}^{-E/RT_i}. \tag{7}$$

The boundary condition at the edge of the hot spot (i.e., at $r = r_0$) is calculated by a second-order approximation giving the boundary value here as

$$\frac{\partial T}{\partial r} = \frac{P}{-k4\pi r_0^2}$$

which becomes

$$\frac{P}{-k4\pi r_0^2} = \left(\frac{-3T_{\text{start}} + 4T_1 - T_2}{2h}\right)$$

and is re-arranged to give

$$\longrightarrow T_{\text{start}} = \frac{1}{3}\left(4T_1 - T_2 + \frac{2hP}{k4\pi r_0^2}\right). \tag{8}$$

The second boundary condition (4) and initial conditions (5) need not be transformed to employ the MoL. The governing equations together with the boundary and initial conditions described above are implemented in MATLAB to produce results that we discuss later.

3.1 Accuracy considerations: the heat-conduction problem without heat generation

To test the code we first consider the simpler problem of heat conduction without the generation of heat by an exothermic reaction; that is, we set $Q = 0$ and then the only phenomenon is the diffusion of heat from the hot spot into the material. Equation 1 becomes

$$\frac{\partial T}{\partial t} = \frac{k}{\rho c_p}\left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r}\right). \tag{9}$$

If we consider the steady-state equation, i.e., $\partial T/\partial t = 0$, then Eq. 9 can be easily solved together with the boundary conditions (3) and (4) to give

$$T_{\mathrm{ss}} = T_{\mathrm{a}} + \frac{P}{k4\pi}\left(\frac{1}{r} - \frac{1}{r_1}\right). \tag{10}$$

It is then possible to use Eq. 10 to determine whether the MATLAB code is producing accurate results and this was tested for values of $r_0 = 4\,\mathrm{mm}$ and $P = 2.84\,\mathrm{W}$; values which were used by Brindley et al. [3] to generate their Fig. 2a [p. 2041]. Here the ambient temperature T_{a} was assumed to be room temperature, typically 300 K and the radius of the reactive material r_1 was chosen to be 50 mm, as was done for all reported calculations in their work (however, it was stated that phenomena behaviour did not depend on the size of the reactant, since numerical tests were preformed at sizes up to $r_1 = 500\,\mathrm{mm}$ without any quantitative change to the critical boundary). Using the above values gives a maximum temperature at $r = r_0$ of

$$T_{\mathrm{ss}} = 819.8\ \mathrm{K}$$

This does not agree with the value illustrated for $t \to \infty$ in [3, Fig. 2a] which shows at r_0, $T \approx 775\,\mathrm{K}$. A query was forwarded to the authors regarding this and they responded that "the calculations are still with the unsteady program at large times... and will only very slowly reach the proper inert temperature of 819.8 K. The work is still ongoing, both on further numerical approaches (involving voidage for instance), and also analytical ideas.". The test case ($Q = 0$) MoL results, on the other hand, agree more closely with the analytical result of Equation 10. This can be seen in Table 1, for 401 nodes at 10,000 s. Note that the change from 51 to 101 nodes (used in the numerical calculations) produces a marked improvement and that continual increase in the number of nodes refines the accuracy even further. Of course, the greater the number of nodes, the longer the MATLAB process takes to run—thus a compromise between accuracy and speed must be conceded.

This table shows a sensitivity of the result to the number of nodes used: an improvement from 4% error to 0.1% error, where error is calculated via the following:

$$\mathrm{Error} = \frac{\mathrm{actual} - \mathrm{numerical}}{\mathrm{actual}} \times 100\%,$$

where '*actual*' is the steady-state analytical solution for $Q = 0$ and '*numerical*' is the steady-state result given by MoL for $Q = 0$. The accuracy requirement for the number of nodes increases as power does and also as the hot-spot radius decreases, so an error evaluation using the analytical steady-state equation is necessary for all parameter sets of P and r investigated. The number of nodes required for each set to ensure an error of $< 0.5\%$ was always used in our calculations.

4 Numerical results

The MoL in MATLAB provides a graphically understandable platform of results. Figures of both the temperature distribution and the reactant percentage at time intervals versus radius were generated to provide a clear illustration of either a steady-state or ignited material. The MATLAB 'movie' function was implemented for extra clarity and concise representation of the phenomena.

Table 1 The MoL results, using an increasing number of nodes showing the maximum temperature (at r_0) converging to the steady state value of 819.8 K

Time	Nodes			
	51	101	151	201
0	300.000	300.000	300.000	300.000
2000	833.856	812.499	807.442	805.411
4000	848.496	826.669	821.451	819.299
6000	851.558	829.455	824.181	822.006
8000	852.161	829.923	824.673	822.603
10000	852.308	829.999	824.748	822.709
Time	Nodes			
	251	301	351	401
0	300.000	300.000	300.000	300.000
2000	804.503	803.903	803.584	803.366
4000	818.435	817.840	817.389	817.325
6000	821.149	820.436	820.101	820.021
8000	821.639	821.029	820.694	820.505
10000	821.712	821.147	820.802	820.573

In the occurrence of ignition for a certain set of parameters (r, P), an obvious jump in maximum temperature is observed with what can be described as a 'flick-like' action, after which the temperature relaxes seemingly monotonically to what will eventually be a steady state. This can be seen in Fig. 1, an example of one of the many temperature distribution graphs generated. This flick at $t = 2,000$ s, it was suggested, is actually the initiation of a combustion wave. Due to the small radius of the material, this wave is quickly constrained by the $T(r_1, t) = T_a$ condition, and the following relaxation of the temperature to a steady state (much lower than the maximum reached) can be attributed to the diffusion of heat from the power source through the vessel's burnt interior. There is no longer any chemical reaction occurring as all the reactant has been depleted during ignition. This can be seen in Fig. 2, which shows the dependence of the

