

The thermal explosion revisited

G. I. BARENBLATT^{†‡}, J. B. BELL[‡], AND W. Y. CRUTCHFIELD[‡]

[†]Department of Mathematics, University of California, Berkeley, CA 94720; and [‡]Lawrence Berkeley National Laboratory, Berkeley, CA 94720

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ABSTRACT The classical problem of the thermal explosion in a long cylindrical vessel is modified so that only a fraction α of its wall is ideally thermally conducting while the remaining fraction $1-\alpha$ is thermally isolated. Partial isolation of the wall naturally reduces the critical radius of the vessel. Most interesting is the case when the structure of the boundary is a periodic one, so that the alternating conductive α and isolated $1-\alpha$ parts of the boundary occupy together the segments $2\pi/N$ (N is the number of segments) of the boundary. A numerical investigation is performed. It is shown that at small α and large N , the critical radius obeys a scaling law with the coefficients depending on N . For large N , the result is obtained that in the central core of the vessel the temperature distribution is axisymmetric. In the boundary layer near the wall having the thickness $\approx 2\pi r_0/N$ (r_0 is the radius of the vessel), the temperature distribution varies sharply in the peripheral direction. The temperature distribution in the axisymmetric core at the critical value of the vessel radius is subcritical.

1. Introduction

We revisit in this Note the classical problem of a thermal explosion in a long circular cylindrical vessel containing an exothermically reacting gas at rest. It was formulated and solved under some assumptions by D. A. Frank-Kamenetsky (refs. 1 and 2; see also ref. 3). In the original formulation, it was assumed that the wall of the vessel is ideally conducting, so that the gas temperature at the boundary is equal to the temperature of the ambient medium. Such a boundary condition made the problem cylindrically symmetric, and the symmetry essentially simplified the solution. In the present Note, the problem is modified in the following way: the wall is partially isolated so that the symmetry is lost. The critical values of the radius of the vessel are determined numerically. Especially instructive results were obtained for the cases when the isolated parts of the boundary are distributed periodically with large angular frequency, and the isolated part of the boundary is large. The scaling laws for the critical values were found. For large angular frequencies, it was found that there exists an axisymmetric core of the temperature distribution that occupies a major part of the vessel. The conditions in this core at the critical case were found to be subcritical.

2. Mathematical Problem Formulation

Assume that a gas at rest is enclosed in a long cylindrical vessel of radius r_0 . An exothermic reaction is going in the gas with the thermal effect Q per unit mass of reacted gas. For the reaction rate, the Arrhenius law is assumed with the activation energy E . If the thermal effect and the activation energy are large, it can be shown (see ref. 3) that an “intermediate-asymptotic” steady state regime is achieved. For this regime,

the gas consumption in the reaction can be neglected, and, from the other side, the temperature distribution in the vessel is steady. Applying the Frank-Kamenetsky large activation energy approximation, a nonlinear equation for dimensionless reduced temperature u is obtained:

$$\Delta u + \lambda^2 e^u = 0, \tag{2.1}$$

where

$$u = \frac{(T - T_0)E}{RT_0^2}, \tag{2.2}$$

the Laplace operator Δ is related to dimensionless variables $\rho = r/r_0$, θ ; r , θ are the polar coordinates. The constant λ is

$$\lambda = \frac{r_0}{l}, \quad l = \left(e^{\frac{E}{RT_0}} \kappa R T_0^2 c / Q E \sigma(T_0) \right)^{1/2}. \tag{2.3}$$

Here T is the absolute temperature, T_0 is the temperature of the ambient medium, R is the universal gas constant, κ is the molecular thermal diffusivity, c is heat capacity of gas per unit volume, and $\sigma(T)$ is the pre-exponential factor in the Arrhenius reaction rate expression: a slow function of temperature.

In the classical problem formulation, it was assumed that the whole wall of the vessel is ideally heat conducting, so that the gas temperature at the boundary is equal to the temperature of the ambient medium. This gives a Dirichlet condition for Eq. 2.1:

$$u(1, \theta) = 0. \tag{2.4}$$

The boundary value problem formulation under this condition is axisymmetric, and this was important for obtaining the analytic solution in an explicit form. D. A. Frank-Kamenetsky showed that the solution to the problem 2.1, 2.4 does exist for $\lambda \leq \lambda_{cr} = \sqrt{2}$ only. Physically it means that a quiet, nonexplosive proceeding of the reaction is possible only if the radius of the vessel is less than a critical one: $r_0 \leq (r_0)_{cr} = \sqrt{2}l$. This condition is known as the *condition of the thermal explosion*.

