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This paper investigates the effect of buoyancy-driven motion on the quasi-steady "slowly reacting" mode of combustion and on its thermal-explosion limits, for gaseous mixtures enclosed in a spherical vessel with a constant wall temperature. Following Frank-Kamenetskii's seminal analysis of this problem, the strong temperature dependence of the effective overall reaction rate is taken into account by using a single-reaction model with an Arrhenius rate having a large activation energy, resulting in a critical value of the vessel radius above which the slowly reacting mode of combustion no longer exists. In his constant-density, convection-free analysis, the critical conditions were found to depend on the value of a Damkohler number, defined as the ratio of the time for the heat released by the reaction to be conducted to the wall, to the homogeneous explosion time evaluated at the wall temperature. For gaseous mixtures under normal gravity, the critical Damkohler number increases through the effect of buoyancy-induced motion on the rate of heat conduction to the wall, measured by an appropriate Rayleigh number $Ra$. In the present analysis, for small values of $Ra$, the temperature is given in the first approximation by the spherically symmetric Frank-Kamenetskii solution, used to calculate the accompanying gas motion, an axisymmetric annular vortex determined at leading order by the balance between viscous and buoyancy forces, which we call the Frank-Kamenetskii vortex. This flow is used in the equation for conservation of energy to evaluate the influence of convection on explosion limits for small $Ra$, resulting in predicted critical Damkohler numbers that are accurate up to values of $Ra$ on the order of a few hundred.

Keywords: Thermal explosion, Laminar reacting flows, word, word, word
in a critical value of $Da_c \approx 3.322$, given by the classical Frank-Kamenetskii (F-K) explosion problem [2]. Effects of buoyancy will now be investigated. Specifically, the aim of the present work is to obtain a qualitative and quantitative description and understanding of the effects of the buoyant motion on the temperature distribution for the quasi-steady, slowly reacting mode of combustion, as well as to determine the associated modifications of the explosion limits.

While the initial work of Frank-Kamenetskii [2] considered constant density, so that heat transfer occurred only by conduction, it was soon acknowledged [3] that in gaseous reactive systems under normal gravity the density differences associated with the small changes in temperature, of the order of the F-K temperature for the slowly reacting mode of combustion, generate a convective motion having a non-negligible effect on the heat-transfer rate if the corresponding Peclet numbers are of order unity or larger. The special non-generic case of a slab configuration, with a reactive gas bounded by two isothermal infinite horizontal walls, has been considered in a number of previous theoretical analyses that address the effects of buoyancy-induced convection on thermal explosions [4–7]. As in the closely related case of the Rayleigh-Benard problem of convection produced by wall-temperature differences, a motionless quasi-steady combustion mode (similar to that described in Part 1) may exist for values of the Rayleigh (or Grashof) number below a critical value, at which a bifurcation occurs to a convective state, as described by linear stability analyses [4, 5]. It was found that convection sets in before the explosion limit is reached and that, contrary to widespread expectations, convection does not always hinder the development of the thermal explosion [6]. While sufficiently small eddies with widths on the order of or smaller than the slab height raise the explosion threshold by enhancing heat removal from the reaction region, long-wave convection rolls favor explosions by promoting the formation of hot spots [6], an aspect of the problem further investigated in [7] with use made of a prescribed velocity field.

Unlike the case of a horizontal infinite slab, for reactive gases in cylindrical and spherical vessels any gravity force at all establishes buoyant motion because the horizontal variations of the density that are present in these configurations generate forces that cannot be compensated by a vertical pressure gradient. Numerical integration has been employed previously [8–10] to investigate influences of the buoyancy-induced motion, measured by a Rayleigh number $Ra$, on the explosion limits. By enhancing heat losses to the wall, the presence of convection postpones the thermal explosion to larger values of $Da$ [11, 12]. Surprisingly, however, the resulting values of $Da_c$ differ by only a few percent from those of the buoyancy-free predictions for values of $Ra$ as large as a few hundred [8–10], an outcome of the numerical integrations that agrees with early observations [3]. This will be clarified by the perturbation analysis given below for small values of $Ra$.

The structure of the paper is as follows. The steady equations for the quasi-steady, slowly reacting mode of combustion in spherical vessels with natural convection, expected from the analysis of Part 1 [1] to occur at the end of an initial transient stage with negligible reactant consumption, are presented in Sec. 2. As in Part 1 of this work, the analysis is carried out for an overall irreversible reaction with an Arrhenius rate having a large activation energy to model the chemistry.

The spherically symmetric F-K temperature field, found at leading order in a perturbation analysis for small $Ra$, induces a toroidal Frank-Kamenetskii vortex, given at leading order by a balance between viscous and buoyancy forces, as described in Sec. 3. Higher-order terms are computed in Sec. 4 to determine the influence of convection on the critical ignition conditions, yielding corrections to the bifurcation diagram that are found to be quantitatively small, in agreement with the
numerical computations. The paper ends in Sec. 5 with concluding remarks.