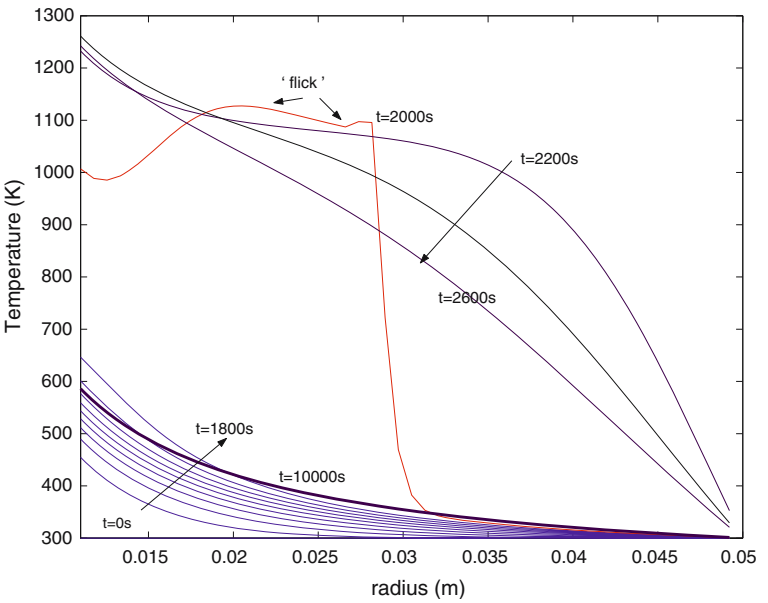

Fig. 1 The temperature distribution at time intervals of 200 s for $r_0 = 0.011$ m, $P = 5$ W, $r_1 = 0.05$ m with 51 nodes

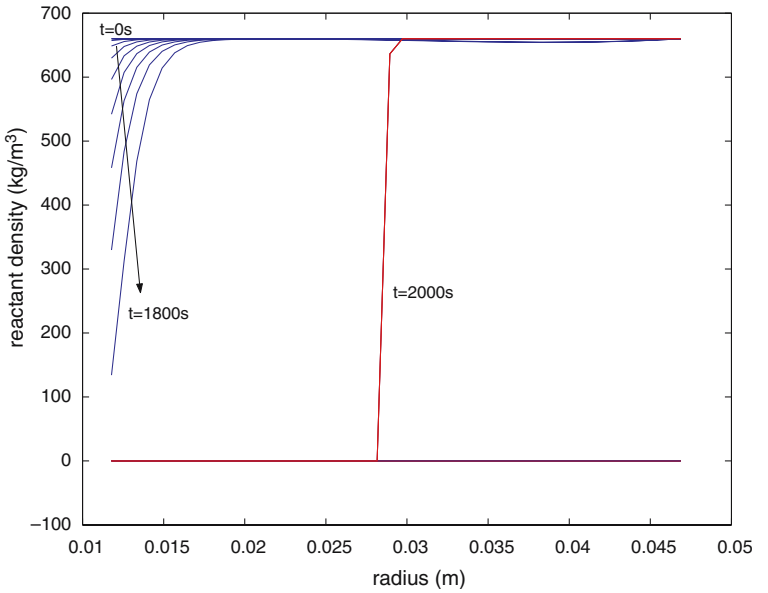

Fig. 2 The dependence of the reactant density to the radial distance. All other parameters and notation are given in Fig. 1

amount of reactant present to the radius at any chosen time. Notice the correlation between Figs. 1 and 2: the material becomes wholly depleted at the same radius and time that we see the ignition flick.

If the radius of the spherical material is increased, it is found that this does not affect whether or not ignition occurs (as long as substantially more nodes are used to retain accuracy). What this increase in radius does achieve, however, is visual confirmation of a travelling-wave front propagating through the medium. Figure 3, which is generated using the same hot-spot radius and power as Figs. 1 and 2, shows the 'initial flick' occurring at $t = 2,100$ s. From there, the maximum temperature of the material does not

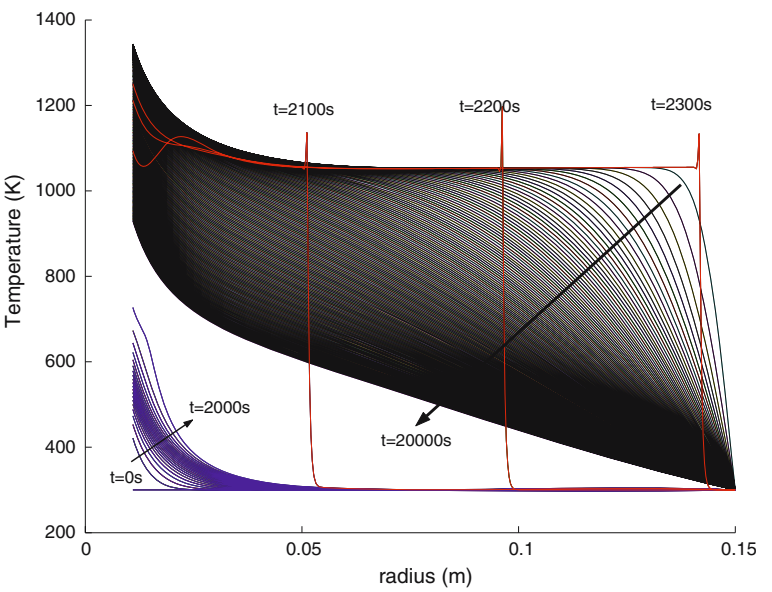

Fig. 3 The temperature distribution at time intervals of 100 s for $r_0 = 0.011$ m, $P = 5$ W, $r_1 = 0.15$ m with 451 nodes of 100 s

