On the Critical Behaviour of Exothermic Explosions in Class A Geometries

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Abstract

The aim of this work is to apply the homotopy perturbation method for solving the steady state equations of the exothermic decomposition of a combustible material obeying Arrhenius, Bimolecular and Sensitised laws of reaction rates. These equations are formulated on some Class A geometries (an infinite cylinder, an infinite slab and a sphere). We also investigate the effect of Frank-Kamenetskii parameter on bifurcation and thermal criticality by means of the Domb-Sykes graphical method.

Keywords: Thermal criticality, chemical kinetics, Homotopy perturbation method, Domb Sykes method.

Nomenclature

a	geometry half width
A	rate constant
C_0	initial concentration of the reactant
E	activation energy
h	Planck's number
j	geometry factor
k	thermal conductivity of the material
K	Boltzmann's constant
$\mathcal{L}$	linear operator
m	numerical exponent
p	embedding parameter
r	radial distance
$\bar{r}$	dimensionless radial distance
Q	heat of reaction
R	universal gas constant
T	absolute temperature
T_0	wall temperature

Greek symbols

ν	vibration frequency
θ	dimensionless temperature
ε	dimensionless activation energy
δ	Frank-Kamenetskii parameter
γ	exponent

Subscripts

cr	critical
max	maximum

1. Introduction

The safety in transport and storage of combustible materials is a key issue in pyrotechnic applications. These materials are often subjected to self-ignition. This internal heating occurs when an explosive substance is brought to a sufficient temperature so that the process of decomposition begins to produce significant exothermic effects. This involves a thermal runaway phenomenon accompanied with an increase of the temperature producing a rapid thermal decomposition. The understanding of the factors that control this phenomenon is of fundamental importance in many industrial processes.

This phenomenon was first introduced in the **1930s** by Semonov, Zeldovith and Frank-Kamentskii and their pioneering contributions were summarized in [1]. Furthermore, Frank-Kamentskii developed the steady-state theory of thermal explosion. In this theory, the Frank–Kamenetskii approximation allows **us** to determine the critical values, which constitute limit values not to be exceeded to avoid the phenomenon of self-ignition. Some studies deal with a chain of several reactions in this phenomenon of self-ignition. Several works in the literature have applied this theory **to** different combustible materials geometries. Boddington et *al.* [2, 3, 4] have considered the special case of two-step parallel exothermic reactions for the infinite slab where solutions by quadrature were possible. Graham-Eagle and Wake [5] considered a system of simultaneous exothermic reactions. They extended the investigations of Boddington et *al*. **[2, 3, 4]** to the other two geometries of the infinite circular cylinder and the sphere. A variational method was used to evaluate the critical values of Frank-Kamentskii parameter and the maximum temperature. The same authors [6] extended the treatment of simultaneous reactions to the case where one reaction is exothermic and the other endothermic, leading to the phenomenon of the disappearance of criticality or transition, which can also happen with a single reaction in the case of very low activation energy. In this case, the Frank–Kamenetskii approximation is no longer valid. They used a variational method **[7]** to determine

numerically the values of the parameter which characterizes the transition in the parameter space.

Recently Ajadi and Gold'shtein [8] employed a three-step reaction kinetics model (initiation, propagation and termination steps). The calculation of the criticality was made by means of a variational method for (infinite slab, infinite cylinder, and sphere) under Arrhenuis laws of reaction rates by using an effective activation energy approximation. Balakrishnan *et al.* [9] calculated the critical values for some non Class A geometries (infinite square rod and cube). Their critical values were found using the finite difference method.

In applied mathematics or engineering problems, numerical methods commonly used such as finite difference, finite element or characteristics method, need large size of computational works due to discretization and usually the effect of round-off error causes loss of accuracy in the results. In addition to this drawback, these methods with limited precision include slow runtimes, numerical instabilities and difficulties in handling redundant constraints.

Analytical traditional methods commonly used for solving these problems can be very useful, especially for the calibration of numerical calculations. Among these approaches, the classical perturbation method is based on the existence of small parameters but the overwhelming majority of linear and nonlinear problems have no small parameters at all.

To overcome this shortcoming, the homotopy perturbation method (HPM) was first introduced by He [10-13]. The main idea of this method is to introduce an embedding parameter $p \in [0,1]$ to construct a homotopy and give solutions of the deformed problem as power series in p . When $p = 0$ the system of equations is reduced to a simplified form which admits exact solutions and at $p = 1$, the system takes its original form and gives the desired solutions. This method has been extensively employed by numerous authors to solve a large

variety of linear and nonlinear problems. We can cite the works [14-17], the list is not exhaustive.

In this paper, we examine the steady-state solutions for the strongly exothermic decomposition of a combustible material of a symmetric class *A* geometries, uniformly heating, under Arrhenius, Biomolecular and Sensitised kinetics, neglecting the consumption of the material.

The contribution of the present work is twofold. First, we calculate the temperature field using the HPM in a symbolic computational language. The second aim is to study the thermal criticality conditions of the problem.

Critical values for different geometries are found by using the Domb- Sykes technique [18] as a useful tool to extract singularities and to perform analytic continuation. This method has shown to be very accurate and simpler to implement, when compared with variational method [6,8] or Hermite-Padé approximants method [19-21].

