Effects of natural convection on thermal explosions in spherical vessels

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1 Introduction

The seminal theory of thermal explosions developed by Frank-Kamenetskii [1] examined the onset of thermal ignition of reactive mixtures enclosed in vessels with isothermal walls by investigating the existence of steady weakly reactive solutions. The corresponding critical conditions for ignition were seen to be determined by the competition between the heat released by the chemical reactions, which accelerates the temperature-sensitive reaction rate, and the heat losses to the container wall. Frank-Kamenetskii addressed initially stagnant systems in which heat transfer occurred solely by thermal conduction. Using a one-step Arrhenius model with large activation energy for the chemical reaction, he was able to characterize the state of the system in terms of a single parameter, a Damköhler number $\mathcal{D}$, defined as the ratio of the characteristic heat-conduction time across the container to the characteristic chemical time needed to increase the temperature by an amount of the order of the so-called Frank-Kamenetskii temperature. For a given geometry, the solution was represented in a diagram showing the maximum temperature increase in the container as a function of $\mathcal{D}$. Regardless of the geometry considered, the lower branch of the resulting curve, departing from the origin of the diagram, was found to exhibit a first turning point at a critical value $\mathcal{D}_c$ of order unity, beyond which the steady weakly reactive solution ceases to exist. Using the value of $\mathcal{D}_c$ together with the definition of the Damköhler number leads to explicit expressions for critical explosion sizes, giving results in close agreement with experimental observations, a truly notable achievement of the early theory given the many different simplifying assumptions involved in its derivation [2]. This remarkable success has motivated recent extensions of the early theory incorporating realistic chemistry in descriptions of hydrogen-oxygen systems that have been shown to predict explosion conditions in spherical vessels in excellent agreement with experiments [3], including critical pressures along the so-called third-explosion limit [4].

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It was soon acknowledged in the initial investigations reported by Frank-Kamenetskii [2] that in gaseous reactive systems under normal gravity conditions the density differences associated with the temperature increase induced by the chemical reaction, although small in thermal-explosion events, may suffice to generate significant convection, thereby questioning the assumption of stagnant fluid present in the original development. The influence of the resulting motion can be measured through a Grashof number Gr based on the characteristic induced velocity associated with the Frank-Kamenetskii temperature increase. For a given Gr, the diagram of peak temperature as a function of $\mathcal{D}$ can be compared with the motionless case to assess the effect of buoyancy on the onset of the thermal explosion. The different curves generated in computations for increasing values of Gr [5–7] were found to be almost indistinguishable from the convection-free results for values of Gr as large as a few hundred, with corresponding values of $\mathcal{D}_c$ differing by only a few percent from those of the buoyancy-free predictions [2], an unexpected outcome that has remained largely unexplained.

Many of the previous theoretical and numerical analyses of thermal explosions have considered simplified geometries, including the infinite slab, the infinite cylinder and the sphere. A number of theoretical analyses have addressed effects of buoyancy on explosions in horizontal infinite slabs [8–11], for which a motionless solution may exist when buoyancy is sufficiently weak. The present work deals instead with reactive gases in spherical vessels, for which a motionless state is never a solution to the conservation equations in the presence of gravity. The effect of convection on the temperature field and the corresponding modifications to the value of $\mathcal{D}_c$ are negligibly small for values of $Gr \ll 1$, in which limit the spherically symmetric Frank-Kamenetskii temperature distribution is recovered at leading order. The accompanying Stokes motion, arising from a balance between the buoyancy force and the viscous stresses, conforms an annular vortex that is symmetric about the equatorial plane, which is described here, including the simplified analytic solutions arising along the lower branch of the reactive curve near the origin (i.e., for $\mathcal{D} \ll 1$). The work is extended to investigate also higher-order perturbations to the Frank-Kamenetskii solution for $Gr \ll 1$ as well as the opposite limit of strong buoyancy $Gr \gg 1$.

