Iranian Journal of Mathematical Chemistry

IJMC

DOI: 10.22052/IJMC.2025.256937.2017
Vol. 17, No. 1, 2026, pp. 1-11
Research Paper

Variational Formulation of Thermal Explosion Problem with Internal Heat Generation

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Keywords:
Exothermic reaction,
Critical condition,
Approximate solution

AMS Subject Classification (2020):
35K57; 35J20

Article History:
Received: 26 May 2025
Accepted: 15 August 2025

Abstract

The article considers the problem of thermal stability in plane symmetry with an exothermic chemical reaction and constant heat release. The dependence of the critical reactivity on the intensity of heat release is investigated. Differential and variational formulations are considered; for the latter, an approximate analytical solution is given that relates the parameters of the problem for critical conditions. A simple Rayleigh-Ritz procedure results in a set of equations expressing the temperature distribution in terms of polynomials. The ignition boundary can be found through second derivatives of the integral, which can be evaluated using some simplifications that are typical for combustion theory. The results are reduced to simple approximations that can be used to estimate the ignition limits in systems with combined heat release.

1 Introduction

The complex behavior of chemical reactors is a widely studied topic due to its practical application. Among the variety of issues, thermal stability can be considered a key problem for the control of reactors. Exothermic reactions may demonstrate critical behavior with a sharp division of parameter space between slow and fast reaction rates. The criticality is concerned with changes in several stationary (or quasi-stationary) solutions of the coupled reaction and transfer equations. Losing low-temperature stability is often marked as a thermal explosion. The first mathematical models of thermal explosion were proposed by Semenov and Frank-Kamenetskiy [1], using zero-dimensional and one-dimensional approximations, respectively. In this work, we consider a modification of the Frank-Kamenetskiy model.

The simplest model of thermal explosion with internal heat generation was considered by Dik and Vilyunov [2, 3], where the heat balance equation with a stationary heat source was

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Academic Editor: Abbas Saadatmandi

approximately solved. It was shown that internal heating leads to the degeneration of thermal explosion. Thermal explosion with melting condition was considered by Aldushin [4]: the phase transition influence on critical condition was estimated.

The Frank-Kamenetskiy model describes thermal explosion in a one-dimensional area (reacting sample, gas vessel). Low-temperature approximation allows us to consider thermal stability without the influence of burnout. The modification taking into account a stationary and uniform heat source was proposed by Margolin [5]. It was shown that the Semenov and Frank-Kamenetsky models give similar results in a wide range of heat generation intensities. Joule heating with temperature-dependent electrical resistance was studied using numerical modelling by Frankel [6]. Propagation of reaction wave in a two-layered system was numerically studied by Bostandzhiyan et al. [7] concerning combustion synthesis.

A slightly different approach was used in the paper by Knyazik et al. [8]. Using a simple transformation, the authors proposed to consider a parabolic solution as a reference and solve the heat balance equation using a two-zone approximation. Numerical modelling was used to investigate the dynamic features of the ignition zone transition from the center to the whole area.

Non-linear reaction-diffusion equations with polynomial sources under reasonable requirements have variational formulations [9, 10]. The method of alternative functional allowing to control the numerical precision was proposed by Zarubin et al. [11, 12]. Thermal explosion with an expanded source was studied by Wake & Rayner [13]. Variational form allows us to study the stability of solutions [14]. Lyapunov functionals for stochastic media were constructed by Fedotov [15].

Application of variational methods to the thermal explosion problem started from the works of Grishin [16] (logarithmic function) and Anisimov & Vitkin [17] (polynomials). Critical conditions for complex reactions were studied by Graham-Eagles and Wake [18] (using polynomials and cosines). Variational problems for non-linear heat sources, including Joule heating, were considered in [19]. Similar variational formulations were also used in other areas [20]. Thermal explosion in viscous reactive flows was studied by Ajadi [21]. Recently, variational methods in thermal explosion were developed by Zarubin et al. [22], taking into account the temperature dependence of heat conduction, arbitrary convex boundaries, and non-local heat transfer [23]. Formulation for exact Arrhenius temperature dependence was proposed by Biyadi et al. [24]. Radiative heat transfer was considered in [25, 26]. Possible generalizations for the adiabatic flame equation are discussed in [27, 28].