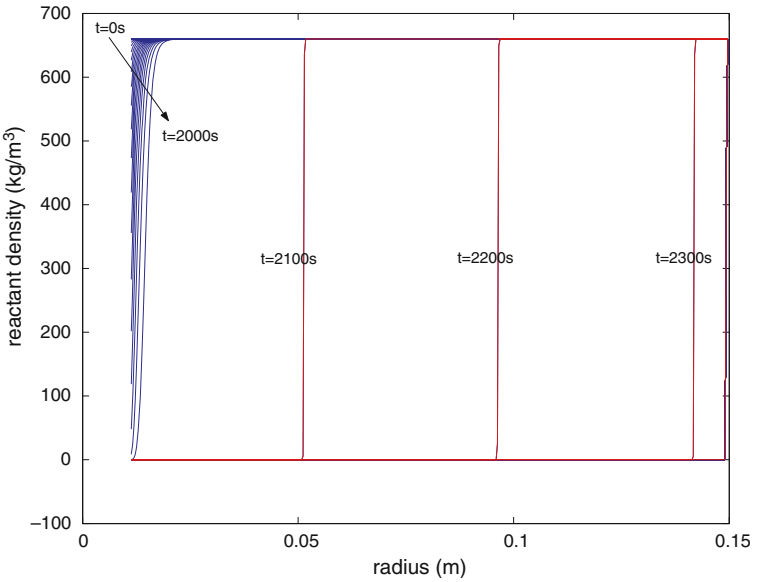

Fig. 4 Reactant density distribution. All notations and parameters are the same as those used in Fig. 3

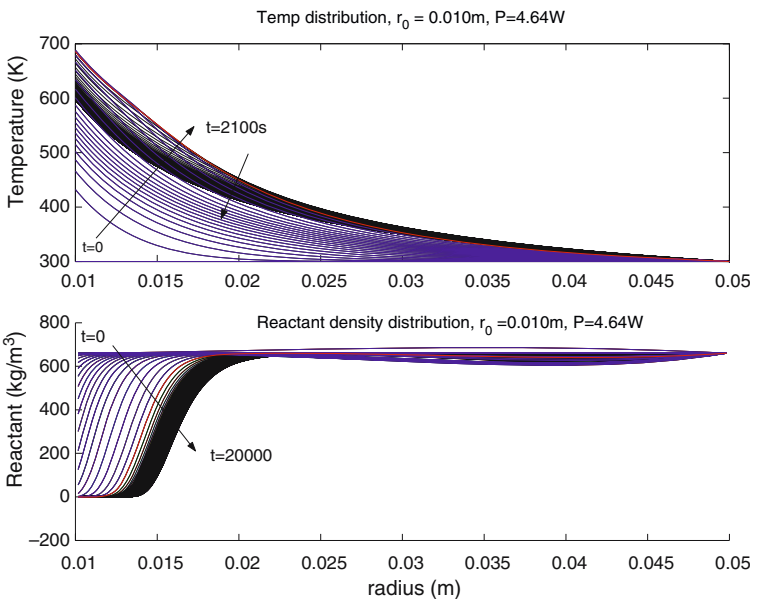

Fig. 5 Temperature and reactant-density distribution in a steady-state system. Here $r_0 = 0.01$ m and $P = 4.64$ W

quickly relax as represented in Fig. 1, but instead the combustion wave moves through the material in time. As this is happening, the reactant material (shown in Fig. 4) is consumed in a corresponding fashion–that is, wholly depleted up to the radius the combustion wave has reached in an amount of time. For example, in Fig. 3 it can be seen that at time $t = 2,200$ s, the combustion wave has travelled to a radius of ≈ 0.1 m. If we consider Fig. 4, it can be determined that at $t = 2,200$ s, the reactant density is 0 (i.e., fully consumed) up to a radius of ≈ 0.1 m, and unchanged (i.e., equal to initial concentration) beyond this point. In this case ($r_0 = 0.11$ m, $P = 5$ W) therefore, it is easy to determine that the material has undergone the process of thermal ignition.

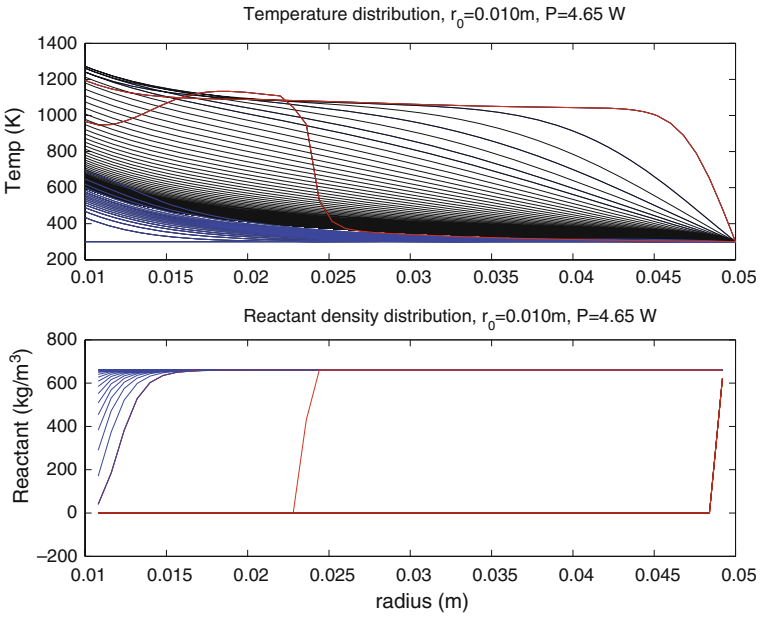

Fig. 6 Temperature and reactant distribution in an ignited system. Here $r_0 = 0.01\,\text{m}$ and $P = 4.65\,\text{W}$