2. Conservation equations for the slowly reacting mode of combustion accounting for buoyancy-induced motion

2.1 Formulation for the transient near-explosion regime

Following Part 1, the reaction-rate expression selected is

\[ \dot{m}/\rho = Y B \exp[-E/(RT)], \]  

where \( \dot{m} \) is the mass of reactant consumed per unit volume per unit time, which, when divided by the density \( \rho \), is a function of the temperature \( T \) and of the reactant mass fraction \( Y \) (variations of which were shown in Part 1 to be negligible), with \( B \) the frequency factor, \( E \) the activation energy, and \( R \) the universal gas constant; correspondingly, the heat-release rate per unit volume is \( q \dot{m} \), where \( q \) denotes the amount of heat released per unit mass of reactant consumed. In terms of the wall temperature \( T_w \), the nondimensional activation energy \( \beta = E/(RT_w) \) again is the large parameter of expansion, with variations of the temperature from the wall value and of the density from its mean value \( \rho_0 \) small, of order \( T_w/\beta \) and \( \rho/\beta \), respectively, so that the non-dimensional temperature increase \( \phi = \beta(T - T_w)/T_w \) is of order unity in this distinguished limit. Leaving out terms of order 1/\( \beta \) as before, departures of transport coefficients from their values evaluated for \( T = T_w \) and \( \rho = \rho_0 \) can be neglected. The principal non-dimensional parameters of the problem are the Damköhler number of order unity,

\[ Da = (a^2/D_T)[(qY)/(c_p T_w)][E/(RT_w)]B \exp[-E/(RT_w)], \]  

where \( a \) is the radius of the sphere and \( D_T \) the thermal diffusivity, and the Rayleigh number (based on the ordering of the temperature difference),

\[ Ra = \frac{\beta^{-1} g a^3}{\nu D_T}, \]  

where \( g \) is the acceleration of gravity and \( \nu \) the kinematic viscosity. This Rayleigh number measures the convective transport in the energy equation, while the Grashof number, \( Ra/Pr \), involving the Prandtl number \( Pr = \nu/D_T \), similarly measures convective transport in the momentum equation. The formulation is entirely non-dimensional, lengths being scaled with \( a \) and time with \( a^2/D_T \).

Irrespective of the shape of the vessel, the equations describing the distributions of temperature and velocity of the reacting gas at the beginning of the transient reaction stage in the distinguished near-explosion regime corresponding to the limit of large activation energies with fixed values of \( Da \) and \( Ra \) of order unity are the continuity, momentum, and energy conservation equations for the slowly reacting mode of combustion, which take the form

\[ \nabla \cdot \mathbf{v} = 0, \]  

\[ \frac{1}{Pr} \left( \frac{\partial \mathbf{v}}{\partial t} + Ra \mathbf{v} \cdot \nabla \mathbf{v} \right) = \nabla^2 \mathbf{v} - \nabla p' + \phi \mathbf{e}_z, \]  

\[ \frac{\partial \phi}{\partial t} + \frac{1}{\gamma} \frac{\partial p'}{\partial t} + Ra \mathbf{v} \cdot \nabla \phi = \nabla^2 \phi + Da e^\phi, \]
in the non-dimensional variables. In the momentum equation (5), \( \mathbf{e}_z \) is the unit vector pointing upwards (against gravity), and \( p' \) represents the pressure differences from the hydrostatic value scaled with \( \rho g n \). The velocity vector \( \mathbf{v} \) is scaled with the characteristic velocity \( v_g = \beta^{-1} g n^2 / \nu \), resulting from a balance between the viscous and buoyancy forces, associated with density variations of order \( \rho_0 / \beta \). Introduction of this characteristic velocity into the definition of \( Ra \) shows that this Rayleigh number also can be interpreted as a Peclet number, \( v_g a / D T \). In this near-explosion regime, associated with small density differences of order \( \rho_0 / \beta \), the contribution of the temporal density variation in mass conservation gives a higher-order correction, of order \( (Ra) \), and therefore it does not appear in (4). In the equation for energy conservation, \( \gamma \), denoting the specific-heat ratio, is present in the term arising from the time derivative of the pressure.

The spatial variations of the pressure \( p' \) appearing in (5) are small compared with the uniform temporal variation of the pressure from \( p_o \), of order \( p_o = \rho_0 / \beta \), as given by \( \bar{p} = \beta (p - p_o) / p_o \), of order unity, required by the equation of state, with \( \bar{p} = \beta (p - p_o) / p_o \), to ensure, in its linearized form

\[
\dot{\bar{p}} + \bar{\phi} = \bar{q}(t)
\]  

that the spatially mean density is \( \rho_1 \), thus leading to the relation

\[
\bar{p} = \bar{\phi}
\]

between \( \bar{p} \) and the non-zero mean value \( \bar{\phi} \) of the temperature.