Thermal explosion with internal heat generation is a crucial process in electrochemical battery failure. Joule heating induced by electric current may heat the electrolyte and initiate exothermic chemical reactions, leading to degradation and deformation of elements. After a shell rupture, toxic and flammable products spread over the battery storage space and can cause serious accidents [29]. To prevent this phenomenon, detailed mathematical models are developed, taking into account material properties and loading schedules [30]. However, it is important to have more fundamental models that allow us to analyze observed processes based on simple presentations of the involved processes.

The aim of the paper is to find an approximate solution to the thermal explosion problem, taking into account a uniform heat source. This problem is a model of the battery thermal degradation phenomenon, and the obtained solution can be a starting point in the analysis of the safety measures of energy storage.

In the present work, we consider the thermal explosion problem with a uniformly distributed heat source. The novelty includes a new variational formulation and new approximate relations between the reactivity and heat transfer parameters. It is also important that these results are concerned with using the variational method, which is more computationally efficient. Moreover,

the paper shows that the variational method leads to a simple approximate formula that can be derived by means of a special function (and its derivatives) which will be defined in paragraph 2.3.

2 Thermal explosion with uniform heat source

This section is organized as follows: the first subsection describes the differential formulation of the problem under study; the second subsection shows its linearized solutions; the third subsection presents the variational formulation of the problem and proposes a simple method to obtain an approximate dependence of the critical reactivity (Fk^*) on the heat source intensity (Po).

The main assumptions used are as follows: Arrhenius dependence of the reaction rate on temperature; negligible reagent consumption; prevailing conductive heat transfer; uniform heat generation by a non-chemical source.

2.1 Differential formulation

The Frank-Kamenetskiy problem concerns the thermal stability of a sample with a non-linear (exponential) heat source. This source is typically associated with an exothermic chemical reaction (or sometimes with viscous flow dissipation). The original statement is as follows:

$$\frac{d^2\theta}{d\xi^2} + \mathrm{Fk}\exp\left(\frac{\theta}{1+\mathrm{Ar}\theta}\right) = 0. \tag{1}$$

Here, θ is temperature, ξ is spatial coordinate, Fk is the Frank-Kamenetskiy number, which is heat generation by chemical reaction at ground temperature, and Ar is the Arrhenius number, which characterizes the sensitivity of chemical reaction rate to temperature (usually, $\mathrm{Ar} \approx 0$).

The boundary conditions are:

$$\frac{d\theta}{d\xi}(0) = 0, \tag{2}$$

$$-\frac{1}{\mathrm{Bi}}\frac{d\theta}{d\xi}(1) = \theta(1). \tag{3}$$

Here, Bi is the usual Biot number from heat diffusion theory [31]. When Bi is large, the Frank-Kamenetskiy problem has a critical value of the Fk parameter, which is close to 0.88.

Margolin [5] and Knyazik et al. [8] considered modification of the problem, including an additional constant heat source:

$$\frac{d^2\theta}{d\xi^2} + \mathrm{Fk}\exp\left(\frac{\theta}{1+\mathrm{Ar}\theta}\right) + \mathrm{Po} = 0. \tag{4}$$

Here Po is the Posnov number, which is a non-dimensional heat generation. This term is usually considered as heating caused by electric current.