2 Problem formulation

Following the classical analyses [2], the thermal explosion in a spherical container with an isothermal wall is formulated in terms of a one-step irreversible reaction, including an Arrhenius rate with an activation temperature T_a that is much greater than the wall temperature T_o, so that their ratio $\beta = T_a/T_o$ can be used as an asymptotically large quantity in describing the solution. Since the reaction-rate increase is associated with a small temperature increment of the order of the Frank-Kamenetskii temperature $(T - T_o) \sim \beta^{-1} T_o \ll T_o$, the associated density decrement with respect to the chemically frozen value $(\rho_o - \rho)/\rho_o$ is of order $\beta^{-1} \ll 1$, as inferred by the equation of state, with the result that the Boussinesq approximation can be used to investigate the resulting motion. The characteristic flow velocity $v_c = \beta^{-1} g a^2/\nu$ follows from a balance between viscous forces and buoyancy forces, with g and ν representing the gravitational acceleration and the unperturbed value of the kinematic viscosity, and a denoting the radius of the spherical vessel. Using v_c and a to scale the velocity and radial coordinate and introducing the dimensionless temperature increment $\Theta = \beta(T - T_o)/T_o$ reduces the momentum and energy equations for the weakly reactive solutions to

$$Gr\, \mathbf{v} \cdot \nabla \mathbf{v} = \nabla^2 \mathbf{v} - \nabla p + \Theta \mathbf{e}_z \tag{1}$$

$$Gr\, Pr\, \mathbf{v} \cdot \nabla \Theta = \nabla^2 \Theta + \mathcal{D}\, e^{\Theta}, \tag{2}$$

where $\mathbf{e}_z$ is the unit vector pointing upwards (against gravity) and p represents the pressure differences from the hydrostatic value scaled with $\rho_o g a/\beta$. Here, $Pr = \nu/D_T$, taken to be unity in the computations below, denotes the Prandtl number of the gas, with D_T correspondingly representing its thermal diffusivity. The Damköhler and Grashof numbers, given respectively by

$$\mathcal{D} = \frac{\beta[q/(c_pT_o)]BY_o e^{T_a/T_o}}{D_T/a^2} \quad \text{and} \quad Gr = \frac{ga^3}{\beta\nu^2}, \tag{3}$$

appear as the main controlling parameters in the formulation. Here, q, c_p, Y_o, and B are the amount of heat released per unit mass of reactant consumed, the specific heat at constant pressure, the initial reactant mass fraction, and the reaction-rate frequency factor. The solution is obtained by integrating (1) and (2) supplemented with the continuity equation $\nabla \cdot \mathbf{v} = 0$ with boundary conditions $\mathbf{v} = \Theta = 0$ at the container walls. For given values of Pr and Gr, at least one solution exists for values of $\mathcal{D}$ below a critical value $\mathcal{D}_c$ and no solution exists for $\mathcal{D} > \mathcal{D}_c$.

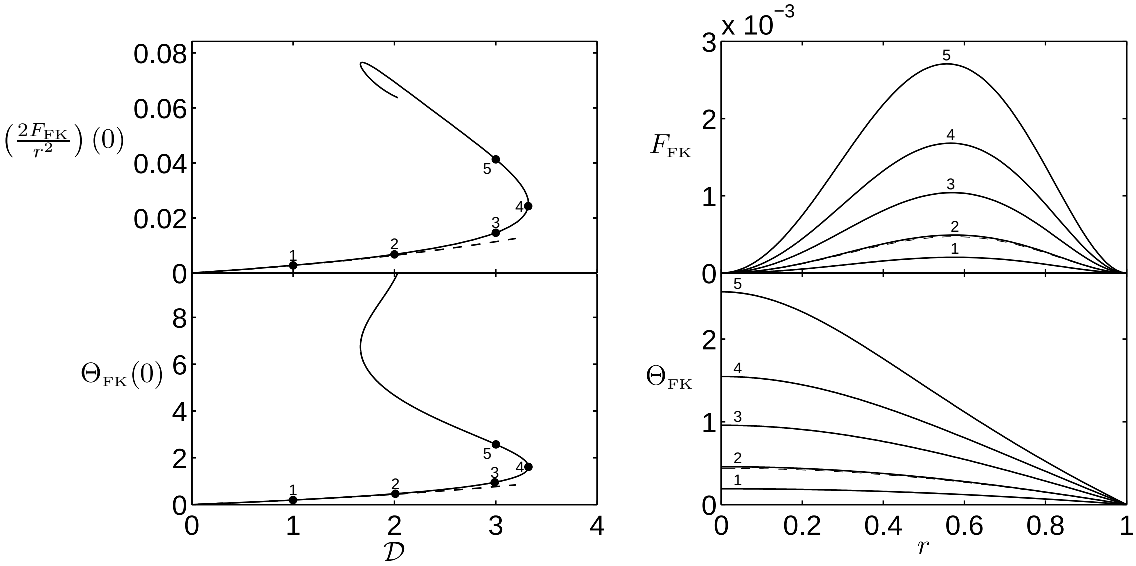

Figure 1: Summary of the Frank-Kamenetskii solution, including as dashed lines the asymptotic results for $\mathcal{D} \ll 1$ obtained by evaluating the first three terms in the expansions for $\Theta_{\rm FK}$ and $F_{\rm FK}$.