The slowly reacting mode of combustion accounting for buoyancy-induced motion is expected to have analytical solutions of (4)–(6) inside the vessel with boundary conditions \( \mathbf{v} = \phi = 0 \) at the vessel walls. Computation of the transient evolution requires specification of the initial conditions. For instance, for a stagnant gas mixture at temperature \( T_o \), with \( T_o - T_\infty \sim T_\infty / \beta \), the conditions at the initial instant, when the wall temperature is raised to \( T_o \), are simply given by \( \mathbf{v} = \phi = \phi_o = 0 \), where \( \phi_o = \beta (T_o - T_\infty) / T_\infty \). The analysis is somewhat more complicated when the wall-temperature rise is larger, such that \( T_o - T_\infty \sim T_\infty \), in which case the heating of the gas by heat conduction from the wall, which proceeds initially with negligible chemical reaction, involves relative variations of the density, temperature, and pressure of order unity. The initial conditions for (4)–(6) then must be obtained by matching with the distributions of temperature and velocity found at the end of this chemically frozen heating period, similar to the discussion in Part 1.

As discussed in [1] for the buoyancy-free case \( Ra = 0 \), the transient evolution depends fundamentally on the Damköhler number. Thus if \( Da \) is smaller than a critical value \( Da_c \) of \( Ra \), the temperature and velocity evolve to approach a steady distribution for moderately large values of \( t \), whereas for \( Da > Da_c (Ra) \) the transient stage ends with a thermal runaway at a finite ignition time. The explosion limits can be obtained either by computing the transient problem or, as proposed by Frank-Kamenetskii, by investigating the existence of steady solutions, the latter being the quantitative approach pursued below for the specific case of a spherical vessel.

It may be worth mentioning here that, for subcritical values of \( Da \leq Da_c (Ra) \), the above system of equations (4)–(6) can be used to describe the quasi-steady evolution of the temperature and velocity during the second much longer reactant-consumption stage occurring for \( t \sim \alpha \alpha \gg 1 \), where \( \alpha = (q Y_o) / (c_p T_\infty) \). As explained in [1], the reactant mass fraction in this stage is nearly uniform and appears only as a factor of \( Da \), equal to its spatially mean value, which evolves on the time scale...
2.2 Steady conservation equations in spherical vessels

For spherical vessels with axisymmetric flow, introduction of spherical coordinates with origin at the vessel center reduces the continuity equation (4) to

\[
\frac{\partial}{\partial r} \left( r^2 \sin \theta \nu_r \right) + \frac{\partial}{\partial \theta} \left( r \sin \theta \nu_\theta \right) = 0, \tag{9}
\]

where \( r \) is the radial distance and \( \theta \) is the angle measured with respect to the vertical direction \( e_z \). Introducing the stream function \( \psi \), such that

\[
\frac{\partial \psi}{\partial r} = r^2 \sin \theta \nu_r \quad \text{and} \quad \frac{\partial \psi}{\partial \theta} = -r \sin \theta \nu_\theta, \tag{10}
\]

reduces energy conservation (6) to

\[
\frac{Ra}{r^2 \sin \theta} \left( \frac{\partial \psi}{\partial \theta} \frac{\partial \phi}{\partial \theta} - \frac{\partial \psi}{\partial r} \frac{\partial \phi}{\partial r} \right) = \frac{1}{r^2 \sin \theta} \left( \frac{\partial \phi}{\partial r} \right)^2 + \frac{1}{r^2 \sin \theta} \left( \frac{\partial \phi}{\partial \theta} \right)^2 + Da e^\psi. \tag{11}
\]

The momentum equation can be written conveniently in terms of the vorticity, which is azimuthal, of magnitude \( \omega \), given in terms of the stream function by

\[
-r \sin \theta \omega = \frac{\partial^2 \psi}{\partial r^2} + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left( \frac{1}{\sin \theta} \frac{\partial \psi}{\partial \theta} \right). \tag{12}
\]

The vorticity distribution then is described by the equation

\[
\frac{Ra}{r^2 \sin \theta} \left( \frac{\partial \psi}{\partial \theta} \frac{\partial \phi}{\partial \theta} - \frac{\partial \psi}{\partial r} \frac{\partial \phi}{\partial r} \right) \left( r \sin \theta \omega \right) = \left[ \frac{\partial^2}{\partial r^2} + \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \left( \frac{1}{\sin \theta} \frac{\partial \psi}{\partial \theta} \right) \right] \left( r \sin \theta \omega \right) - r \sin^2 \theta \frac{\partial \psi}{\partial r} - \sin \theta \cos \theta \frac{\partial \phi}{\partial \theta} \tag{13}
\]

obtained by taking the curl of (5), where the temperature gradient serves as the source term. The boundary conditions for (11)–(13)

\[
\psi = \frac{\partial \phi}{\partial r} = \phi = 0 \quad \text{at} \quad r = 1 \quad \text{for} \quad 0 \leq \theta \leq \pi, \tag{14}
\]

correspond to a vessel with an isothermal wall with non-slip flow. The solutions, bounded and symmetric with respect to the vertical axis of the sphere, must be analytical, so that additional boundary conditions are

\[
\psi = \frac{\partial \phi}{\partial \theta} \left( \frac{1}{\sin \theta} \frac{\partial \psi}{\partial \theta} \right) = \frac{\partial \phi}{\partial \theta} = 0 \quad \text{at} \quad \theta = 0, \pi \quad \text{for} \quad 0 \leq r \leq 1. \tag{15}
\]
3. The Frank-Kamenetskii vortex