Knyazik et al. [8] proposed a transformation:

$$\psi = \theta + \frac{\mathrm{Po}}{2}\xi^2.$$

The usual approximation is $\mathrm{Ar} = 0$. The previous equation can then be rewritten in the following form:

$$\frac{d^2\psi}{d\xi^2} + \mathrm{Fk}\exp(\psi)\exp\left(-\frac{\mathrm{Po}}{2}\xi^2\right) = 0.$$

It can be seen that the heat source strongly influences the chemical reaction in the vicinity of the center (since the Gauss function fades quickly). When Po is large, the area of greater influence of the heat source is about $\sqrt{\frac{2}{\mathrm{Po}}}$ (that is, the difference between the inert (Fk = 0) and reactive systems is the largest in this range).

At the same time, the boundary condition for large Bi gives $\psi(1) = \frac{\mathrm{Po}}{2}$. Using dimensional arguments, one can show that the critical value of Fk should depend on Po in the following way:

$$\mathrm{Fk}^* \approx \mathrm{Fk}^0 \exp\left(-\frac{\mathrm{Po}}{2}\right). \tag{5}$$

Here, Fk^0 is the critical value for the classical Frank-Kamenetskiy problem.

2.2 Linear theory

Linearization allows us to find approximate solutions and to study their stability [32, 33]. In some cases, relations between parameters can be obtained even in a linear approximation.

The first approximation, the crudest one, is to replace the non-linear term in Equation (4) by a constant. The problem turns into a well-known linear equation that can be solved immediately:

$$\theta(\xi) = \frac{\mathrm{Po} + \mathrm{Fk}}{2}\left(1 - \xi^2 + \frac{2}{\mathrm{Bi}}\right).$$

The solution has no critical parameters (except trivial singularity at Bi = 0).

The next approximation is keeping a linear term in the Taylor expansion. The truncated series gives an equation:

$$\frac{d^2\theta}{d\xi^2} + \mathrm{Fk}\,(1+\theta) + \mathrm{Po} = 0.$$

This equation has the following solution:

$$\theta(\xi) = \left(1 + \frac{\mathrm{Po}}{\mathrm{Fk}}\right)\left[\frac{\cos\left(\xi\sqrt{\mathrm{Fk}}\right)}{\cos\left(\sqrt{\mathrm{Fk}}\right) - \frac{\sqrt{\mathrm{Fk}}}{\mathrm{Bi}}\sin\left(\sqrt{\mathrm{Fk}}\right)} - 1\right].$$

It can be shown that this expression tends to the previous solution when Fk → 0. The key difference between solutions is that the latter has a non-trivial singularity when the denominator in the first term inside brackets tends to zero:

$$\cos\left(\sqrt{\mathrm{Fk}^*}\right) - \frac{\sqrt{\mathrm{Fk}^*}}{\mathrm{Bi}}\sin\left(\sqrt{\mathrm{Fk}^*}\right) = 0.$$

For Bi → ∞, this relation gives the critical value of the Frank-Kamenetskiy number as $\mathrm{Fk}^* = \pi^2/4$, which is almost three times higher than the critical value for the exponential term (0.88...). The approximate solution also does not result in dependence of Fk^* on Po.

Estimation of critical conditions (i.e., evaluation of Fk dependence on Po) cannot be obtained using linear approximations. However, there is another way to establish these relations.

2.3 Variational formulation

It was shown (in the mentioned works [11, 16, 18], etc.) that the Frank-Kamenetskiy problem (1) can be formulated in a variational form when Ar = 0 and Po = 0:

$$\delta \int_0^1 \left[\frac{1}{2}\left(\frac{dy}{d\xi}\right)^2 - \mathrm{Fk}\exp(y)\right] d\xi = 0.$$

Here, y is a function from the functional space satisfying boundary conditions (2) and (3). A constant heat source in (4) is a constant, which becomes a linear term in the variational formulation, as follows:

$$\delta \int_0^1 \left[\frac{1}{2} \left(\frac{dy}{d\xi} \right)^2 - \mathrm{Fk} \exp(y) - \mathrm{Po} y \right] d\xi = 0. \tag{6}$$

Applying the Euler–Lagrange equation to these extrema conditions gives the required equations. It should be noted that the extrema are not necessarily global; this feature will be discussed later.

