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Numerical study of an exothermic reaction with convective boundary conditions

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Abstract. In this work, we used a finite element method to solve the boundary value problem governing the ignition of a solid reactant undergoing slow oxidation for some non-class A geometries (infinite square rod and cube). We also examine the effect of Frank-Kamenetskii parameter on bifurcation and thermal stability by means of the block Lanczos method.

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1. Introduction

Energetic materials are a class of material that can release in a short time a high amount chemical energy stored in their molecular structure. The problem of self-ignition of energetic materials is of great importance in the field of thermal hazard of storage of explosives and pyrotechnics. The self ignition occurs by self-heating as a result of increase in temperature due to exothermic internal reactions. This leads to a release of heat, accompanied by a rise in temperature and a high velocity of thermal decomposition. Very often, this process causes a thermal runaway generating an explosion which can give rise to a devastating detonation.

In many problems of practical importance in the combustion theory, the critical values associated with thermal runaway are related either to the determination of the critical ambient temperature or to the critical initial temperature. This constitutes the structural basis for thermal stability of energetic materials.

This phenomenon was initially studied analytically by Frank-Kamenetskii [1] who developed the quasi-stationary theory of thermal explosion for determining the critical conditions for self-ignition of explosive hazardous materials.

After the pioneer work of Frank-Kamenetskii, various aspects of this phenomenon were studied intensively by many authors during past several decades, (see for examples refs [2-7]). When reactant consumption is neglected, the equation for the heat balance in a bounded domain Ω, can be written as:

$$k\Delta T + C_0 Q A \exp(-\frac{E}{RT}) = c\frac{\partial T}{\partial t} \quad \text{in } \Omega \tag{1}$$

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with the boundary condition

$$k\frac{\partial T}{\partial n} + H(T - T_a) = 0 \quad \text{on} \quad \partial\Omega, \tag{2}$$

where T is the absolute temperature, k is the thermal conductivity, c is the thermal capacity, C_0 is the initial concentration of the reactant species assumed to be constant, Q is the heat of reaction.

According to the Arrhenius law, the rate constant on the absolute temperature has the form $A\exp(-\frac{E}{RT})$, where E is the activation energy, R the universal gas constant and A is the pre-exponential factor, T_a is the ambient temperature, H is the heat transfer coefficient. $\frac{\partial T}{\partial n}$ is the normal derivative of the temperature, $\mathbf{n}$ is the outward normal vector on the boundary.

Following Balakrishnan et *al.* [4], we introduce the following dimensionless variables in Eq. (1):

$$u = (\frac{E}{RT_a^2})(T - T_a), \quad \varepsilon = (\frac{RT_a}{E}), Bi = \frac{Ha_0}{k}, \quad \delta = \frac{Q\sigma AEa_0^2}{kRT_a^2}\exp(-\frac{E}{RT_a})$$

and obtain the dimensionless governing equation together with the appropriate boundary condition as:

$$\Delta u + \delta\exp(\frac{u}{1+\varepsilon u}) = 0 \quad \text{on} \quad \Omega, \tag{3}$$

$$\frac{\partial u}{\partial n} + Biu = 0 \quad \text{on} \quad \partial\Omega, \tag{4}$$

δ is the Frank-Kamenetskii parameter, Bi is the Biot number and a_0 is some characteristic length scale of the problem.

One of the most commonly used formulation is to adopt the approximation of Frank-Kamenetskii leading to a linearized exponential Arrhenius law, in which ε is supposed so small to be neglected. So the problem (3-4) is reduced to the following nonlinear eigenvalue problem:

$$\begin{cases} \Delta u + \delta\exp(u) = 0 & x \in \Omega, \\ \frac{\partial u}{\partial n} + Biu = 0 & x \in \partial\Omega. \end{cases} \tag{5}$$

The problem (5) has been studied both analytically and numerically by various authors, for simple class A geometries (infinite cylinder, infinite slab, and a sphere) and also for some non class A geometries. Thermal ignition in all these geometries can be formulated using a single variable [1, 4, 8-9].