The temperature distribution \( \phi = \phi_\text{FK}(r) \) obtained from the Frank-Kamenetskii problem

\[
\frac{1}{\rho}(r^2 \phi_\text{FK})'' = -D\phi \, e^{\phi_\text{FK}}, \quad \phi_\text{FK}'(0) = \phi_\text{FK}(1) = 0, \tag{16}
\]

corresponds to the leading term in the perturbation analysis for small Rayleigh numbers. As shown in our accompanying paper [1] the solution can be determined most conveniently in terms of \( u = D\phi_\text{FK} \) and \( w = -r(\phi_\text{FK})'/(dr) \) with the temperature drop from its peak value at the center of the vessel \( \lambda = \phi_0 - \phi_\text{FK}(r) \) used as a new independent variable, such that

\[
r^2 = \frac{u(\lambda)e^\lambda}{D\phi_\text{FK}}. \tag{17}
\]

The spherically symmetric temperature field \( \phi = \phi_\text{FK}(r) \) generates a convective flow with toroidal vorticity. This F-K vortex, determined at leading order by the balance \( \nabla^2 v - \partial r = 0 \) between buoyancy and viscous forces, has a simple dependence on \( \theta \), given by

\[
\omega_\text{RFK} = -\sin \theta \, \Gamma_\text{RFK}/r \quad \text{and} \quad \psi_\text{RFK} = \sin^2 \theta \, F_\text{RFK}, \tag{18}
\]

the latter implying

\[
v_r = 2 \cos \theta F_\text{RFK}/r^2 \quad \text{and} \quad v_\theta = -\sin \theta F_\text{RFK}/r, \tag{19}
\]

with the radial variations \( \Gamma_\text{RFK}(r) \) and \( F_\text{RFK}(r) \), to be determined below. Thus, (12) and (13), for \( Ra = 0 \), reduce to the ordinary differential equations

\[
\Gamma_\text{RFK}'' - \frac{2}{r^2} \Gamma_\text{RFK} = w \tag{20}
\]

and

\[
F_\text{RFK}'' - \frac{2}{r^2} F_\text{RFK} = \Gamma_\text{RFK}, \tag{21}
\]

to be integrated with the boundary conditions

\[
|F_\text{RFK}(0)/r^2| \neq \infty \quad \text{and} \quad F_\text{RFK}(1) = F_\text{RFK}'(1) = 0, \tag{22}
\]

resulting from the boundedness of the velocity at the center of the vessel and from the no-slip condition at the wall.

Similar to the approach taken in [1] when solving (16), it is convenient to employ as variables \( \Gamma_\text{RFK} = \Gamma_\text{RFK}/r^2 \) and \( F_\text{RFK} = F_\text{RFK}/r^3 \), invariant under the radial dilatation used when defining \( w \), yielding the equations

\[
r^2 \Gamma_\text{RFK}'' + 4r \Gamma_\text{RFK}' = w, \tag{23}
\]

\[
r^2 F_\text{RFK}'' + 8r F_\text{RFK}' + 10 F_\text{RFK} = \Gamma_\text{RFK} \tag{24}
\]
The homogeneous equation associated with (23) has solutions proportional to \( r^3 \), which must be disregarded as incompatible with the boundedness condition stated in (25), because they would lead to solutions \( \tilde{F}_\kappa \propto 1/r^k \). Therefore, the general solution to (23) is of the form

\[
\tilde{F}_\kappa = \tilde{F}_\kappa(1) + \frac{\sigma}{w} (r - 1),
\]

where the constant \( \tilde{F}_\kappa(1) \), to be determined later using the condition \( \tilde{F}_\kappa(1) = 0 \), represents the value of \( \tilde{F}_\kappa \) at the center, and \( \tilde{F}_\kappa(1) \) is a particular solution satisfying \( \tilde{F}_\kappa(0) = 0 \). The latter can be obtained by defining \( \gamma(\lambda) = \tilde{F}_\kappa(\lambda) \) and \( \sigma(\lambda) = r \tilde{F}_\kappa(\lambda) \), with \( \lambda \) and \( r \) related by (17), and then writing (23) in the alternative form

\[
\frac{d\gamma}{d\lambda} = \frac{\sigma}{w} \quad \text{and} \quad \frac{d\sigma}{d\lambda} = \frac{w - 3\sigma}{w}, \quad \gamma(0) = \sigma(0) = 0.
\]

The numerical integration with the behaviors \( \gamma = \sigma/2 = \lambda/5 \) and \( w = 2\lambda \) for \( \lambda \ll 1 \) give results shown in Fig. 1, with other curves exhibited there to be derived and discussed later.