In the present Note, the following modification of the problem 2.1, 2.4 is proposed: only a fraction α of the wall is heat conducting, whereas the fraction $1 - \alpha$ is thermally isolated. The simplest formulation corresponds to a mixed problem (Fig. 1a)

$$\begin{aligned} u(1, \theta) &= 0 & \text{at } 0 \leq \theta \leq 2\pi\alpha \\ \partial_\rho u(1, \theta) &= 0 & \text{at } 2\pi\alpha < \theta \leq 2\pi. \end{aligned} \tag{2.5}$$

More interesting is the case when the isolated part of the wall is not concentrated on a single arc but is distributed periodically (Fig. 1b): the boundary $\rho = 1$ is divided into N segments

$$\frac{2\pi}{N}(1+k) \geq \theta \geq \frac{2\pi}{N}k, \quad k = 0, 1, \dots, N-1. \tag{2.6}$$

The fraction α of each segment is left heat conducting, while the fraction $1 - \alpha$ becomes isolated. In this case, the mixed boundary condition at $\rho = 1$ has the form:

$$\begin{aligned} u(1, \theta) &= 0 & \text{at } \frac{2\pi}{N}k \leq \theta \leq \frac{2\pi\alpha}{N} + \frac{2\pi}{N}k, \\ \partial_\rho u(1, \theta) &= 0 & \text{at } \frac{2\pi\alpha}{N} + \frac{2\pi}{N}k < \theta \leq \frac{2\pi}{N}(k+1). \end{aligned} \tag{2.7}$$

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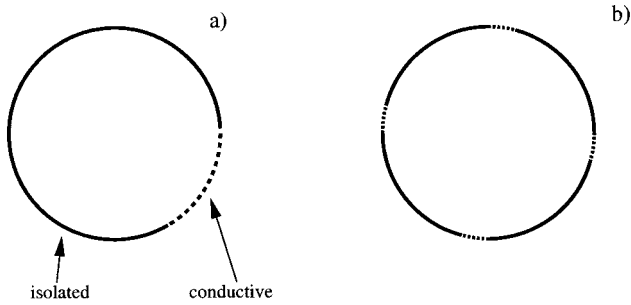


FIG. 1. A fraction of the wall is isolated. (a) Isolated (5/6) and conducting (1/6) parts are connected. (b) Isolated and conducting parts are distributed periodically.

The central question addressed in the present Note is: what are the asymptotic laws for the critical radius if $N \rightarrow \infty$ and $\alpha \rightarrow 0$ (i.e., the period of the boundary condition tends to zero, and the isolation is close to a complete one)? Remember, for the complete isolation the critical radius is equal to zero.

3. The Numerical Method

To answer the questions posed above, we must numerically evaluate the critical values λ_{cr} for fixed N and α . There are two special items in determining λ_{cr} : the singularity of the linearized Eq. 2.1:

$$\Delta \delta u + \lambda^2 e^u \delta u = 0 \tag{3.1}$$

at the critical value of $\lambda = \lambda_{cr}$, and the dependence of the values of λ_{cr} obtained by discretization upon the number of points, n , used to discretize each dimension. Here δu is the perturbation of the solution.

For fixed values of N , n , and α , we determined a trajectory of solutions u versus λ by solving Eq. 2.1 with a Newton-Raphson method. This method requires the solution of the linearized Eq. 3.1 which becomes singular at $\lambda = \lambda_{cr}$. In practice it prevents us from approaching the critical point closely. Therefore, to make an accurate determination of λ_{cr} , an extrapolation procedure was used. It was assumed that for λ approaching λ_{cr} , a parabolic approximation is valid:

$$\lambda_{cr}^2 - \lambda^2 = C(\|u\| - u_0)^2. \tag{3.2}$$

The parameters λ_{cr} , C and u_0 were determined to fit the last 10 points on the trajectory u versus λ approaching λ_{cr} . The approximation 3.2 happened to be satisfactory. Typically the fit 3.2 is accurate to a few parts in 10^6 .