3 The Frank-Kamenetskii solution

The original analysis of Frank-Kamenetskii, neglecting the effect buoyancy, is recovered in the present formulation by setting $Gr = 0$ in (2) to give

$$\Theta''_{\rm FK} + \frac{2}{r}\Theta'_{\rm FK} = -\mathcal{D}\, e^{\Theta_{\rm FK}} \begin{cases} \Theta_{\rm FK}(0) \neq \infty \\ \Theta_{\rm FK}(1) = 0 \end{cases} \tag{4}$$

for the spherically symmetric Frank-Kamenetskii temperature field $\Theta_{\rm FK}(r)$, where the prime denotes differentiation with respect to r. The resulting temperature profile peaks at the center, with a value that depends on $\mathcal{D}$, as shown in Fig. 1, which also displays five different temperature profiles $\Theta_{\rm FK}(r)$ corresponding to the conditions indicated by the dots along the criticality curve $\Theta_{\rm FK}(0) - \mathcal{D}$. For the spherical geometry considered here the curve exhibits multiple turning points, the first one emerging for $\mathcal{D} \simeq 3.322$ when $\Theta_{\rm FK}(0) \simeq 1.61$. Since the remaining turning points occur always for $\mathcal{D} < 3.322$, for practical purposes only the first turning point becomes relevant in defining the critical explosion conditions, which are therefore given by $\mathcal{D}_c = 3.322$.

The spherically symmetrical temperature field obtained by Frank-Kamenetskii for $Gr = 0$ generates a convective flow in the form of an annular vortex, symmetric about the equatorial plane, with a velocity

$\mathbf{v} = (v_r, v_\theta)$ that can be determined by integrating (1) with $Gr = 0$. This velocity field can be expressed as $v_r = 2\cos\theta F_{\mathrm{FK}}/r^2$ and $v_\theta = -\sin\theta F'_{\mathrm{FK}}/r$ in terms of the Frank-Kamenetskii stream function $\psi_{\mathrm{FK}} = F_{\mathrm{FK}}(r)\sin^2\theta$, where F_{FK} is determined by integration of

$$F''''_{\mathrm{FK}} - 4F''_{\mathrm{FK}}/r^2 + 8F'_{\mathrm{FK}}/r^3 - 8F_{\mathrm{FK}}/r^4 = -r\Theta'_{\mathrm{FK}} \begin{cases} F_{\mathrm{FK}}(0)/r^2 \neq \infty \\ F_{\mathrm{FK}}(1) = F'_{\mathrm{FK}}(1) = 0 \end{cases} . \quad (5)$$

The solution depends on $\mathcal{D}$ through the function Θ'_{FK}, giving the results shown in Fig. 1. The stream function F_{FK} is seen to be very small, revealing that, as a result of the spherical confinement, the induced viscous flow is very slow. To illustrate further this effect, the figure includes the variation with $\mathcal{D}$ of the velocity at the center of the container, given by the value of $(2F_{\mathrm{FK}}/r^2)$ at $r = 0$. Besides, to complete the description of the Frank-Kamenetskii solution the temperature isolines and the streamlines corresponding to $\mathcal{D} = 2$ are plotted in Fig. 2.