Let us consider a simple approximation: temperature is a quadratic function of the coordinate. Then $y(\xi, a) = a\left(1 - \xi^2 + \frac{2}{\mathrm{Bi}}\right)$ is a one-parametric family of trial functions. Under positive values of Po, parameter a is related to the maximum (center) temperature. Noting the integral as I, one can write:

$$I(a) = \frac{2}{3}a^2 - \mathrm{Fk}\frac{\sqrt{\pi}}{2\sqrt{a}} \exp\left[a\left(1 + \frac{2}{\mathrm{Bi}}\right)\right] \mathrm{erf}(\sqrt{a}) - a\mathrm{Po}\left(\frac{2}{3} + \frac{2}{\mathrm{Bi}}\right).$$

Let us introduce a new function for simplification of formulas:

$$\Phi(a, \mathrm{Bi}) = \frac{\sqrt{\pi}}{2\sqrt{a}} \exp\left[a\left(1 + \frac{2}{\mathrm{Bi}}\right)\right] \mathrm{erf}(\sqrt{a}). \tag{7}$$

Then the stationary condition gives the equation for determining a:

$$I'(a) = \frac{4}{3}a - \mathrm{Po}\left(\frac{2}{3} + \frac{2}{\mathrm{Bi}}\right) - \mathrm{Fk}\Phi'(a, \mathrm{Bi}) = 0. \tag{8}$$

Here, the prime symbol denotes differentiation with respect to a. If $\mathrm{Fk} \to 0$, then $a = \frac{\mathrm{Po}}{2} + O(\frac{1}{\mathrm{Bi}})$, which is close to the exact solution.

The critical condition corresponds to $I''(a^*) = 0$:

$$I''(a^*) = \frac{4}{3} - \mathrm{Fk}^*\Phi''(a^*, \mathrm{Bi}) = 0. \tag{9}$$

From here, we can make a substitution and combine the stationarity and criticality conditions:

$$\frac{4}{3}a^* - \mathrm{Po}\left(\frac{2}{3} + \frac{2}{\mathrm{Bi}}\right) - \frac{4}{3}\frac{\Phi'(a^*, \mathrm{Bi})}{\Phi''(a^*, \mathrm{Bi})} = 0. \tag{10}$$

The latter equation gives the approximate relation between Po and critical maximum temperature a. Using implicit formulas, we can then construct the dependence between Po and Fk^*.

Instead of solving the cumbersome equations, we can search for a local minimum satisfying the following condition:

$$I(a^* - \delta a) \geq I(a^*) \leq I(a^* + \delta a).$$

This minimization problem could be solved using contraction techniques, for example, the bisection method. To this end, we need to specify a convex interval containing the minimum point. It is relatively easy to estimate the boundaries of such an interval by calculating I over a grid of a values. Then the algorithm is reduced to the estimation of $I'(a)$ at the boundaries of a convex interval.

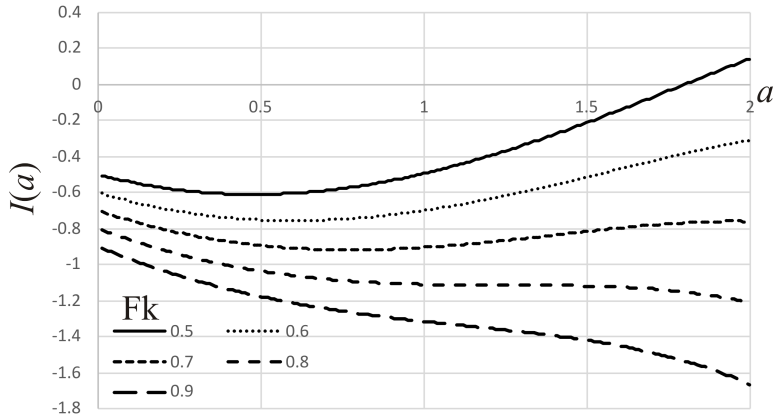

Figure 1: Dependence of I on a (parabolic approximation) under $\mathrm{Po} = 0.2$ and different values of Fk ($\mathrm{Bi} = 1000$).