Thermal stability analysis in the theory of combustion, with or without consideration of consumption and diffusion of the reactant, has generally been performed using the Frank-Kamenetskii number as a parameter of bifurcation, ε is kept fixed. But this number dissimulates the role of the most practically significant control variable, namely the ambient temperature. Several authors [10-13] have assessed thermal ignition models with regard to multiplicity and stability of these steady states using the dimensionless ambient temperature.

In terms of the following dimensionless quantities:

$$u = \frac{RT}{E}, U = \frac{RT_a}{E}, \quad \lambda = \frac{Ra_0^2QC_0A}{KE}$$

the governing steady state of heat balance are reduced to:

$$\begin{cases} \Delta u + \lambda \exp(-\frac{1}{u}) = 0 & x \in \Omega, \\ \frac{\partial u}{\partial n} + Bi(u - U) = 0 & x \in \partial\Omega. \end{cases} \tag{6}$$

The aim is to determine the critical value of U, say U_{crit}, beyond which multiple solutions of the problem (6) occur.

In this paper, we first show how to use the finite element method to find an approximation of the solution of the problem (6). This can be done using a fixed point iteration method that we will describe below. Then, we deal with the study of the stability and bifurcation using the block Lanczos algorithm.

2. Computational method

2.1. Variational Formulation. In this section we first derive a variational formulation of the boundary value problem (6.1) subject to Robin boundary condition (6.2).

We multiply the equation (6.1) by a test function $v \in H^1(\Omega)$, integrate and use Green's formula to obtain:

$$\int_\Omega \nabla u \nabla v \, dx + Bi \int_{\partial\Omega} u \, v dS + \int_\Omega \lambda \exp(-\frac{1}{u}) \, v dx = BiU \int_{\partial\Omega} v dS, \quad \forall v \in H^1(\Omega) \tag{7}$$

It is obvious that the left term of this expression is not a bilinear form. Therefore the Lax-Milgram theorem cannot be applied here.

To solve this problem, we must linearize (6) in order to bring the solution to a series of linear problems converging to the solution of the initial nonlinear problem. We have several approaches to deal with such problems in particular Picard's method or Newton's method. In this work, an efficient finite element procedure based on Picard's method is developed for analyzing the problem (6).

2.2. Picard's method. Picard's method ca be used as an alternative way for solving a nonlinear PDE problem.

We here briefly recall the principle of Picard's method (or successive iterations), which is a variant of the fixed-point method.

Let the matrix system:

$$K\mathbf{u} = \mathbf{f} \tag{8}$$

as a result of the finite element discretization of a non-linear problem, where K is the stiffness matrix, $\mathbf{u}$ the unknown vector and $\mathbf{f}$ the 'load vector'. K may be linear or not.

The residue R, which is defined by: $R = \mathbf{f} - K\mathbf{u}$ is a measure of the distance from the solution.

The methods of solving nonlinear systems are all iterative. At every step, we calculate $d\mathbf{u}$, the solution increment defined by: $d\mathbf{u} = \mathbf{u}^{i+1} - \mathbf{u}^i$, where i is the index of the iteration.

We study the convergence of the system by successive approximations. This convergence can be measured by a standard of the increment or a norm of R $d\mathbf{u}$. The solution increment can also be seen as a descent direction, down to the zero residue, i.e. to the solution.

The method of Picard is a fixed point method [14]. It is defined by:

$$K(\mathbf{u}^i)\mathbf{u}^{i+1} = \mathbf{f}(\mathbf{u}^i) \quad \text{given } \mathbf{u}^0 \tag{9}$$

which can also be written as:

$$K(\mathbf{u}^i)d\mathbf{u} = R(\mathbf{u}^i)$$

for the solution increment $d\mathbf{u}$.

An accuracy of ε is required and the process may be terminated by setting the criterion:

$$\max \|\mathbf{u}^{i+1} - \mathbf{u}^i\| \ < \varepsilon$$

where $\| \ . \ \|$ is the Euclidian norm.

Rather than being recalculated at each iteration, the matrix K can be maintained constant over a number of sub-iterations.