The structure of this paper is as follows. Section 2 presents the boundary value problem governing the ignition of a viscous combustible material for symmetric class *A* geometries. In section 3, we will apply the homotopy perturbation method to this nonlinear boundary value problem. The fourth section is assigned to a brief description of the Domb and Sykes method and its application to calculate the bifurcation points. Section 5 is devoted to the results obtained by the proposed method and comparison with other works will be performed. Conclusions will appear in Section 6.

2. Mathematical model

We consider the steady-state solutions for the strongly exothermic decomposition of a viscous combustible material. Neglecting the reactant consumption, the equation for the temperature $T(r)$ may be written in terms of physical variables together with the boundary conditions, as:

$$\frac{d^2T}{dr^2}+\frac{j}{r}\frac{dT}{dr}+\frac{QC_0A}{k}\left(\frac{KT}{\upsilon h}\right)^m e^{-\frac{E}{RT}}=0 \tag{1}$$

$$\frac{dT}{dr}(0)=0\,,\quad T(a)=T_0 \tag{2}$$

with T the absolute temperature, T_0 the wall temperature, k the thermal conductivity of the material, Q the heat of reaction, A the rate constant, E the activation energy, R the universal gas constant, C_0 the initial concentration of the reactant species, h the Planck's number, K the Boltzmann's constant, υ the vibration frequency, a the geometry half width and r the radial distance in the normal direction. m is the numerical exponent, such that $m=-2, 0, \frac{1}{2}$ represent numerical exponent for Sensitised Arrhenius, and Bimolecular kinetics (Boddington *et al*. [3], Makinde [20], Bowes [22], Bebernes and Eberly [23], Okoya [24]) and j = 0, 1, 2 is the geometry factor representing respectively Slab, Cylindrical and Spherical geometries.

The following dimensionless variables and parameters are introduced in Eq. (1):

$$\theta=\frac{E(T-T_0)}{RT_0^2}\quad,\quad \bar{r}=\frac{r}{a},\quad \varepsilon=\frac{RT_0}{E}\,,\qquad \delta=\frac{QEAa^2C_0K^mT_0^{m-2}}{\upsilon^m h^m Rk}e^{-\frac{E}{RT_0}}$$

to obtain the dimensionless governing equation together with the corresponding boundary conditions. We get the following equations:

$$\frac{d^2\theta}{d\bar{r}^2}+\frac{j}{\bar{r}}\frac{d\theta}{d\bar{r}}+\delta(1+\varepsilon\theta)^m\exp\left(\frac{\theta}{1+\varepsilon\theta}\right)=0 \tag{3}$$

$$\frac{d\theta}{d\bar{r}}(0)=0\,,\ \theta(1)=0 \tag{4}$$

where δ, ε represent the Frank Kamenetskii and activation energy parameters respectively. Hereafter we will suppress the bar symbol for clarity.

3. Homotopy perturbation method solution

In this section, we will apply the homotopy perturbation method (HPM) to nonlinear ordinary differential equation (3). According to this method, we can construct a homotopy as follows:

$$(1-p)\mathcal{L}(\phi)+p\left(\frac{d^2\phi}{dr^2}+\frac{j}{r}\frac{d\phi}{dr}+\delta(1+\varepsilon\phi)^m\exp\left(\frac{\phi}{1+\varepsilon\phi}\right)\right)=0 \tag{5}$$

where

$$\mathcal{L}(\phi)=\frac{d^2\phi}{dr^2}+\frac{j}{r}\frac{d\phi}{dr} \tag{6}$$

Applying the perturbation technique [25], we can assume that the solution of Eq. (5) can be expressed as a series in *p*

$$\phi(r,p)=f_0(r)+pf_1(r)+p^2f_2(r)+... \tag{7}$$

where $p \in [0,1]$ is an embedding parameter. When $p = 0$, we can obtain the initial guesses; when $p = 1$, Eq. (5) turns out to be the original one.

Setting $p = 1$, we obtain an approximate solution of (3):

$$\theta=\lim_{p\to 1}\phi(r,p)=f_0(r)+f_1(r)+f_2(r)+... \tag{8}$$

One has to substitute relation (7) into the governing equation (5), collect the powers of p and obtain a sequence of differential equations and boundary conditions. The solution for the temperature field for Sensitised, Arrhenius and Bimolecular reaction rates for different geometries are given as

$$\phi(r,p,j=0)=-\frac{1}{2}p\delta\left(r^2-1\right)+\frac{1}{24}p^2\delta^2\left(r^2-1\right)\left(r^2-5\right)(m\varepsilon+1)+O\left(p^3\right) \tag{9}$$

$$\phi(r,p,j=1)=-\frac{1}{4}p\delta\left(r^2-1\right)+\frac{1}{64}p^2\delta^2\left(r^2-1\right)\left(r^2-3\right)(m\varepsilon+1)+O\left(p^3\right) \tag{10}$$

$$\phi(r,p,j=2)=-\frac{1}{6}p\delta\left(r^2-1\right)+\frac{1}{360}p^2\delta^2\left(r^2-1\right)\left(3r^2-7\right)(m\varepsilon+1)+O\left(p^3\right) \tag{11}$$

According to Eq. (8) we get the solution for the temperature field for the three reaction rates.