The analysis continues by discarding singular solutions of the form \( \tilde{F}_\kappa \propto 1/r^5 \) when solving the homogeneous problem associated with (24), so that the general
solution for the stream function reduces to

\[ \tilde{F}_{\infty} = \frac{v_o}{27/2} + \frac{\bar{p}_o}{10} + \bar{F}_{\infty}, \]  

(28)

where \( \bar{\Gamma}_{\infty} \) and \( \tilde{F}_{\infty}(r) \) are the particular solutions associated with \( \tilde{\Gamma}_v \) and \( \tilde{F}_{\infty} \), respectively. The first term in (28), arising from the solution to the homogeneous problem, represents a uniform vertical velocity, as can be seen by using \( \tilde{F}_{\infty} = r^2 \bar{F}_{\infty} = r^2v_0/2 \) to yield \( v_v = v_v \cos \theta \) and \( v_\theta = -v_v \sin \theta \). Since the other two terms in (28) give a vanishing velocity at \( r = 0 \), the constant \( v_v \) to be determined below from the no-slip condition at the wall, turns out to be the velocity induced at the center, which is the peak velocity in the vessel.

The function \( \tilde{F}_{\infty} \) can be determined by introducing \( f(\lambda) = \tilde{F}_{\infty}(r) \) and \( g(\lambda) = r \bar{F}_{\infty}(r) \) and integrating the autonomous problem

\[ \frac{df}{d\lambda} = \frac{g}{w} \quad \text{and} \quad \frac{dg}{d\lambda} = \gamma - 7g - 10f, \quad f(0) = g(0) = 0, \]

(29)

obtained by rewriting (24) with \( \lambda \) replacing the radial coordinate. Since \( \lambda = 0 \) is a singular point, the numerical integration must be initiated using the approximations \( 2f = g = \lambda/70 \) for \( \lambda \ll 1 \).

The functions \( f(\lambda) \) and \( g(\lambda) \), shown in Fig. 1, increase with \( \lambda \) and with the radial distance from the center, as dictated by (17). Their higher values at \( \lambda = \phi_o \), corresponding to \( r = 1 \), are given by \( f(\phi_o) = -(v_0/2 + \bar{\Gamma}_v/10) \) and \( g(\phi_o) = v_v \), as resulting from the boundary conditions \( \tilde{F}_{\infty}(1) = \bar{F}_{\infty}(1) = 0 \) and (28). Therefore, the functions \( f(\lambda) \) and \( g(\lambda) \) can be used to compute the variation with \( \phi_o \) of

\[ v_v = g(\phi_o) \quad \text{and} \quad \frac{\bar{\Gamma}_v}{10} = f(\phi_o) + \frac{1}{2}g(\phi_o), \]

(30)
given in Fig. 1. The range of values of \( \lambda \), or \( \phi_o \), considered extends beyond the first turning point of the explosion diagram, associated with the value \( \lambda = 1.607 \), marked in Fig. 1 with a vertical dotted line.

Once the values of \( \bar{\Gamma}_v \) and \( v_v \) corresponding to a given \( Da \) are identified from Fig. 1 (complemented with the curve \( Da(\phi_o) \) shown in Fig. 4 of the accompanying paper [1]), the functions \( \gamma(\lambda) \) and \( f(\lambda) \) can be used to obtain the functions

\[ \Gamma_{\infty}/r = r(\bar{\Gamma}_v + \Gamma_{\infty}) \quad \text{and} \quad F_{\infty} = r^2 \left( \frac{v_v}{27/2} + \frac{\bar{p}_o}{10} + \bar{F}_{\infty} \right), \]

(31)

with (17) employed to relate \( \lambda \) and \( r \). The resulting profiles are shown in Fig. 2(b) for five different values of \( \phi_o \) along the explosion curve of Fig. 2(a), including the critical turning point \( \phi_o = 1.607 \) (with \( Da = 3.322 \)), the points \( Da = 1 \) and \( Da = 2 \) along the lower branch, and the points corresponding to \( Da = 3 \) both below and above the turning point. The curves in Fig. 2(a) for the three non-zero Rayleigh numbers are to be discussed later.

The results can be used to evaluate the vorticity and stream function given in (18), with sample streamlines and isovorticity lines given in Fig. 3 for the critical case \( Da = Da_c = 3.322 \). A distinctive qualitative feature of the solution is that, as a result of geometrical effects of the spherical vessel, the resulting nondimensional values of the peak velocity \( v_v \), which are scaled with our original estimate \( v_v = \beta^{-2}g\sigma^2/\nu \) (resulting from the balance between viscous forces and buoyancy forces),
are very small, of the order of $10^{-2}$, as shown in Fig. 1. This unexpectedly slow motion has a weak effect on the temperature field through the convective transport of heat, leading to modifications of the explosion limits that remain small even for moderately large values of $Ra$ of order $10^2$, as found in numerical integrations of the original equations [8-10].