Typical dependence of I on a is shown in Figure 1. The dependence is non-monotonous in a range of parameters where a stationary solution exists. If the values of Fk and Po are not compatible with a stationary reaction, then $I'(a) < 0$ for all values of $a > 0$. It can be shown from Equation (7) that $\Phi(0, \mathrm{Bi}) = 1$, and then $I(0) = -\mathrm{Fk}$. The critical value of Fk for $\mathrm{Po} = 0.2$ is close to 0.7: the corresponding curve has an inflection point near 1. It should be noted that the results correspond to the case $\mathrm{Ar} = 0$; otherwise, the explicit formula of the integral $I(a)$ requires additional special functions (for example, exponential integral). Several approximations for the Arrhenius integral were proposed in papers on thermal analysis [34, 35]. Expansions with respect to parameter Ar were proposed in [36].

3 Results and discussion

In this section, we will compare the results obtained by different techniques and derive simple formulas from the calculations. The developed variational method, with its numerical advantages, is demonstrated. Graphical presentation of stationarity and criticality conditions is shown. The calculations were carried out using the resources of the High-Temperature Circuit Multi-Access Research Center (MESI SB RAS).

First, we can check that if $\mathrm{Po} = 0$, then a^* is calculated by solving Equation (10) for $\mathrm{Bi} \to \infty$ is 1.21, and Fk^* is 0.89, which is close to the exact solution. Further, we can calculate the implicit dependence of Fk^* on Po using Equations (8) and (9). To this end, we vary a and solve a linear system of equations, which results in the following:

$$\mathrm{Po} = \frac{2}{1 + \frac{3}{\mathrm{Bi}}} \left(a - \frac{\Phi'(a)}{\Phi''(a)} \right),$$

$$\mathrm{Fk}^* = \frac{4}{3\Phi''(a)}.$$

Then, we can plot values of corresponding parameters, which are shown in Figure 2. The dependence of critical values Fk^* on Po is close to exponential, as expected from the qualitative treatment given above. Expectedly, at low values of Bi, the reacting system is more sensitive

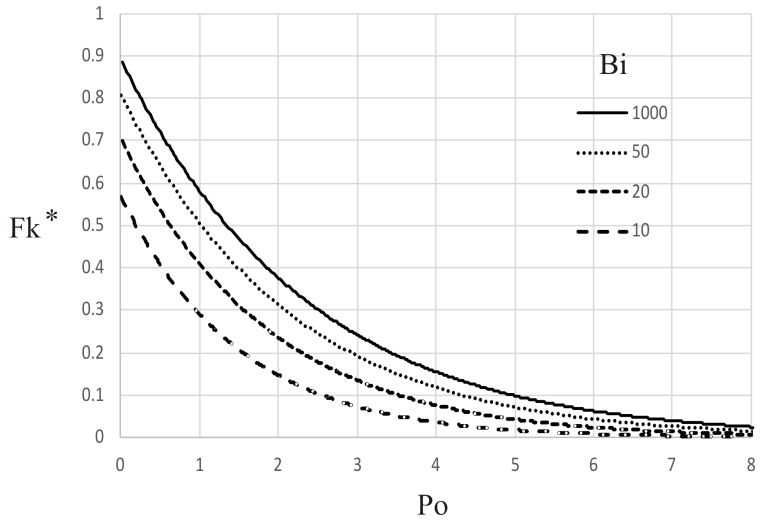

Figure 2: Implicit dependence of Fk^* on Po using parabolic approximation (the values of Bi are on the legend).