4. Bifurcation study by Domb Sykes method

The modelling of physical phenomena often results in nonlinear problems for some unknown function, say $f(\lambda)$ depending on a parameter λ. Usually, the problems cannot be solved exactly. The solutions of these nonlinear systems are dominated by their singularities which must be real and positive in order to have a physical sense.

We suppose that up to the point λ_0, the solution is analytic in the interval]- λ_0 , λ_0 [, then one can solve the problem by expanding the solution in a power series

$$f(\lambda) = \sum_{n=0}^{\infty} c_n \lambda^n \tag{12}$$

If an infinite number of c_n is known, the radius of convergence, which is the distance from the origin to the nearest singularity, limiting the range of validity of the series (12), can be calculated using D'Alembert's ratio.

However, for most nonlinear problems it is rare to find an unlimited number of terms of the power series (12). The nearest singularity cannot therefore be determined precisely.

Using symbolic calculus codes, it is now possible to calculate a sufficient number of terms in the series, to study precisely the solution, and there exist a variety of methods devised for extracting the required information of the singularities from a finite number of series coefficients. The most frequently used methods are the ratio-like methods, such as the Domb-Sykes method [26, 27], Neville-Aitken extrapolation [26], and semi numerical approximant methods, such as Padé approximants [28] or Hermite- Padé approximants [19-21].

Herein we are concerned with the bifurcation analysis by analytic continuation as well as with the dominant behavior of the solution by using partial sum (7). This may be done calculating the nearest singularity to the origin. A useful tool for extracting this singularity is the Domb-Sykes method. This technique is easier to implement than others and when it is improved, it

gives a precise value of the singularity. This method, successfully applied in various problems [29-32], is presented here.

According to Fuchs (Bender and Orszag [27]), there are two possible forms:

$$f(\lambda)=\sum_{n=0}^{\infty}c_n\lambda^n=\begin{cases}\alpha(\lambda_0-\lambda)^{\gamma} & \text{for } \gamma\neq 0,1,\cdots,n,\ldots\\ \alpha(\lambda_0-\lambda)^{\gamma}\ln(\lambda_0-\lambda) & \text{for } \gamma=0,1,\cdots,n,\ldots\end{cases} \tag{13}$$

The radius of convergence λ_0 of the series (12) may be found by d'Alembert ratio:

$$\lambda_0=\lim_{n\to\infty}\left|\frac{c_{n-1}}{c_n}\right| \tag{14}$$

Of course only a finite number of coefficients c_n are known, so that it is difficult to obtain precisely the limit. Domb and Sykes have suggested that the inverse ratio $\left|\frac{c_n}{c_{n-1}}\right|$ has the following expansion:

$$a_n=\left|\frac{c_n}{c_{n-1}}\right|=\frac{1}{\lambda_0}\left[1-\frac{1+\gamma}{n}\right]+o\left(\frac{1}{n}\right)=h(1/n) \tag{15}$$

Domb and Sykes have pointed out that it is more reliable to plot $\left|\frac{c_n}{c_{n-1}}\right|$ versus 1/*n*, i.e. to bring $n\to\infty$ to the origin, rather than plotting $\left|\frac{c_{n-1}}{c_n}\right|$ versus *n*. The plot of $\left|\frac{c_n}{c_{n-1}}\right|$ versus 1/*n* is known as Domb-Sykes plot. In this plot, the intersection of the straight line $a_n=h(1/n)$ with the axis 1/*n* = 0 is exactly $\frac{1}{\lambda_0}$ and the slope of this line gives the exponent γ. Unfortunately, a_n is often a slowly converging series. A good way to improve the convergence is to use the Richardson extrapolation (Bender and Orszag [27]), which is appropriate for this kind of sequences and can be defined as follows.

If a_n can be written in the following form: $a_n = S_0 + \frac{S_1}{n} + \frac{S_2}{n^2} + \ldots$, then the Richardson extrapolation is:

$$s_{mn} = \sum_{k=0}^{m} \frac{(-1)^{k+m} a_{n+k} (n+k)^m}{k!(m-k)!} \tag{16}$$

This new sequence has a quicker convergence to the limit S_0.

5. Results and discussion

Using a computer algebra system, we obtained the first 30 terms of the solutions series (9)-(11).

In order to verify numerically whether the proposed methodology leads to high accuracy, we evaluate the numerical solutions using a collocation method proposed by Shampine *et al*. [33].

In Figures1-3 below, we have plotted the numerical and the HPM solutions in all class A geometries and for all numerical exponent *m*. These figures show a perfect agreement of both solutions.