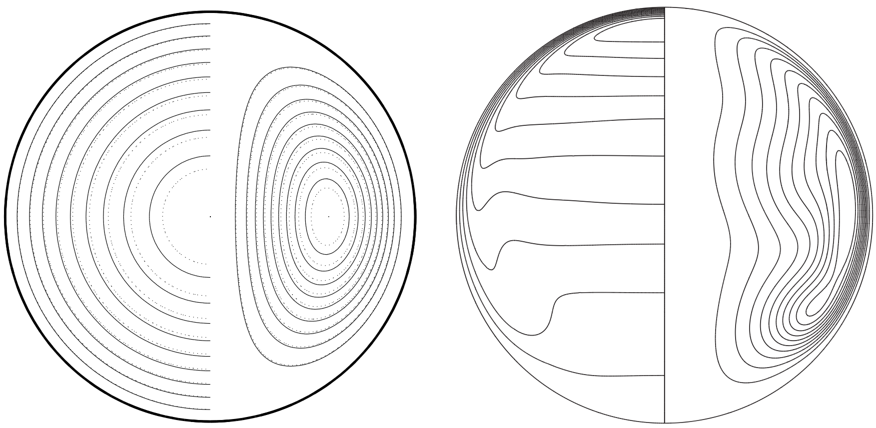

Figure 2: Temperature isolines (left hemispheres) and stream lines (right hemispheres) determined numerically for $\mathcal{D} = 2$ and $Gr = 0$ (left-hand-side sphere) and for $\mathcal{D} = 21$ for $Gr = 10^6$ (right-hand-side sphere); the dotted lines on the left-hand-side plot are obtained from evaluations of the three-term asymptotic expansions for $\mathcal{D} \ll 1$.

For small values of the Damköhler number the solution can be determined by introducing regular expansions of the form $\Theta_{\mathrm{FK}} = \mathcal{D}(\Theta_{\mathrm{FK}_0} + \mathcal{D}\Theta_{\mathrm{FK}_1} + \mathcal{D}^2\Theta_{\mathrm{FK}_2} + \cdots)$ and $F_{\mathrm{FK}} = \mathcal{D}(F_{\mathrm{FK}_0} + \mathcal{D}F_{\mathrm{FK}_1} + \mathcal{D}^2 F_{\mathrm{FK}_2} + \cdots)$. Using a Taylor expansion for the exponential in (4) provides a series of linear equations for the functions Θ_{FK_i} that can be solved sequentially subject to the conditions $\Theta_{\mathrm{FK}_i}(0) \neq \infty$ and $\Theta_{\mathrm{FK}_i}(1) = 0$. A similar sequential procedure applies to the solution for the stream function, with the linear problems arising at different orders obtained from (5) after evaluation of the buoyancy force on the right-hand side with use made of the expansion for Θ_{FK}. Up to three terms were computed analytically following this procedure. The corresponding asymptotic predictions are compared in Figs. 1 and 2 with the numerical results. The asymptotic expansions can be seen to remain accurate even for values of $\mathcal{D}$ of order unity.

4 Summary of results for $Gr > 0$

Deviations from the Frank-Kamenetskii solutions emerge for any nonzero value of Gr. Although the extent of the modifications to the heat-transfer rate can be expected from (2) to be of order Gr, the observed departures are much smaller because the buoyancy-induced flow is very slow. An exact quantification involves the numerical integration of (1) and (2). Results corresponding to different values of Gr, similar to those of previous numerical studies [5–7], were obtained with a finite-difference computation. Because of the pseudo-transient method employed here in seeking convergence to a steady solution, the branch of unstable solutions found beyond the first turning point is not accessible in the numerical integrations, so that the results shown in figure 3 are limited to the lower branch of solutions extending from $\mathcal{D} = 0$ to $\mathcal{D} = \mathcal{D}_c$. A notable feature of the results is that, as anticipated above, the deviations with respect to the Frank-Kamenetskii curve are very small up to relatively large values of Gr.