4. Effect of convection on the critical ignition conditions

4.1 Perturbation scheme

The small deviations from the F-K solution resulting from the presence of slow fluid motion for $Ra \ll 1$ can be addressed formally by expanding the different fluid variables in powers of $Ra$. The slow motion found at leading order, as represented by the small value $v_o \simeq 1/30$ for $Ra = 1.607$, is due to geometrical effects, not taken into account in the rough estimate $v_g \approx (g/\beta(\alpha^2/r))$ of the buoyant velocity. The resulting corrections to the explosion limits, although formally applicable only for small values of $Ra$, in fact become significant only for moderately large values of $Ra \sim 10^2$, and they remain accurate even for these large values, as occurs occasionally in perturbation expansions, although seldom ever to so great an extent.

Since the Rayleigh-number correction modifies the value of the critical Damkohler number above which F-K solutions no longer exist, from its value at zero Rayleigh number, in the vicinity of the critical point, it is inconvenient to seek solutions for $\phi(r)$ by specifying $Da$. Instead, it is better to prescribe the temperature at the center of the vessel $\phi_0$, as known and then pose the problem as that of finding the perturbed Damkohler number $Da$ that, for a given value of $Ra$, results

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2}
\caption{The functions $\Gamma_{FK}/r$ and $F_{FK}$ obtained from (31) for the conditions corresponding to the five points indicated along the F-K explosion curve on the left-hand side, where the thin curves represent the Damkohler-number prediction obtained from (51) for $Ra = (100, 200, 300)$ and the small dots represent the variation of the temperature at the vessel center obtained by integration of (11)-(14) for different values of $Da$.}
\end{figure}
in a temperature at the center of the vessel equal to $\phi_o$. The monotonic variation of $Dn$ with $\phi_o$, plotted in figure 4 of [1], shows that the leading-order solution exists for all values of $\phi_o$, so that the problem is well-posed when formulated in this manner.

In order to calculate the dependence $Dn(\phi_o, Ra)$ for fixed $\phi_o$ and small $Ra$, the expansions

$$Dn = Dn_{FK}[1 + Ra\delta_1 + Ra^2\delta_2 + O(Ra^3)]$$

and

$$\phi - \phi_{FK} = Ra\phi_1 + Ra^2\phi_2 + O(Ra^3)$$
$$\omega - \omega_{FK} = Ra\omega_1 + Ra^2\omega_2 + O(Ra^3)$$
$$\psi - \psi_{FK} = Ra\psi_1 + Ra^2\psi_2 + O(Ra^3)$$

are introduced. The terms in these expansions are determined by solving sequentially the different problems that arise at different orders in powers of $Ra$ when (32) and (33) are introduced into (11)–(13), as indicated below, with the condition $\phi_j = 0$ applied at all orders $j = 1, 2, \ldots$ for the temperature perturbations from $\phi_o$ at the center $r = 0$. Although a term proportional to $Ra$ is included in (32) for consistency with (33), it will be found that $\delta_1 = 0$, so that the corrections to the explosion curve $Dn(\phi_o)$ are of order $Ra^3$; they will be seen to be important only for $Ra \sim 10^4$ because of the result that $\delta_2 \approx 10^{-6}$.

### 4.2 First-order temperature perturbation

When the expansions (32) and (33) are used in (11), the terms of order $Ra$ yield a linear equation for $\phi_1$, to be integrated with boundary conditions $\phi_1 = 0$ at
The resulting equation includes perturbations proportional to \( \cos \theta \), arising from the convection term, along with perturbations independent of \( \theta \), that are proportional to the Damköhler-number perturbation \( \delta_1 \), consistent with a perturbed temperature of the form \( \phi_1 = \cos \theta H_1(r) + H_1(r) \). The function \( H_1 \), however, and its accompanying Damköhler-number perturbation \( \delta_1 \), which are determined from the problem

\[
r^2 H''_1 + 2r H'_1 + u(H_1 + \delta_1) = 0, \quad H_1(0) = H_1(1) = 0, \quad (34)
\]

derived from (11) and (14), must be identically zero. This can be seen readily by comparing the above equation with

\[
r^2 u'' + 2ru' + u(w - 2) = 0, \quad (35)
\]

equivalent to the autonomous system giving \( u \) and \( w \) in terms of \( r \) in (22) of [1]. Specifically, since \( H_1 + \delta_1 \) and \( w - 2 \) satisfy the same second-order linear equation, the solution to (34) can be expressed in general as

\[
H_1 + \delta_1 = C_A Y_A + C_B Y_B, \quad (36)
\]

involving two integration constants \( C_A \) and \( C_B \) and the two independent solutions

\[
Y_A = w - 2 \quad \text{and} \quad Y_B = (w - 2) \left( \int_0^r \frac{w(4 - w)}{r^2(w - 2)^2} \frac{dr}{r} \right), \quad (37)
\]

of (35). Because of the term \( 1/r \), the function \( Y_B \) is singular at the origin, so that the condition \( H_1(0) = 0 \) requires that \( C_B = 0 \), and also determines the value \( C_A = -\delta_1/2 \), as follows from (36) with \( H_1(0) = 0 \) and \( Y_A(0) = w(0) - 2 = -2 \). The solution therefore reduces to \( H_1 = -\delta_1 u/2 \). Since, however, \( w \) is positive for \( r > 0 \) (see Fig. 2 of [1]), the boundary condition \( H_1 = 0 \) at \( r = 1 \) can be satisfied only if \( \delta_1 = H_1 = 0 \). This completes the proof. With \( \delta_1 = 0 \), the perturbations to the Damköhler number emerge only at the following order in the expansion (32).