When convergence of this procedure occurs, then the solution satisfies (8). The advantage of this method lies in the existence of a large enough radius of convergence that allows us not to worry too much about the initial approximation. In return, its convergence rate remains often too slow for practical applications of interest. We can then use a relaxation method speeding a little bit the scheme (9). The latter is defined as follows:

$$\begin{cases} K(\mathbf{u}^i)(\mathbf{u}^* = \mathbf{F}(\mathbf{u}^i), & \mathbf{u}^0 \text{ given} \\ \mathbf{u}^{i+1} = \alpha\mathbf{u}^{i+1} + (1-\alpha)\mathbf{u}^*, & 0 \leq \alpha \leq 1. \end{cases} \tag{10}$$

2.3. Method of solution. After the choice of appropriate finite elements by a method adapted from discretization in space, the system of nonlinear equations resulting system (7) is solved using Picard's method.

The discrete problem arises as usual by restricting $V = H^1(\Omega)$ to a discrete space V_h according to some mesh with some element type. Similarly, we let u_d be the discrete solution, so the discrete nonlinear problem is then written as:
Find u_d such that:

$$F(u_d, v) = L(v), \quad \forall v \in V_h,$$

with $u_d = \sum_{j=1}^{N} \alpha_j \Phi_j$ where Φ_j are some basic shape functions. Since F is a nonlinear function of u_d, the variational statement gives rise to a system of nonlinear algebraic equations.

We simply use a known previous solution in the nonlinear terms so that these terms become linear in the unknown u_d.

More precisely, given a solution u^k from iteration k, we seek a new (hopefully improved) solution u^{k+1} in iteration $k+1$ such that u^{k+1} solves the linear problem,

$$\begin{cases} \Delta u^{k+1} + \lambda \exp(\ -\frac{1}{u^k}) = 0 & x \in \Omega, \\ \frac{\partial u^{k+1}}{\partial n} + Bi(u^{k+1} - U) = 0 & x \in \partial\Omega. \end{cases} \tag{11}$$

The iterations require an initial guess u^0. The hope is that $u^k \longrightarrow u_d$ as $k \to \infty$, and that u^{k+1} is sufficiently close to the exact solution u_d of the discrete problem after just a few iterations.

We can easily formulate a variational problem for u^{k+1} from the last equation. Equivalently, we can approximate $e^{-\frac{1}{u}}$ to obtain the same linear variational problem. Therefore, the problem consists of seeking $u^{k+1} \in V_h$ such that:

$$a(u^{k+1}, v) = L(v) \ , \quad \forall v \in V_h \tag{12}$$

with:

$$\begin{aligned} a(u^{k+1}, v) &= \int_\Omega \nabla u^{k+1} \nabla v \; dx + Bi \int_{\partial\Omega} u^{k+1} \; v dS, \\ L(v) &= BiU \int_{\partial\Omega} v dS - \lambda \int_\Omega v \exp(-\frac{1}{u^k}) \; dx. \end{aligned}$$

The iterations can be stopped when $\varepsilon = \| \; u^{k+1} - u^k \; \| < tol$ where tol is a small tolerance, or when the number of iterations exceed some critical limit. The latter case will pick up divergence of the method or unacceptable slow convergence.

Note that the existence and uniqueness of the solution of problem (12) is established through the Lax-Milgram theorem [15]. Thus, Picard's linearization reads as follows:

Find u^{k+1} such that:

$$\int_\Omega \nabla u^{k+1} \nabla v \; dx + Bi \int_{\partial\Omega} u^{k+1} v dS = BiU \int_{\partial\Omega} v dS - \lambda \int_\Omega v \exp(-\frac{1}{u^k}) \; dx,$$

$$\forall v \in V_h, \quad \text{given } u^0.$$

3. Stability and bifurcation

3.1. Lanczos method. The Lanczos algorithm was originally proposed by Lanczos as a method for the computation of eigenvalues of symmetric and non-symmetric matrices. The idea was to reduce a general matrix to a tridiagonal form, from which the eigenvalues could be easily determined, For symmetric matrices, the Lanczos algorithm has been studied extensively ([16]- [17]).

Let $H = \int_\Omega \frac{1}{2}(\nabla u)^2 - \lambda G(u)) dx$ the functional related to (6.1) where G is a primitive of $exp(-\frac{1}{u})$.