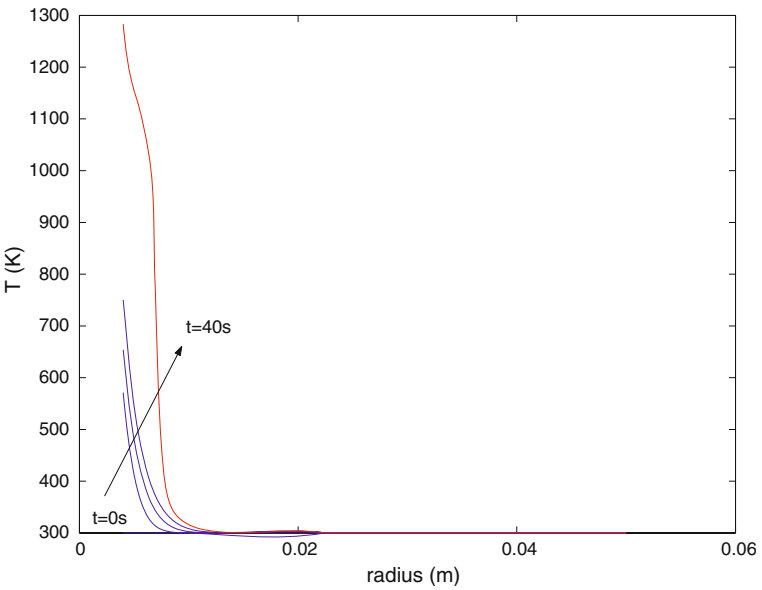

Fig. 7 Temperature distribution over 40 s for $r_0 = 0.004\,\text{m}$, $P = 4.5\,\text{W}$, $r_1 = 0.05\,\text{m}$ with 1001 nodes used in the numerical scheme

We now turn our attention to investigate the unexpected result presented in [3] which we reproduce later in our Fig. 16. To do this, a radius lying within the range exhibiting an apparently 'safe' region between two distinct critical values for the power is chosen for investigation. This value is chosen as $r_0 = 0.004\,\text{m}$, as it lies on many of the points tested by Brindley et al. [3]. The 'control' chosen is $r_0 = 0.010$, used to provide agreement between the MoL results and those given in the paper for a more accepted result—results gained in the questionable region using the MoL will thus be more credible. A value $r_0 = 0.010$ is chosen because

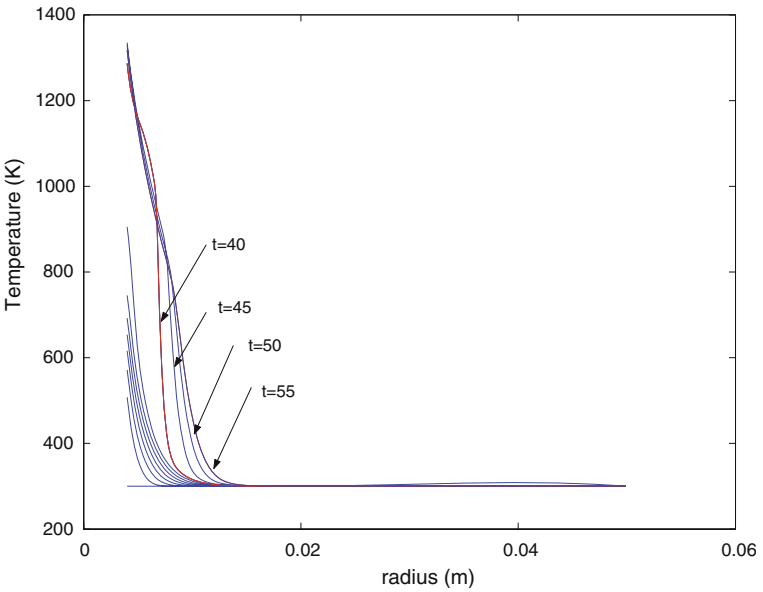

Fig. 8 Temperature distribution over 55 s. Parameter values are the same as those in Fig. 7

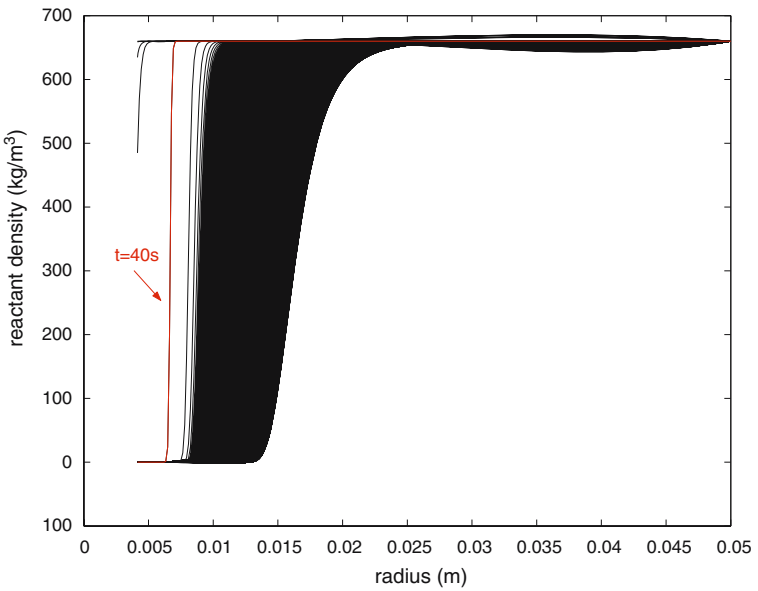

Fig. 9 Reactant-density distribution over 20,000 s for parameter values as those used in Figs. 7 and 8

it lies well within the range of values investigated in [3], but is on the lower end of the 'nodes needed for accuracy' scale.

Considering the control case first ($r_0 = 0.010$ m) through pairs of temperature and density plots given as Figs. 5 and 6, it is easily determined via visual investigations that ignition takes place somewhere between $P = 4.64$ and 4.65 W. Since it takes considerable time to obtain these results accurate to 2 decimal places, ignition will be said to occur at the higher bound, 4.65 W, instead of examining to an increased number of decimal places (the higher of two values given to 2 decimal places between which ignition is seen to take place will be continually regarded as the point of ignition throughout the remainder of this paper, for consistency).