to heat generation due to poorer heat transfer. The curves presented in Figure 2 can be approximated using the following formula:

$$\mathrm{Fk}^* \approx \left(0.91 - \frac{3.26}{\mathrm{Bi}}\right) \exp\left[-\mathrm{Po}\left(0.44 + \frac{2.61}{\mathrm{Bi}}\right)\right]. \tag{11}$$

For large Bi values, this formula is close to the approximation (5). However, this formula, as well as the approach it is based on, gives high errors at low Bi values. The parabolic approximation poorly describes the temperature profile with weak external heat transfer. As for large Bi values, the parabolic profile is very close to the exact solution, and the dependence of Fk^* on Po is quite well reproduced by the variational approach.

Increasing the number of terms in polynomial approximation does not lead, however, to better agreement between variational approach results and direct search of critical conditions. For comparison, we calculated Fk^* by directly solving the differential problem using the bisection method to search for critical conditions [37]. Figure 3 shows the results. The dependence, which is closer to the exact solution, can be approximated by a formula:

$$\mathrm{Fk}^* \approx 0.88 \exp\left(-0.45\mathrm{Po} - \frac{2.09}{\mathrm{Bi}}\right). \tag{12}$$

The formulas (11) and (12) demonstrate similar dependence on Po with significant differences in dependence on Bi. The coefficient of Po is 0.44–0.45, which is close to 0.5, as expected from the dimensional analysis (5). This fact can be another justification for the approach proposed by Knyazik et al. It should be noted that at very low values of Bi, approximation (12) also becomes invalid.

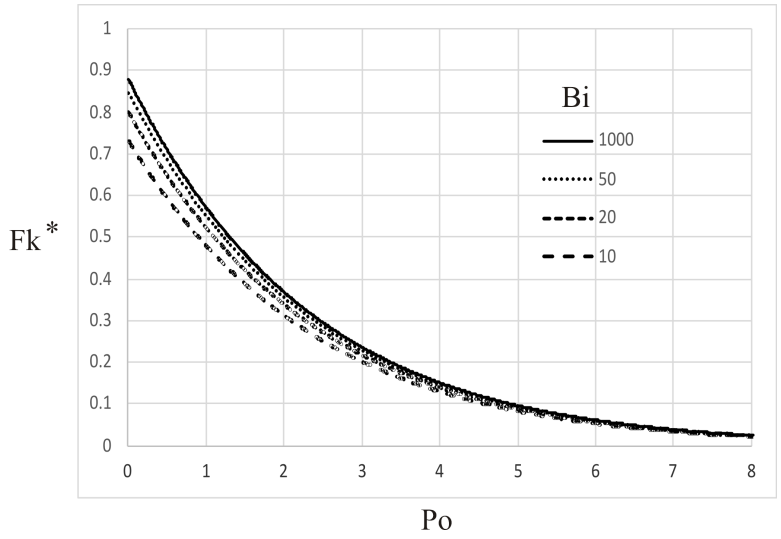

Figure 3: Implicit dependence of Fk^* on Po obtained by direct calculations (the values of Bi are on the legend).

4 Concluding remarks

Different formulations of the problem of thermal explosion with an additional heat source are proposed. It is shown that in the linear approximation, there is no dependence between the parameters of the problem on the boundary of thermal stability. A variational formulation is investigated, which allows one to obtain the dependence between the critical reactivity and the intensity of heat release in an implicit form. This dependence turns out to be exponential, and the coefficient at the intensity of heat release is equal to 0.44-0.45, which is close to the simplified estimate (0.5). However, the dependence on the intensity of heat transfer in the region of low values is reproduced more poorly.

Conflicts of Interest. The author declares that he has no conflicts of interest regarding the publication of this article.

Acknowledgment. The research was carried out under State Assignment Project (no. FWEU-FWEU-2026-0010) of the Fundamental Research Program of the Russian Federation 2021-2030.

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