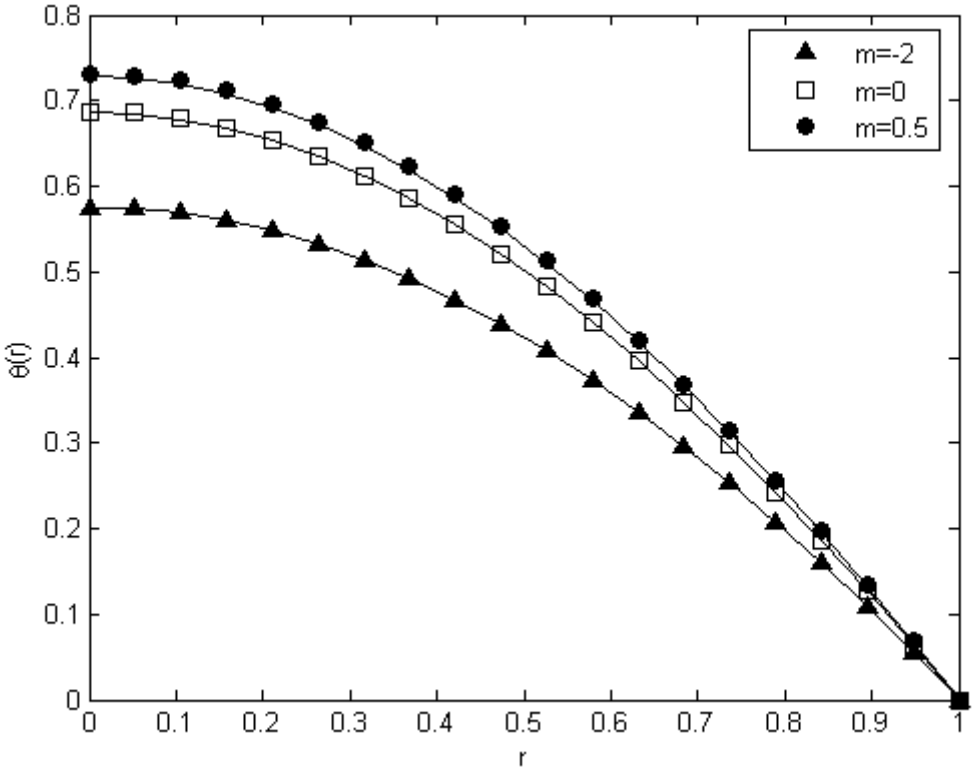

Figure 1. Comparison of slab numerical and HPM results of the boundary value problem (3-4) for ε = 0.1, δ = 0.5. Solid lines: Numerical solutions; Symbols: HPM solutions.

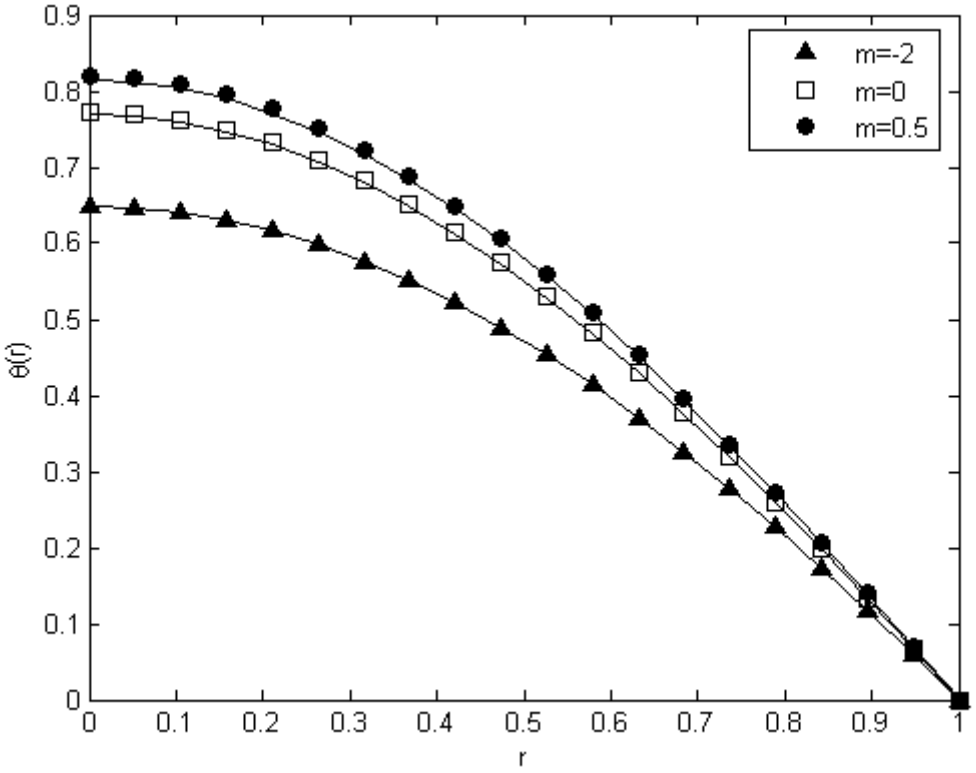

Figure 2. Comparison of cylinder numerical and HPM results of the boundary value problem (3-4) for ε = 0.1, δ=1.8. Solid lines: Numerical solutions; Symbols: HPM solutions.