In the investigated case ($r_0 = 0.0004\,\text{m}$), however, it is not so easy to determine the value of the critical power that leads to ignition. As can be seen in Figs. 7 and 8, there is a suspicious 'flick' similar to, but not as pronounced as, that seen previously in Fig. 1. This flick, however, does not give rise to a combustion wave, although it involves a large jump in temperature between times $t = 30\,\text{s}$ and $40\,\text{s}$ followed by relaxation; it does not display a sudden complete reactant consumption throughout the entire material at any time up to 20,000 s (Fig. 9). Intuitively, due to the incomplete reaction consumption, one would assume no ignition to occur. However, what about the surprisingly large temperature increase? Are there other values of P that are even more difficult to quantify visually as ignition or steady-state scenarios? To eliminate any doubt, a sensitivity analysis as described by Nelson and Sidhu [6] is undertaken to determine the critical values of P.

4.1 Sensitivity analysis

'In classical thermal-explosion theory reactant consumption is ignored. The difference between subcritical (slow-reaction) and supercritical (combustion) behaviour is then well defined. When reactant consumption occurs, this demarcation disappears. . .historically a number of authors have addressed the issue of how to define criticality in such problems. . .in the 1980's it was realised that the most satisfactory method is to identify criticality using sensitivity analysis" [6].

The idea of sensitivity analysis is based on taking an experimental measurement m chosen to describe the system, and together with a variable parameter ϕ find the *absolute sensitivity* of the measurement to changes in the parameter. The method has the advantages of applicability to any measurable parameter, and ease of use. This absolute sensitivity is defined by

$$s(m, \phi) = \left| \frac{\mathrm{d}m}{\mathrm{d}\phi} \right|. \tag{11}$$

Criticality is defined as the value of the parameter ϕ at which the absolute sensitivity takes a maximum value, i.e., the value of the parameter ϕ at which the measurement m is most sensitive to changes in ϕ. This can be considered as the value of ϕ at which the derivative is greatest, hence the use of the derivative in the definition.

For the hot-spot model, the measurement characterising the system is chosen to be ρ_{total}. This is the value of the integral of the normalised (i.e., $\times \frac{1}{600}$) reactant density *spline* (the curve fitted to the data set of nodal positions and final densities) at $t = 20,000\,\text{s}$, from node 2 to node $(n-2)$, where n is the number of nodes used in the lattice (determined by the parameters P and r_0 via accuracy considerations).

Density was chosen as the system measurement rather than temperature as it holds greater meaning for criticality: although the temperature may rise to a great height, this is not always an indicator of ignition, whereas the situation of wholly depleted reactant at any stage indicates with much greater confidence that ignition has occurred. Power (P) is the varying parameter for which ρ_{total} is calculated.

At each node (the x-values), the normalised values of 'Xdata' (local reactant density ranging from 0 to 1) is calculated over time, and the value at the final time ($t = 20,000\,\text{s}$) forms the set of corresponding y-values. The resulting data set of x- and y-values is fitted using a cubic spline which is then integrated to find the average density ρ_{total} for that value of P, using the approach from [8]. This is carried out for fixed values of both $r_0 = 0.010\,\text{m}$ (our control case) and $r_0 = 0.004\,\text{m}$ (case under investigation), for values of $P = 2.5$ to 6 in increments of 0.5. This process is repeated in the regions where a large change can be seen from one P-value to the next, such as 5.2 to 5.3 as seen in Fig. 10. The increments become smaller in these regions (0.1 and then 0.01) so that the point at which the change in ρ_{total} occurs can be found to lie somewhere within two values accurate to two decimal places. Intuitively one would expect this to be the point at which conditions become critical for ignition. (The values found for ρ_{total} and P for the control and investigation cases are plotted and given in Figs. 10 and 11).

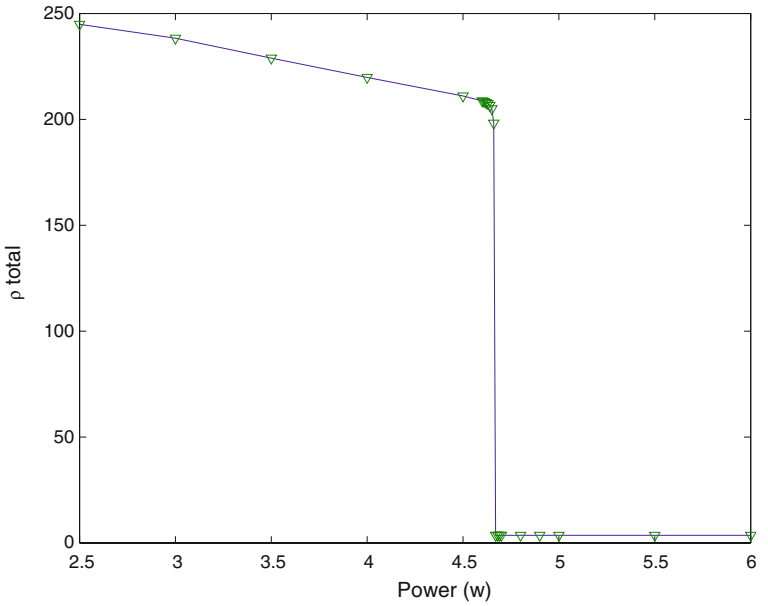

Fig. 10 Input data for the sensitivity analysis: control case, $r_0 = 0.010\,\text{m}$ and $r_1 = 0.05\,\text{m}$ with 251 nodes