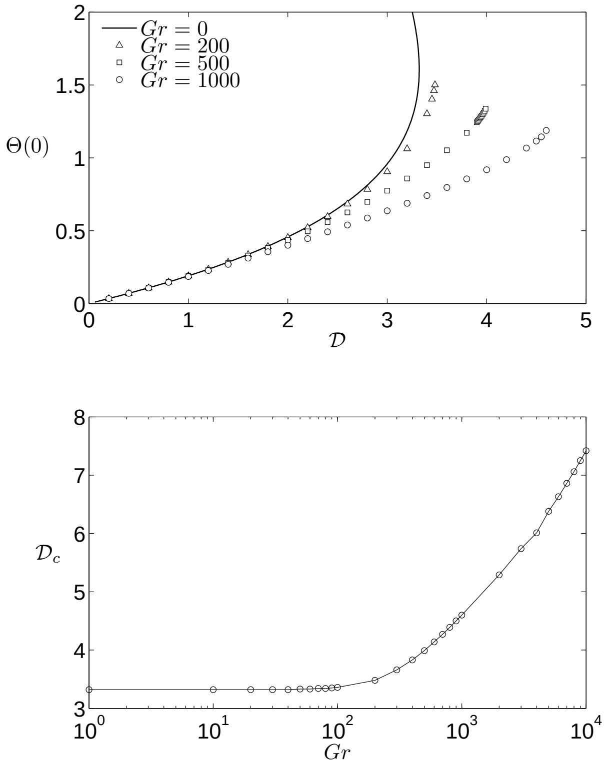

Figure 3: Results of integrations of (1) and (2).

The small deviations from the Frank-Kamenetskii solution resulting from the presence of fluid motion can be formally addressed by considering the asymptotic limit $Gr \ll 1$. In order to quantify the shift of the curve near the turning point, including the modifications to the predicted value of $\mathcal{D}_c$, it is necessary to proceed by selecting the solution corresponding to a given point along the Frank-Kamenetskii curve $\Theta_{\rm FK}(0) - \mathcal{D}_{\rm FK}$ and posing the problem as that of finding the Damköhler number $\mathcal{D}$ that, for a given value of $Gr \ll 1$, results in a temperature at the center of the container $\Theta(r = 0)$ equal to $\Theta_{\rm FK}(0)$. Hence, besides expansions for the temperature and stream function of the form $\Theta - \Theta_{\rm FK} = Gr\Theta_1 + Gr^2\Theta_2 + O(Gr^3)$ and $\psi - \psi_{\rm FK} = Gr\psi_1 + Gr^2\psi_2 + O(Gr^3)$ the analysis requires consideration of an expansion for the Damköhler number variation from its Frank-Kamenetskii value $\mathcal{D} - \mathcal{D}_{\rm FK} = Gr\delta_1 + Gr^2\delta_2 + O(Gr^3)$. The terms in the above expansions are determined by solving sequentially the different problems that arise at different orders in powers of Gr. The solution reveals, in particular, that δ_1 is identically zero, so that the shift $\mathcal{D} - \mathcal{D}_{\rm FK}$ is only of order Gr^2. The corresponding factor δ_2, a function of $\mathcal{D}$, turns out to be a very small number that increases for increasing values of $\Theta_{\rm FK}(0)$, reaching a value $\delta_2 = 2.2 \times 10^{-6}$ at the turning point of the original Frank-Kamenetskii curve. This small value is in agreement with the numerical results shown in Fig. 2.

The opposite limit $Gr \gg 1$ is also worth investigating. The numerical integrations shown on the right-hand side of Fig. 2, corresponding to $Gr = 10^6$ and $\mathcal{D} = 21$, reveal that the flow structure for $Gr \gg 1$ includes a central region of slowly moving hot gas, bounded by a high-velocity near-wall boundary layer driven by buoyancy. The scales and dominant balances applying in each region can be anticipated by an order-of-magnitude balance. Thus, since the temperature increment must be $\Theta \sim 1$ for the thermal explosion to develop, in the boundary layer the buoyancy force in (1) is of order unity, inducing streamwise velocities v_θ of order $Gr^{-1/2}$ in a near-wall layer of characteristic thickness $Gr^{-1/4}$, as inferred from the convection-diffusion balance in (1). The flow in the central region, outside the boundary layer, is induced by the boundary-layer entrainment, with characteristic velocities of order $Gr^{-3/4}$. This slow motion does not induce an appreciable pressure disturbance inside the container, where (1) reduces to $-\nabla p + \bar{\Theta}\mathbf{e}_z = 0$, with $\bar{\Theta}$ denoting the temperature in the container outside the boundary layer. This result indicates that the pressure p and the temperature $\bar{\Theta}$ are only a function of z (the latter dependence is clearly seen in the stratified temperature field of Fig. 2). Convection dominates the heat transfer in this central region, as follows from (2). This equation also indicates that the characteristic Damköhler number at ignition is of order $\mathcal{D} \sim Gr^{1/4}$, so that an appropriate convection-reaction balance leading to the needed temperature increment $\bar{\Theta} \sim 1$ can be reached in the container interior, while the boundary layer remains chemically frozen in this limit, because the transport rates there are larger than the reaction term by a factor of order $Gr^{1/4}$, as can be seen from (2).

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