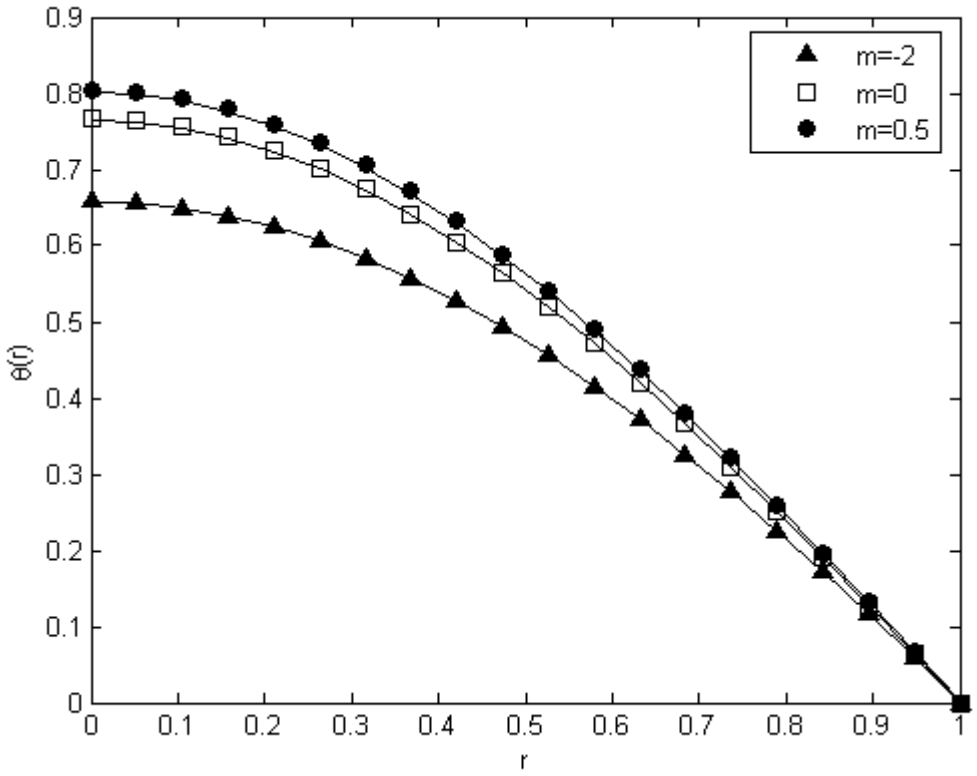

Figure 3. Comparison of sphere numerical and HPM results of the boundary value problem (3-4) for ε = 0.1, δ=2.8. Solid lines: Numerical solutions; Symbols: HPM solutions.

The obtained solutions, in comparison with the numerical solutions admit a remarkable accuracy. A clear conclusion can be drawn from the numerical results that the HPM provides highly accurate numerical solutions for nonlinear differential equations.

For a further information, Figure 4 illustrates a comparison between the exact solutions known for the special cases (m = 0, ε = 0, j = 0, j = 1) when a closed form is available [8] and the series solutions obtained by using the homotopy perturbation method. The obtained results are found to be in good agreement with the exact solutions. We can notice that the deviation between HPM and exact solution do not exceed 0.2 % for both slab and cylindrical geometries.

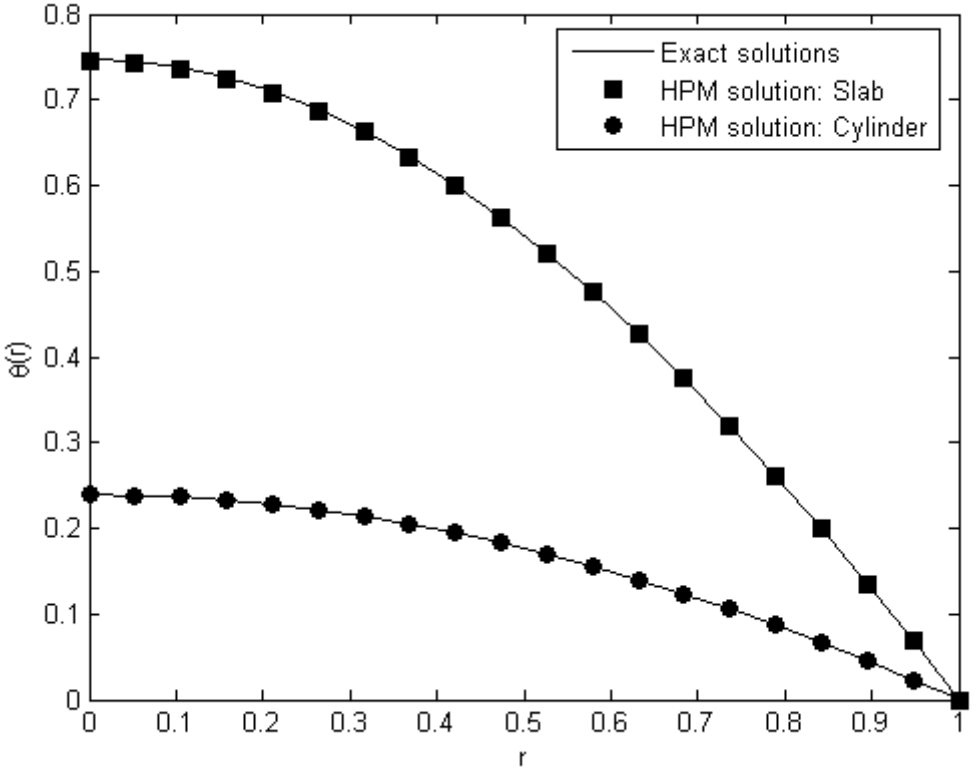

Figure 4. Comparison of HPM results and exact solutions of the boundary value problem (3-4) for ε = 0, m = 0 and δ = 0.8.