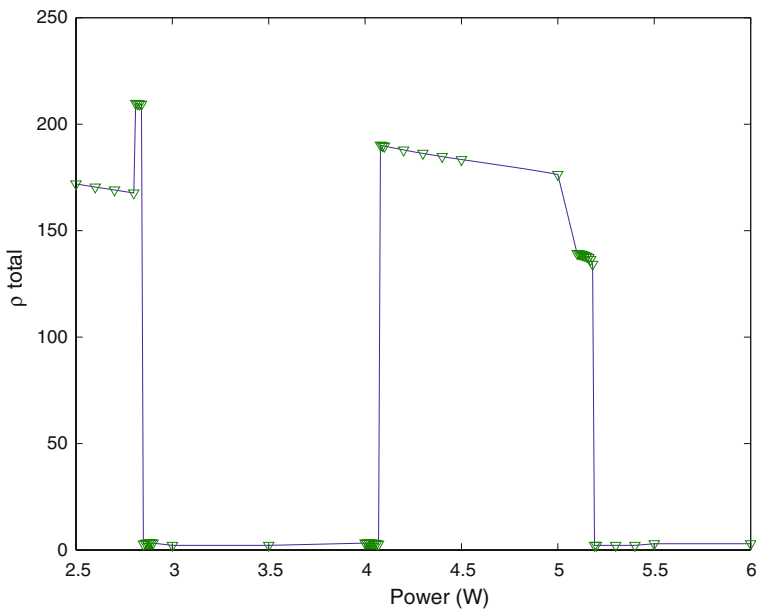

Fig. 11 Input data for the sensitivity analysis: investigation case, $r_0 = 0.004\,\text{m}$ and $r_1 = 0.05\,\text{m}$ with 251 nodes

This expectation is verified by the sensitivity analysis. To apply the analysis we follow Nelson and Sidhu [6] and create a separate data file containing the values of ρ_{total} for corresponding values of P. We calculate the absolute value of the derivative of ρ_{total} with respect to P, and plot these against P as shown in Figs. 12 and 13 for $r_0 = 0.010\,\text{m}$ and $r_0 = 0.004\,\text{m}$, respectively. In all cases here we use $r_1 = 0.05\,\text{m}$.

Considering firstly Fig. 12 for the $r_0 = 0.010\,\text{m}$ case, we observe that it illustrates the expected behaviour, namely a safe region of lower P-values extending to a critical value of P above which the material ignites. This value of P is clearly illustrated by the obvious 'spike', indicating the point at which the derivative

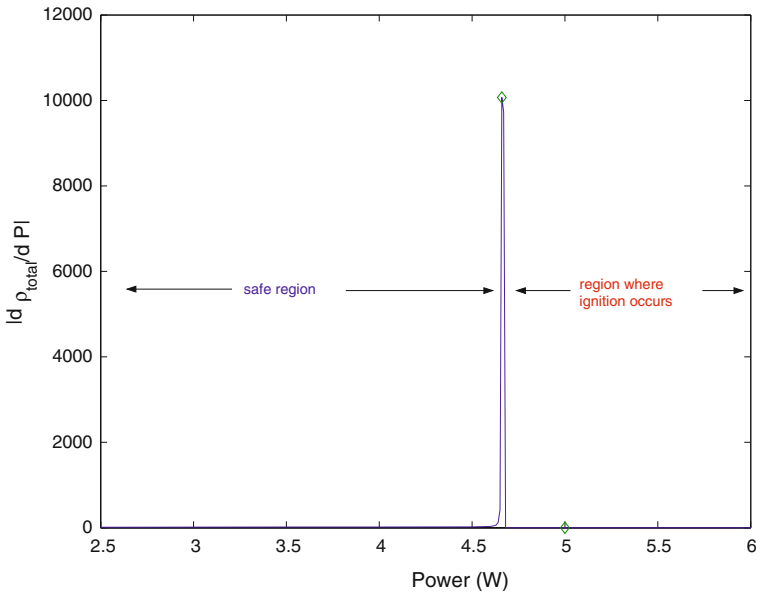

Fig. 12 Sensitivity-analysis result for the control case with $r_0 = 0.01$ m and $r_1 = 0.05$ m (absolute value of the first derivative over a non-uniform grid)

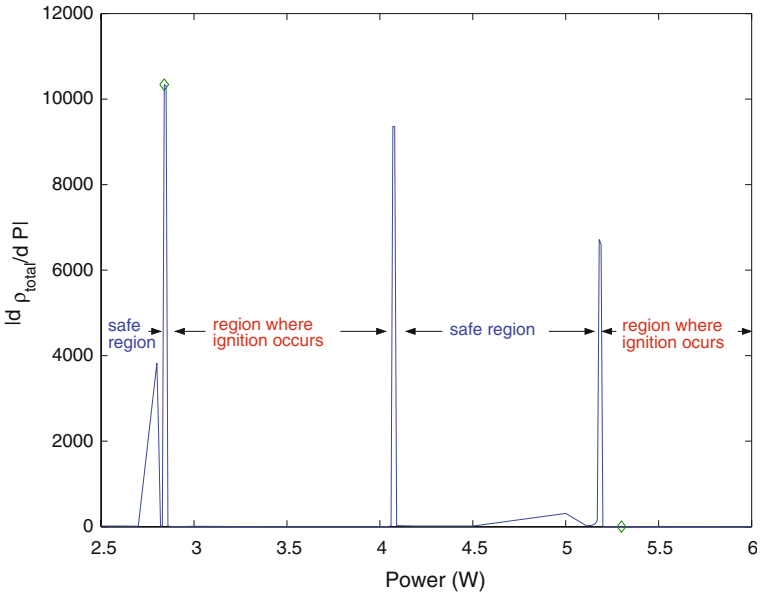

Fig. 13 Sensitivity analysis for the investigation case with $r_0 = 0.004$ m and $r_1 = 0.05$ m

$\left|\frac{\mathrm{d}m}{\mathrm{d}\phi}\right| = \left|\frac{\mathrm{d}\rho_{\mathrm{total}}}{\mathrm{d}P}\right|$ is the greatest. This agrees with the visual deduction made in maximum temperature versus radius graphs, since Figs. 5, 6 show the first occurrence of the initiation of a combustion wave to lie within $P = 4.64$ W and $P = 4.65$ W, as stated earlier.