The Rayleigh quotient is defined as:

$$R(x) = \frac{x^T J x}{x^T M x}, \tag{13}$$

where J is the Hessian, or energy sensitivity, matrix ($J_{ij} = \frac{\partial^2 H}{\partial \alpha_i \partial \alpha_j}$), M is the basis function overlap matrix ($M_{ij} = \int \phi_i \phi_j$). Both J and M are symmetric. Moreover, M usually called 'mass matrix' in finite element literature is positive definite. We are concerned by the minimum of the Rayleigh quotient $R(x)$ given by the i^{em} eigenvalue α_i:

$$\beta_i = \min \frac{x^T J x}{x^T M x}. \tag{14}$$

The minimization problem (14) is readily converted to a constrained generalized eigenproblem:

$$J x_i = \beta_i M x_i. \tag{15}$$

where the eigenvalues are necessarily real. It is convenient to order them from the smallest to the largest. Negative eigenvalues signal instability with respect to infinitesimal variations of u described by the corresponding eigenvector x_i. Established techniques of matrix transformation for solving (15) (see, for example [18]) destroy matrix sparsity and afterwards require storage of both J and M. Few computers have a large enough central memory to contain the entire problem, and so iterative methods must be used.

For (15) we prefer the block-Lanczos method developed by Golub and Underwood [19] and adapted as explained below. For the simple eigenproblem:

$$Az_i = \mu_i z_i, i = 0, ..., N, \tag{16}$$

where A is a positive definite and symmetric matrix, the method computes the lowest several eigenvalues and their eigenvectors. In each iteration the only operation involving A is multiplication by a vector, and thus any sparsity of A is preserved. The rate at which the method converges on an eigenvalue depends on the initial estimate of the eigenvector, on the spacings of the eigenvalues, and on their spread $|\beta_N - \beta_1|$ [20].

To convert (15) to the standard form (16) we transform it by means of the relations

$$\widehat{J} = J + sM = LL^T, \quad s > 0, \tag{17}$$

$$\frac{1}{\mu_i} = \beta_i + s, \tag{18}$$

where s is chosen large enough that $\widehat{J}$ is positive definite, and where L is the lower triangular matrix resulting from a Cholesky factorization of $\widehat{J}$.

The result is:

$$LL^T x_i = \frac{1}{\mu_i} M x_i \quad \Longrightarrow \quad \mu_i L^T x_i = L^{-1} x_i M. \tag{19}$$

Posing $y_i = L^T x_i$ so (19) becomes :

$$\mu_i y_i = L^{-1} M L^{T^{-1}} y_i. \tag{20}$$

The Lanczos method makes it possible to generate m vectors $q_1, ..., q_m$ which are orthonormal. So let Q be an orthonormal matrix formed by $q_1, ..., q_m : Q = [q_1, ..., q_m]$ matrix of N rows and m columns. By performing the variable change $y_i = Qz_i$ the expression (20) becomes

$$\mu_i Q z_i = L^{-1} M L^{T^{-1}} Q z_i \tag{21}$$

involved

$$\mu_i z_i = A z_i, \quad \text{where } A = L^{-1} M L^{T^{-1}} Q. \tag{22}$$

The matrix A is positive definite and symmetric and its eigenvalues and eigenvectors are found by the block-Lanczos method. The eigenvectors and eigenvalues of the original form of the stability problem (14) are recovered from

$$x_i = L^{-T} Q z_i, \quad \beta_i = -s + \frac{1}{\mu_i}. \tag{23}$$

If A in (22) were actually formed it would not be sparse. However, this matrix is not stored in the computer's memory. Just the sparse matrices L and M are stored, and the product of A with any vector is constructed by operations involving only solution of triangular systems of equations and matrix-vector multiplications. Profile storage methods [21] prove advantageous.

The eigenvectors x_i are finite element approximations to the normal modes of the absolute temperature, from which come J. The eigenvalues β_i if positive are related to the natural oscillation frequencies in the respective modes. Stability is of course governed by the sign of the lowest eigenvalues $\beta_1 = \frac{1}{\mu_1} - s$ which, if negative, indicates instability.

3.2. Numerical results. In this section we first present the spatial discretization using finite element method with $P1$ continuous piecewise linear functions. Then we develop all the steps of the FreeFem++ code to solve the problem by using the technique of mesh adaptation. At the end we present some numerical results.