In the following we will focus on the calculation of radius of convergence δ_{cr} of the series solutions (9)-(11). Up to the point δ_c, the solution θ is analytic, and has a real singularity at the value $\delta_{cr.}$ It would be interesting to know the exact nature of the singularity, and if this singularity corresponds to a bifurcation point. Let the maximum temperature $\theta_{\max} = \theta(0, \delta, \varepsilon)$ be a characteristic quantity which qualifies the solution. We rearrange $\theta_{\max}$ in a series of δ to write:

$$\theta_{\max} = \sum_{n=0}^{30} c_n(\varepsilon)\delta^n \tag{17}$$

where different coefficient c_n for $\varepsilon = 0$ are given in table 1. Seeing Figure 5, one can say that the ratio $a_n = \left| \frac{c_n}{c_{n-1}} \right|$ in the simple geometry of infinite circular cylinder converges; however, it is extremely difficult to conclude to a precise value of radius of convergence. It is necessary to use some process of convergence acceleration. The Richardson extrapolation is appropriate for this kind of convergence. The results of Table 2 show that the radius of convergence is $\delta_{cr} = 2$. Two solution branches are therefore identified with a bifurcation point at δ_{cr}.Two solution branches (type I and II) are identified with a bifurcation point at δ_{cr} (i.e. turning point) as shown in a sketch of bifurcation diagram in Figure 6.

n	c_n	***n***	c_n	***n***	c_n	***n***	c_n	***n***	c_n
1	**0.25**	**7**	**2.337e-4**	**13**	**1.455e-6**	**19**	**1.290e-8**	**25**	**1.338e-10**
2	**4.687 e-2**	**8**	**9.588e-5**	**14**	**6.515e-7**	**20**	**5.978e-9**	**26**	**6.310e-11**
3	**1.302 e-2**	**9**	**4.024e-5**	**15**	**2.939e-7**	**21**	**2.778e-9**	**27**	**2.982e-11**
4	**4.272e-3**	**10**	**1.720e-5**	**16**	**1.334e-7**	**22**	**1.296e-9**	**28**	**1.412e-11**
5	**1.538e-3**	**11**	**7.465e-6**	**17**	**6.096e-8**	**23**	**6.064e-10**	**29**	**6.700e-12**
6	**5.874e-4**	**12**	**3.279e-6**	**18**	**2.798e-8**	**24**	**2.845e-10**	**30**	**3.184e-12**

Table 1. Coefficients c_n of $\theta_{\max}$, $\varepsilon = 0$, *m* = 0, *j* = 1.

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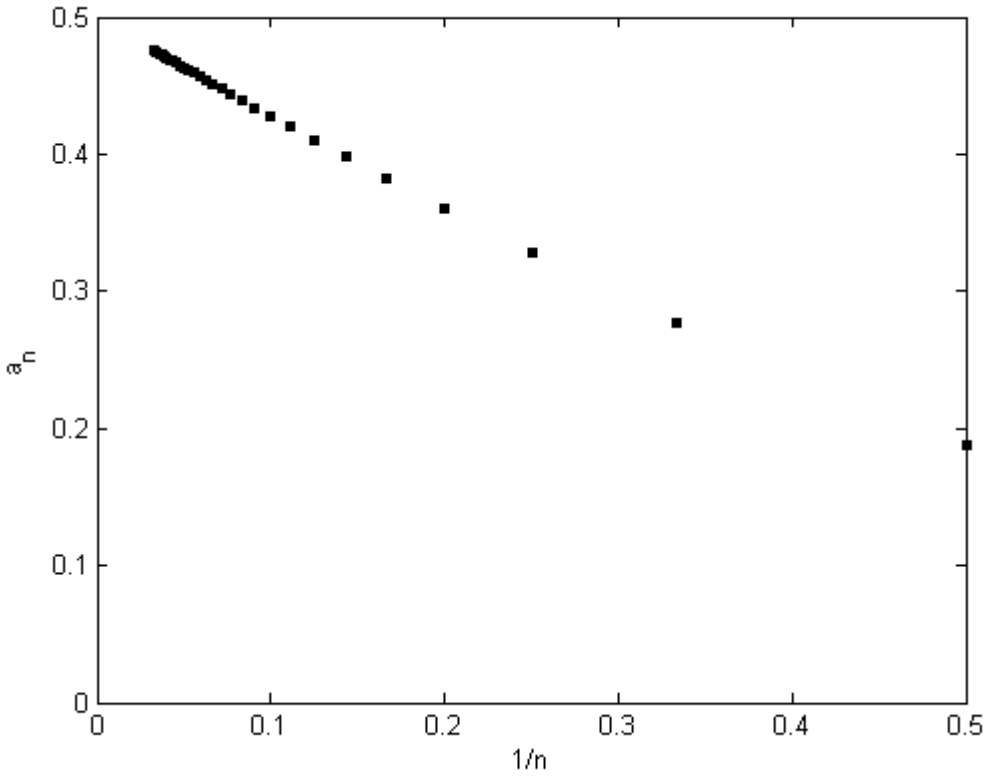