If we now consider Fig. 13 for the $r_0 = 0.004$ m case, a very different situation is illustrated. What can be seen is the existence of a number of critical points for $P \approx 2.8$ W, ≈ 4.1 W and ≈ 5.3 W, implying a system transition from a safe region to an unsafe (ignition occurring) as one would expect, but then moving again into an apparently safe region and again into an unsafe one.

4.2 Accuracy improvement

It is noticeable in Fig. 13 that there exists some possible 'noise' in the data. In the ranges $P \approx 2.7$–2.8 W and $P \approx 4.5$–5.1 W there are regions where the first derivative is shown to be much larger than zero (up to 4,000 in the first region and 400 in the second), but comparatively small to the largest 'spikes' deemed to be the critical values of P. It was not known initially why these regions exist, and there is no instance of any such regions occurring in the control case. To ensure that the cubic-spline approximation does not introduce any computational artefacts, the number of nodes used was raised from 251 to 501 (since, as seen previously, the number of nodes can significantly affect the accuracy of the result). This increase in the number of nodes makes a large difference to the sensitivity-analysis outcome as seen in Figs. 14 and 15. These results are greatly improved as the mysterious discrepancies have disappeared and indicate that our hypothesized regions of noise simply were that: incorrect data representation that could be attributed to accuracy limitations.

By repeatedly using the MoL and sensitivity-analysis techniques described above, a graph of critical conditions for ignition for r_0 versus P is generated for the range 2.5–6 W. This is displayed in Fig. 16. This curve is a close representation of the figure given in [3], which was the initial point of interest due to the unexpected result it illustrates. By using a method totally independent to that used by Brindley et al. [3], and generating a near-replica of this graph, their results are thus corroborated—although counter-intuitive, they appear to be correct and the critical behaviour has been independently confirmed through the use of sensitivity analysis.

5 Discussion

By applying the MoL and sensitivity analysis to a model representing a spherical power source centrally located within a concentric sphere of combustible material, it has been possible to replicate the novel results reported in [3]. From these results it is shown that there exists a range of r_0 over which a number of

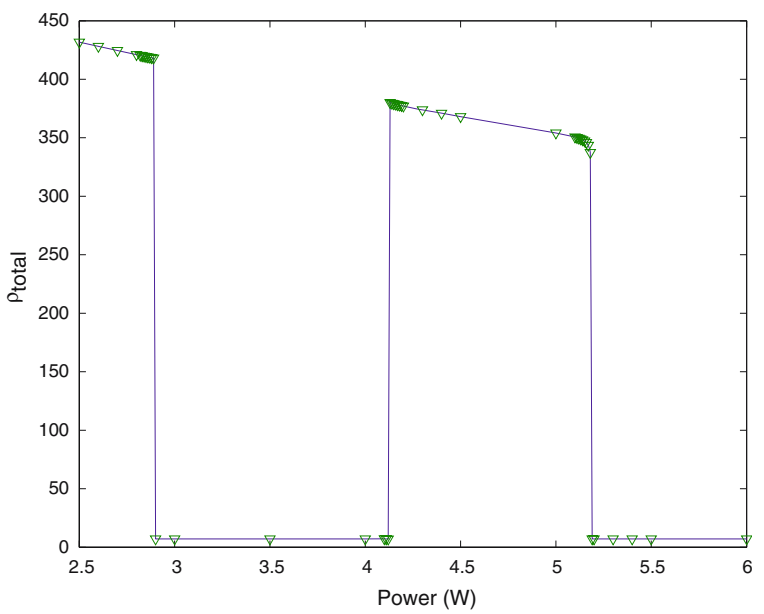

Fig. 14 Improved input sensitivity for the investigation case for $r_0 = 0.004$ m and $r_1 = 0.05$ m with 501 nodes

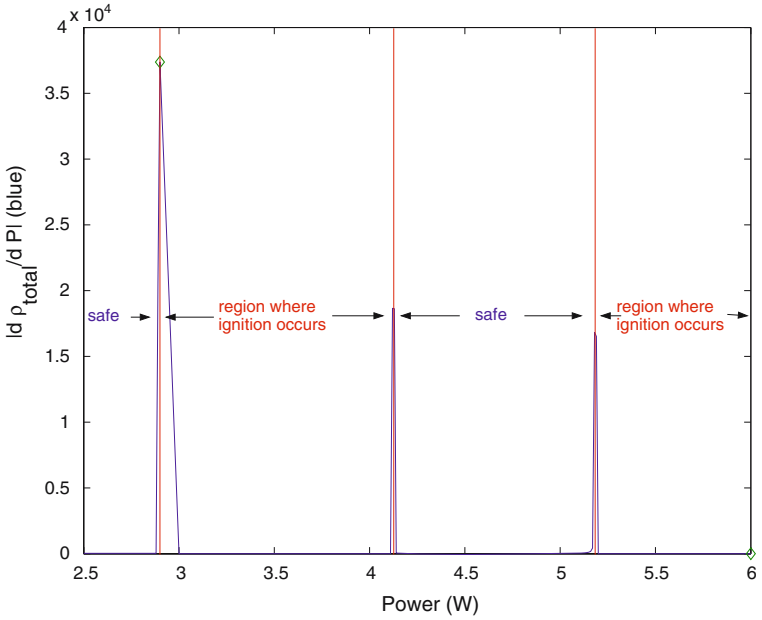

Fig. 15 Improved sensitivity-analysis result for the investigation case $r_0 = 0.004\,\text{m}$ and $r_1 = 0.05\,\text{m}$ with 501 nodes