To discretize (12), let T_h denote a regular, quasi uniform triangulation of Ω with triangles of maximum size $h < 1$. Let $V_h = \left\{ v_h \in C^0(\bar{\Omega}), v_{h/T} \in P_1(T), \forall T \in T_h \right\}$ denote a finite dimensional subspace of $H^1(\Omega)$ where P_1 is the set of polynomials of degrees ≤ 1 .

Let Ω be the rectangle $[0,1] \times [0,1]$, the triangulation T_h of Ω is automatically generated by using a FreeFem command.

Biot number (Bi)	critical values λ_{crit}
1	4,1
50	13,4
100	13,8
∞	14,3

Table 1. Critical values for various Biot numbers.

We have applied Lancsoz algorithm to compute critical values of λ. We observe that these values increase as the Biot number increases, the variation being shown in Table 1.

In Figures 1-3, we notice that the critical value λ_{crit} increases with the increase of Bi.

These figures are obtained by using the Lanczos algorithm taking account the solution obtained by FreeFem.

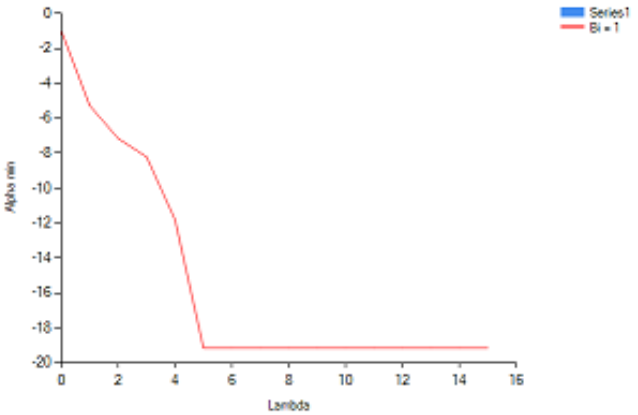

FIGURE 1. The critical value for Bi = 1.

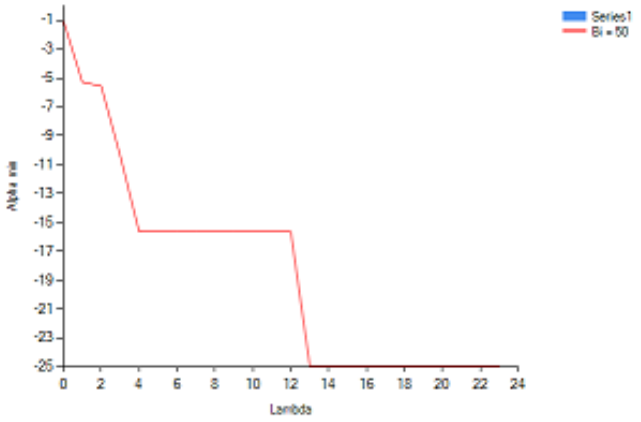

FIGURE 2. The critical value for Bi = 50,100.

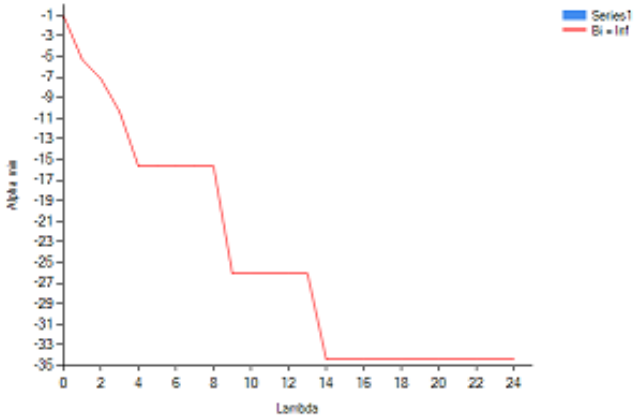

FIGURE 3. The critical value for Bi = 100.

4. Conclusion

In this work, we have applied the Lanczos algorithm implemented together with the finite element formulation for studying the thermal stability of an exothermic explosion in some non class A geometries. In these calculations, we were particularly interested in studying the effect of the Biot number on critical values of a specific parameter governing a strong exothermic explosion.

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