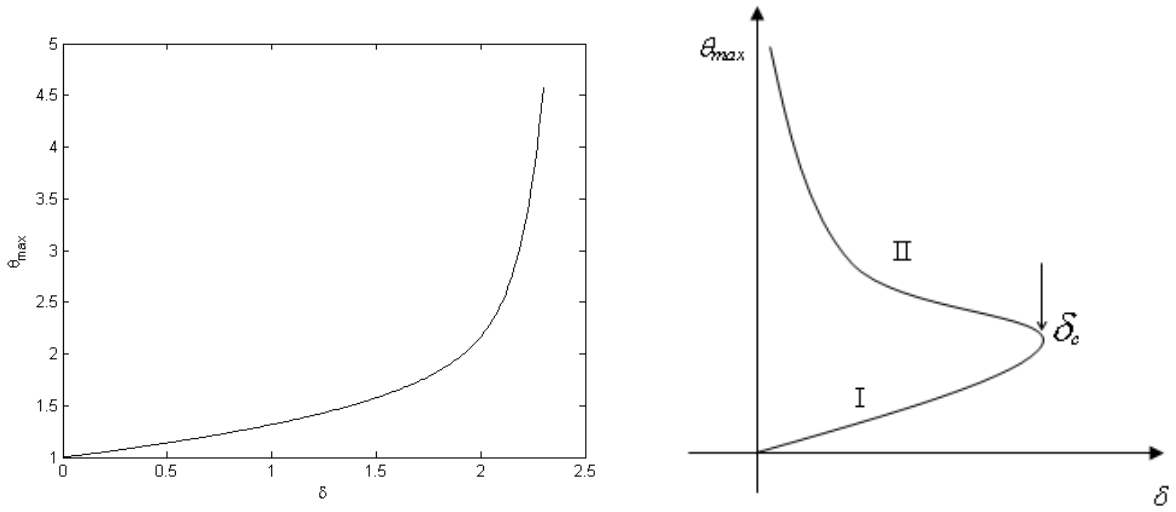
Figure 5. Sykes-Domb plot for $\theta_{\max}$, $\varepsilon = 0$, m = 0, j = 1.

n	m = 1	m = 2	m = 3	m = 4
20	0.4994	0.4999	0.5	0.4999
21	0.4994	0.5	0.4999	0.5
22	0.4995	0.5	0.5	0.4999
23	0.4995	0.5	0.4999	0.5
24	0.4995	0.4999	0.5	0.4999
25	0.4996	0.5	0.4999	0.5

Table 2. Values of the Richardson extrapolation s_{nm} for δ_{cr} , $\varepsilon = 0$, $m = 0, j = 1$.

It is possible to calculate the limited series defining $\theta_{\max} = \theta_{\max}(\delta)$ by a division procedure. This function is plotted in Figure 7.

The graph of θ_{max}, as shown in Figure 6(a), is not compatible with the expected singularity (13). The curve θ_{max} versus θ should be in the form of Figure 6(b). To analyse the paradox, the inverse function $\delta = \delta(\theta_{max})$ is considered. Its series is given by inverting the series θ_{max}. One can see in Figure 7 that this function has a horizontal tangent for the values in the case j = 1, (θ_{max} = 1.38435, δ = 1.99984).



(a) (b)

Figure 6. θ_{max} versus δ; (a) calculated, (b) expected form.