The above procedure yielded an estimate for λ_{cr} as a function of N , α and the number of discretization points n . It is natural to remove the dependence of λ_{cr} of nonphysical, computational parameter n . For this purpose, another extrapolation was used. In our numerical approximations, the second-order accurate discretizations of the operators was employed. If the boundary conditions were smooth, the solution u would approach a limit with an error of the order of $1/n^2$. But the boundary conditions are nonsmooth, the derivative of the solution is discontinuous at the boundary, and it causes the order of the approximation to decrease. Extensive numerical calculations have shown λ_{cr}^2 to vary linearly with $1/n$. Therefore, the following iterative procedure was used: the value $\lambda_{cr}(N, \alpha, n)$ in 3.2 was calculated for three different values of n . These three values were then fit to a linear function $a + b/n$. If the fit was poor, as might happen if the values of n were too small, the procedure was repeated with larger values of n and so on, until a satisfactory fitting was obtained. The extrapolation $n \rightarrow \infty$ is simply the value of the coefficient a .

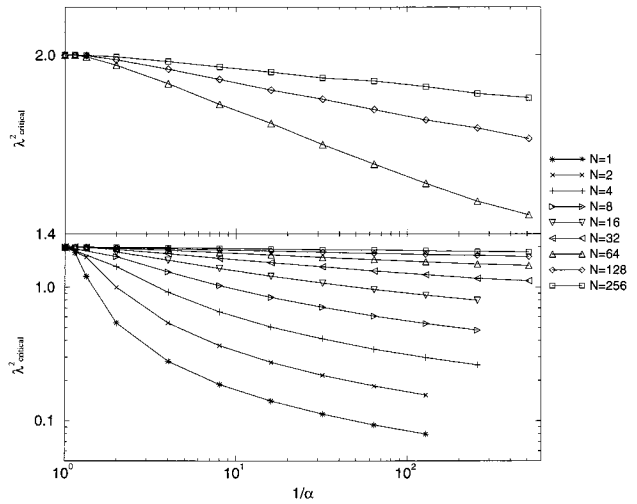


FIG. 2. Dimensionless critical radius as the function of conducting fraction α for different angular frequencies. It is seen that at large frequencies the critical radius is practically α -independent up to very small values of the conducting fraction α .

4. Results of the Numerical Analysis

The results obtained by numerical solution are represented on Figs. 2-4. Three instructive properties are revealed.

(i) On the graph of Fig. 2, the values of λ_{cr}^2 are presented for growing values of N as the functions of $1/\alpha$. It is seen that the critical value λ_{cr} , i.e., the critical radius of the vessel for large N , is practically insensitive to α up to α very small. For small N , the dependence of λ_{cr} on α is strong. Clearly, for any N , $\lambda_{cr} = 0$ for $\alpha = 0$, but it is instructive that for instance, for $N = 256$ when only 1/512 (0.2%) part of the boundary is

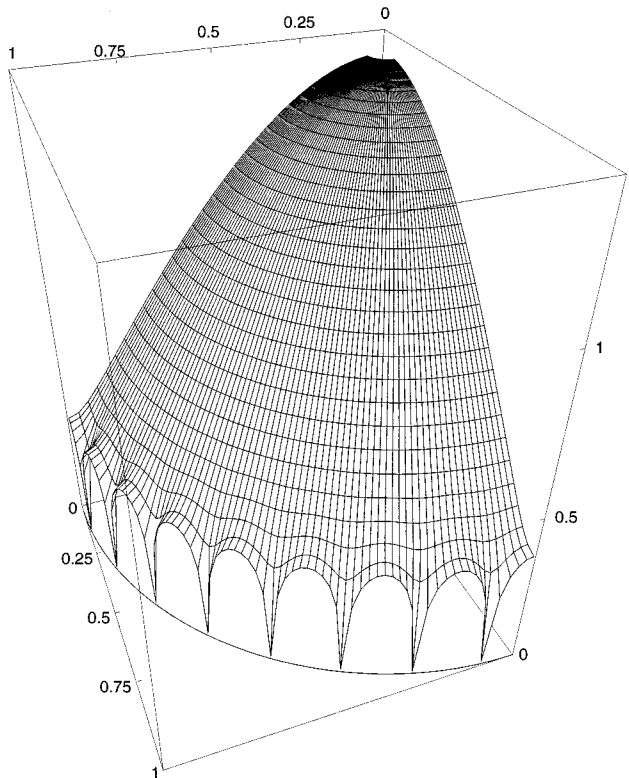


FIG. 3. The solution reveals an axisymmetric internal central core and "rugged" boundary layer ($N = 32, \alpha = 1/32$).