The resulting form of the temperature perturbation, \( \phi_1 = \cos \theta H_1(r) \), indicates that at this order the presence of convection does not modify the mean temperature in the vessel, nor does it perturb the total heat-loss rate to the wall. Instead, the main effect of buoyancy is that of breaking the spherical symmetry of the problem, which was observed to be broken in previous numerical studies [13], causing the appearance of regions of hotter fluid in the upper half of the vessel and, correspondingly, regions of colder fluid in the lower half. The associated function \( H_1 \) is determined from the problem

\[
\frac{H''_1}{r} + \frac{2H_1}{r^2} + (u - 2) \frac{H_1}{r^2} = -2w \left( \frac{v_0/2}{r^2} + \frac{\Gamma_w}{10} + \tilde{F}_{\infty} \right), \quad H_1(0) = H_1(1) = 0, \quad (38)
\]

the solution to which is given in general by the sum of a linear combination of the solutions to the homogeneous problem and three particular solutions, associated with the three terms on the left-hand side of (38). As can be seen by inspection, in view of (35), the particular solution associated with \( v_0 \) is given simply by \( H_1 = -(2 - w/2)v_0r \). The other two particular solutions, associated with the terms involving \( \Gamma_w \) and \( \tilde{F}_{\infty} \), can be expressed more conveniently in terms of alternative variables \( \tilde{H}_c \) and \( \tilde{H}_v \), defined from \( H_1 = -(\tilde{v}_c/10)r^2 \tilde{H}_c \) and \( H_1 = -r^3 \tilde{H}_v \), which
are introduced to preserve the invariance of (38) under a radial dilatation. The general procedure followed above then leads in this case to the problems

$$\frac{dh_x}{d\lambda} = \frac{k_\lambda}{w} \quad \text{and} \quad \frac{dk_y}{d\lambda} = -\frac{7k_x + (u + 10)h_x - 2w}{w}, \quad h_x(0) = k_y(0) = 0 \quad (39)$$

and

$$\frac{dh_x}{d\lambda} = \frac{k_x}{w} \quad \text{and} \quad \frac{dk_y}{d\lambda} = -\frac{7k_x + (u + 10)h_x - 2w}{w}, \quad h_x(0) = k_y(0) = 0, \quad (40)$$

where $h_x(\lambda) = \tilde{H}_x(r)$, $h_x(\lambda) = r\tilde{H}_x(r)$, $k_x(\lambda) = \tilde{K}_x(r)$, and $k_x(\lambda) = r\tilde{K}_x(r)$, respectively. Near the origin, which is a singular point, the asymptotic behaviors $2h_x = k_x = 2x/7$ and $4h_x = k_x = 2x^2/945$ arise, which must be accommodated by the numerical solutions of (39) and (40).

The homogeneous equation associated with (38) admits diverging solutions $H_1 \propto r^{-2}$ near the origin, incompatible with the boundary condition $H_1(0) = 0$, along with linearly increasing solutions $H_1 \propto r$, which can be described more conveniently in terms of $H_1 = rH'_1$, where $H'_1(r)$ satisfies $r^2H'_1 + 4rH''_1 + uH'_1 = 0$. The problem reduces to that of integrating, for $h(\lambda) = \tilde{H}_1(r)$ and $k(\lambda) = r\tilde{K}_1(r)$, the equations

$$\frac{dh}{d\lambda} = \frac{k}{w} \quad \text{and} \quad \frac{dk}{d\lambda} = -\frac{3k + uh}{w} \quad (41)$$

with the boundary values near the origin evaluated from

$$h = 1 - \frac{3}{5}\\lambda + \cdots \quad \text{and} \quad k = -\frac{6}{5}\\lambda + \cdots \quad \text{at} \quad \lambda \ll 1, \quad (42)$$

the convenient normalization of $h$ having been selected to be unity at $r = 0$.