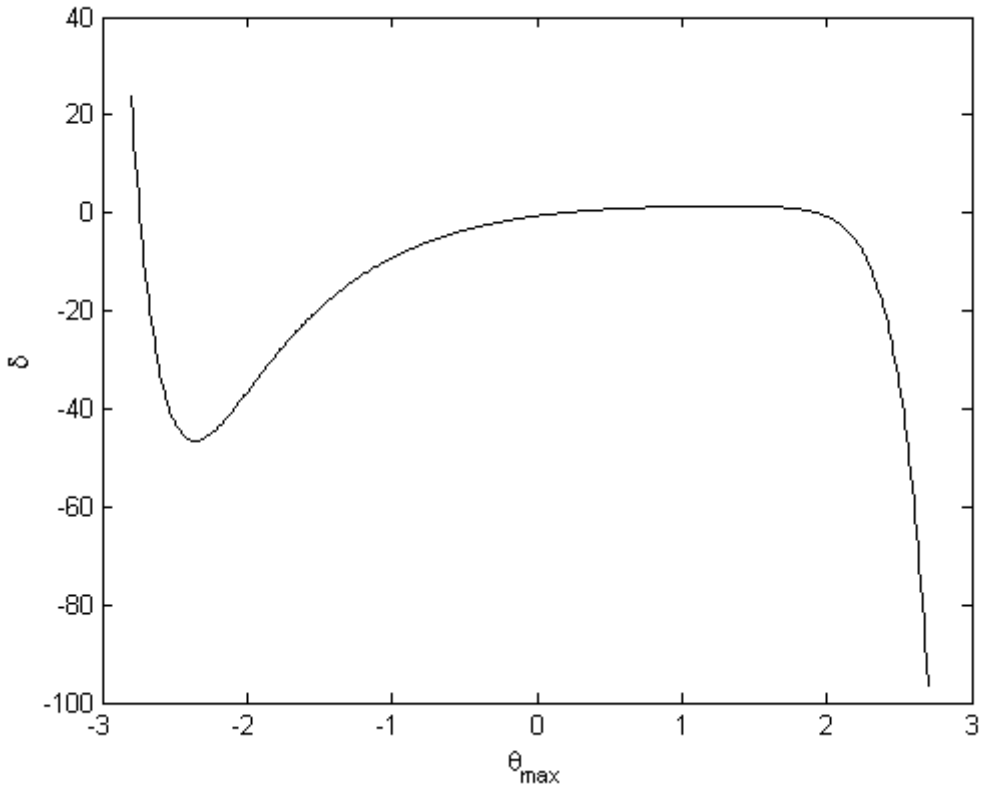


Figure 7. Plot of the function $\delta = \delta(\theta_{max})$.

We summarize the results of all Class A geometries in table 3,4 and 5 while $\theta_{cr} = \theta(0, \delta_{cr}, \varepsilon)$ is calculated by improving the series (17) by a suitable Padé approximant [27].

j	δ_{cr}	γ_{cr}	θ_{cr}
0	0.87846	0.5	1.18684
1	2	0.5	1.38629
2	3.32324	0.5	1.61782

Table 3. Computation showing criticality for Sensitised, Arrhenius and Bimolecular reactions for $\varepsilon = 0$ and for all Class A geometries.

The critical values (δ_{cr} and θ_{cr}) were computed using the improved Domb-Sykes applied to the temperature field obtained by HPM in all Class A geometries.

In order to verify the accuracy of this approximate method, we compare the critical values obtained using this method with the exact methods [1,2], variational methods [5,8] and

Hermite-Padé approach [19-21] for the special case of $\varepsilon = 0$.The method used in the present work gives results approximately equal to those discussed in literature, as shown in Table 4.

	Exact method	Variational method	Hermite-Padé Approach	Present work
Slab	$\delta_{cr} = 0.87846$ $\theta_{cr} = 1.186$	$\delta_{cr} = 0.878355$ $\theta_{cr} = 1.16716$	$\delta_{cr} = 0.87845$ $\theta_{cr} = 1.18684$	$\delta_{cr} = 0.87846$ $\theta_{cr} = 1.18684$
Cylinder	_____ _____	$\delta_{cr} = 1.99998$ $\theta_{cr} = 1.3801197$	$\delta_{cr} = 2$ $\theta_{cr} = 1.38629$	$\delta_{cr} = 2$ $\theta_{cr} = 1.38628$
Sphere	_____ _____	$\delta_{cr} = 3.32199$ $\theta_{cr} = 1.60746$	_____ _____	$\delta_{cr} = 3.32324$ $\theta_{cr} = 1.61782$

Table 4. Comparison of critical values obtained by different methods.

Tables 5-7 depict the variations of δ_{cr} and θ_{cr} with ε. In these tables, we observe that the magnitude of thermal criticality conditions for a viscous combustible material with high activation energy ($\varepsilon = 0$) is lower than the one for moderate values of activation energy ($\varepsilon = 0.01$, $\varepsilon = 0.1$). This implies that for moderate values of activation energy, the criticality varies from one reaction to another, as shown in Tables 5-7. Explosion in Bimolecular reactions seems to occur faster than in Arrhenius and Sensitised reactions.

m	ε	δ_{cr}	θ_{cr}
- 2	0.01	0.90624	1.24566
	0.1	1.31389	2.22239
0	0.01	0.8878	1.19841
	0.1	0.98819	1.52434
0.5	0.01	0.88331	1.19541
	0.1	0.93221	1.42024

Table 5. Variation of δ_{cr} and θ_{cr} with respect to ε (Slab).

m	ε	δ_{cr}	θ_{cr}
- 2	0.01	2.06415	1.58738
	0.1	3.01620	2.65419
0	0.01	2.02216	1.45337
	0.1	2.26129	1.80247
0.5	0.01	2.01192	1.41353
	0.1	2.13219	1.677327

Table 6. Variation of δ_{cr} and θ_{cr} with respect to ε (Cylinder).

m	ε	δ_{cr}	θ_{cr}
- 2	0.01	3.45641	1.67123
	0.1	4.32871	2.1177
0	0.01	3.42112	1.62756
	0.1	4.22245	2.21856
0.5	0.01	3.15167	1.63331
	0.1	4.04567	2.23811

Table 7. Variation of δ_{cr} and θ_{cr} with respect to ε (Sphere).

5. Conclusion

We studied the problem of exothermic explosion of a viscous combustible in Class A geometries under Arrhenius, Bimolecular and Sensitised laws of reaction rates with the homotopy perturbation method. The results show that this method provides excellent approximation of the solution of this nonlinear system with high accuracy.

A bifurcation study is performed with the Domb-Sykes graphical method to calculate the critical value of this problem. The results show that these critical values increase with the activation energy.

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