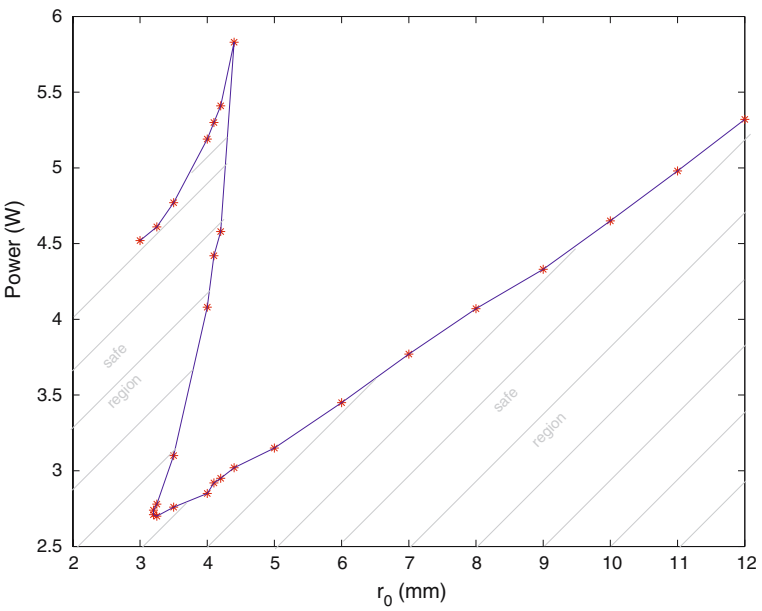

Fig. 16 Critical power values for varying hot-spot radii and and $r_1 = 0.05\,\text{m}$

distinct critical values for P exist, with an unexpected safe region present between regions where ignition would usually be thought to occur.

It is to be noted that this situation does not arise if reactant consumption is ignored. Results of a brief investigation showed that in the range $P = 2$–$6\,\text{W}$, for a hot-spot size of $0.004\,\text{m}$, the critical power for ignition (occurring between 3.5 and 3.75) lies between the first two critical values of P found in the reactant consumption case ($P = 2.85$ and $4.08\,\text{W}$). Suppression of ignition is not seen in the range $P = 2$–$6\,\text{W}$ (investigated in $0.25\,\text{W}$ increments). This implies that the reason for the behaviour depends on the state of

the reactant; this is confirmed in the authors' later work [4], discussed below. Additionally, this investigation shows that, for application to industrial practices, the inclusion of reactant consumption is crucial.

The unexpected result, as the authors state, is "of particular concern" [3, p. 2040], implying that "over a specific range, growth in size of the source, keeping total power constant, causes a transition from safe operation to an unsafe condition at which the total destruction [i.e., spontaneous thermal ignition of] is the consequence". For example, if choosing $P = 4$ W, one can see that this is well within safe limits (shaded area of Fig. 16) for $r_0 \approx 2$–4 mm. Then suddenly, at $r_0 = 4$ mm, the parameters lie within an unsafe region. The expected result would be that, if the radius for a corresponding power lies below the critical condition, it would remain so for increasing values of r_0, as is the case for $r_0 = 0.005$ m onwards. We note that the phenomenon of reducing a control parameter to make something safer and then finding out that this has not happened does occur in other combustion situations as well (see [9–11] for some examples).

The result poses potential problems in practice, as described by Brindley et al. [3]. These include problems such as, (for example) those arising in a rotary drier where there is a buildup of inert residues from the degradation of material around a hot bearing (thus increasing r_0, radius of the hot spot) or alternatively, a buildup of residue at a local site in a damaged power cable passing through a potentially reactive particulate substance of appropriate exothermicity. If a system containing a fixed 'hot spot' is considered, i.e., a source of constant power at a site of specific size and a problem is encountered, the natural inclination to suppress the hazard may be to lower the power. If the original power of perceived problem was, for example, $P = 4.5$ W, lowering the power to below 4 W would lead the parameter set into an unsafe region, with the hazardous consequence of the onset of ignition.

In [4] a possible physical explanation for the finding that ignition behaviour is suppressed for small-radius hot spots is presented, based on the developing inert temperature profiles upon which the effect of the exothermic reaction is superimposed. In this work it is shown qualitatively that the unexpected suppression of ignition for small radius hot spots is

> *"due to the competition between the sufficiently fast burning of an annulus of a reactant near the surface of the hot spot to match that required for self-propagation, and sustaining such a high temperature away from the surface through a temperature profile which is decaying through diffusion. The subtle balance between these two effects must be maintained in order to establish a propagating combustion front".*

From this statement it can be concluded that, for the parameter range $r_0 \approx 3.2$–4.5 mm, the required equilibrium between the fast burning of the reactant near the hot-spot surface and the maintenance of a high temperature away from the surface is not obtained, and hence the system 'falls out' of the expected-behaviour regime. It would be interesting to see this result experimentally confirmed, thus highlighting the urgent need to avoid the accidental ignition of reactive material that can easily accumulate near a hot-spot surface in the process engineering industry. [Nelson, as quoted in [4]].

6 Conclusion

In this paper we have considered a spherical, reactive hotspot problem where the critical conditions for thermal ignition cannot be determined analytically. This problem is a potentially significant source of unwanted ignition in many manufacturing and storage industries where localized heat sources impinge upon organic material. We have reported in detail on our computational approach, using the MoL to verify the unexpected behaviour for small-sized power sources (in which, over a range of source radii, there can be two distinct critical values for the power, with a seemingly safe region occupying some of the range between them). We have provided extensive results to clearly demonstrate the accuracy of the MoL, provided sufficient nodes are included. Furthermore, we have used a sensitivity analysis to provide a rigorous definition of criticality in a system with reactant consumption. This definition relies on finding

the maximum rate of change of a system measurable with respect to a system parameter. In the present case, we found that the integral of the reactant density (as a measure of total system reactivity) should be examined as a function of applied heating power and that this permits extremely clear identification of criticality. As well as emphasising that the previously obtained results can be independently verified, we also reiterate that these results can have serious safety implications for chemical-plant operations and also suggest that this would not be a suitably reliable method for the detonation of cluster bombs.

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