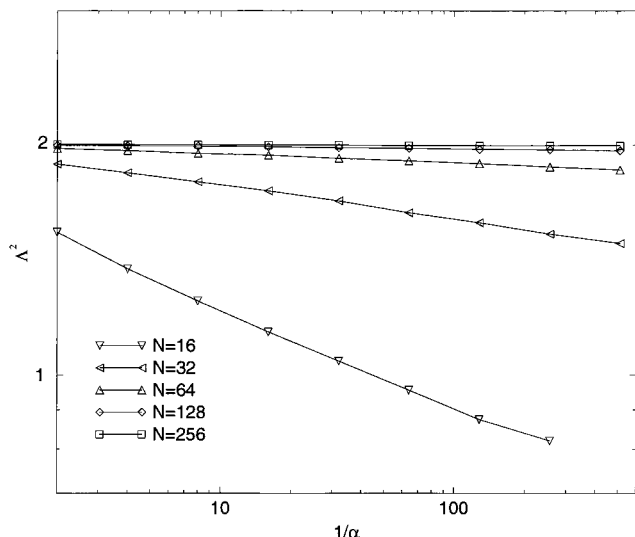


FIG. 4. The temperature distribution in the internal central core is a subcritical one: $\Lambda^2 < 2$.

heat conducting, the critical value of the radius is only 4% less than the critical radius for the wholly heat-conducting wall.

(ii) For large N , starting, say, from $N = 32$, there exists an internal core $0 \leq \rho \leq \rho_*$ where the solution is close to axisymmetric one (see Fig. 3). The value ρ_* was selected so that $|u_{\max}(\rho_*, \theta) - u_{\min}(\rho_*, \theta)| < 10^{-4}$. Introducing the mean value $u_* = (u_{\max} + u_{\min})/2$, we notice that for $0 \leq \rho \leq \rho_*$, the solution is close to axisymmetric, so that Eq. 2.1 and the boundary condition at $\rho = \rho_*$ assume the form

$$\frac{1}{\rho} \frac{d}{d\rho} \rho \frac{du}{d\rho} + \lambda^2 e^u = 0, \quad u = u_* \text{ at } \rho = \rho_*. \quad [4.1]$$

Transforming the variables

$$u = u_* + v, \quad R = \frac{\rho}{\rho_*}, \quad [4.2]$$

we reduce the problem to a classic one:

$$\frac{1}{R} \frac{d}{dR} R \frac{dv}{dR} + \Lambda^2 e^v = 0, \quad v(1, \theta) = 0, \quad [4.3]$$

where $0 \leq R \leq 1$, $\Lambda^2 = \lambda^2 \rho_*^2 e^{u_*}$. We calculate now the values of $\Lambda_{cr}^2 = \lambda_{cr}^2 \rho_*^2 e^{u_*}$ where λ_{cr} is the critical value obtained in previous calculations. The graphs Λ_{cr}^2 as the functions of $1/\alpha$ for different N are presented on Fig. 4. It can be seen that the values of Λ_{cr}^2 are always less than 2 (within the limits of our

Table 1. The parameters of the scaling law

N	$S(N)$	$t(N)$
32	2.03	0.10
64	2.03	0.055
128	2.02	0.028
256	2.01	0.015

numerical accuracy). This means that the “rugged” boundary layer near the wall $\rho = 1$ controls the approach to criticality. The thickness of this layer is of the order of the length of the segment $2\pi/N$. The angular derivative $\partial_\theta u$ in the boundary layer is large. The value u_* decreases as N increases.

(iii) The intermediate power laws are observed for λ_{cr}^2 at large N and small α :

$$\lambda_{cr}^2 = S(N)\alpha^{t(N)}. \quad [4.4]$$

The values of $S(N)$ and $t(N)$ for various N are given in Table 1.

5. Conclusion

Nonaxisymmetric modification of the problem of thermal explosion in a cylindrical vessel is formulated. The boundary is partly isolated, and only partly ideally conducting. Special attention is paid to the case of periodic distribution of the isolated and conducting parts. The critical values of radius and other relevant properties are obtained numerically.

It is shown that for the period small in comparison with the vessel radius the critical value of radius of the vessel is practically insensitive to the relative size of the open area of the wall up to its very small values. The temperature distribution in the central core is axisymmetric and subcritical even at globally critical conditions: the criticality is due to a thin boundary layer near the wall where the temperature distribution is highly nonaxisymmetric. Intermediate power laws are obtained for the critical radius as the function of the relative open area of the wall.

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