The solution to (38), constructed by collecting the different contributions listed above, reads

$$H_1 = r \left[ (h_0 + 2v_0)\tilde{H}_1 - \left( 2 - \frac{w}{2} \right) v_y \right] - r^3 \left( \frac{\pi}{10} \tilde{K}_x + \tilde{K}_y \right), \quad (43)$$

where the integration constant appearing as a factor of the homogeneous solution has been written in terms of $h_0 = \tilde{H}_1(0)$, the magnitude of the temperature gradient at the center. Its value can be evaluated for a given $\phi_0$ from

$$h_0 = -2g(\phi_0) + \left( 2 - \frac{w}{2} \phi_0 \right) \frac{g(\phi_0)}{h(\phi_0)} \left( f(\phi_0) \right) \frac{g(\phi_0)}{h(\phi_0)} + \frac{h_x(\phi_0)}{h(\phi_0)} \quad (44)$$

obtained by using the boundary condition $H_1 = 0$ at $r = 1$ in (43). As expected, the resulting value, plotted in Fig. 1, is found to be quantitatively small in the range of values of $\phi_0$ corresponding to the first turning point of the explosion curve.

4.3 Modified explosion curve

As discussed above, corrections to the Damköhler number emerge only at order $Re^2$. The factor $S_2$ in the expansion (32) is obtained from consideration of higher-order terms in the expansions (33). The vorticity and stream-function perturbations
take the form
\[ \omega_1 = -\sin \theta \cos \theta \Gamma_1/r \quad \text{and} \quad \psi_1 = \sin^2 \theta \cos \theta F_1, \]

in terms of functions \( \Gamma_1(r) \) and \( F_1(r) \), as follows when collecting terms of order \( R_0 \) in (12) and (13). Similarly, the temperature perturbation \( \phi_2 \) can be seen from inspection of (11) to involve two different terms, according to \( \phi_2 = \cos^2 \theta H_2 + \mathcal{H}_2 \).

The functions \( H_2(r) \) and \( \mathcal{H}_2(r) \) satisfy
\[ r^2 H_2'' + 2r H_2' + (u - 6) H_2 = 2 F_{\nu_0} H_1' - F_{\nu_0} H_1 - 3 F_1 w/r - \frac{1}{2} u H_1^2 \]

and
\[ r^2 H_2'' + 2r H_2' + u H_2 = -\delta_2 u - 2 H_2 + F_{\nu_0} H_1 + F_1 w/r, \]

subject to \( H_2(0) = H_2(1) = \mathcal{H}_2(0) = \mathcal{H}_2(1) = 0 \).

The above homogeneous problem determines \( \delta_2 \) as an eigenvalue. Although \( F_1 \) appears in both equations, the resulting value of \( \delta_2 \) is independent of the corrections to the fluid motion (and, therefore, independent of the Prandtl number). This can be seen by considering
\[ r^2 (H_2 + \frac{1}{3} H_2)^2 + 2r (H_2 + \frac{1}{3} H_2) + u (H_2 + \frac{1}{3} H_2 + \delta_2) = \mathcal{F}, \]

obtained from a linear combination of (46) and (47), with \( \mathcal{F} = \frac{2}{3} (F_{\nu_0} H_1') - \frac{1}{2} u H_1^2 \).

As can be inferred from (35), the general solution to (48) can be expressed in the form
\[ H_2 + \frac{1}{3} H_2 + \delta_2 = B_A Y_A + B_B Y_B + \frac{1}{3} Y_A \int_0^r Y_B F dr - \frac{1}{3} Y_A \int_0^r Y_B F dr, \]

involving the integration constants \( B_A \) and \( B_B \) and the independent solutions \( Y_A \) and \( Y_B \) of the homogenous equation, given in (37). Since the boundary conditions \( \mathcal{H}_2(0) = \mathcal{H}_2(0) = 0 \) suffice to determine the value of the two integration constants \( B_A = -\delta_2/2 \) and \( B_B = 0 \), the additional boundary conditions \( \mathcal{H}_2(1) = \mathcal{H}_2(1) = 0 \) require that
\[ \delta_2 = \frac{(w - 2)}{2w} \left[ \left( \int_0^w \frac{(w - 4) - w}{(w - 2)^3} dr - \frac{1}{3} \int_0^w \frac{2}{(w - 2)^3} dr \right) \right]^2 \]

\[ \int_0^w \frac{(w - 4) - w}{(w - 2)^3} dr - \frac{1}{3} \int_0^w \frac{2}{(w - 2)^3} dr \]

obtained by evaluating (49) at \( r = 1 \). The resulting value of \( \delta_2 \) is a function of \( \phi_0 \), shown in Fig. 1.

The function \( \delta_2(\phi_0) \) can be used to evaluate the modified explosion curve
\[ D_a = D_{\text{aw}} (1 + R_0^2 \delta_2), \]

where \( D_{\text{aw}}(\phi_0) = w(\phi_0) \), given by \( u(\lambda) \) for \( \lambda = \phi_0 \). Because of the very small value \( \delta_2 \approx 10^{-6} \) encountered in the perturbation analysis in powers of \( R_0 \), important perturbations in the critical Damköhler number occur only for values of the
Rayleigh number of order $10^3$. Considerations of (29), (39), and (40) suggest that the smallness of the factor $\delta_3 \sim 10^{-6}$ is a cumulative result of the sequence of ordinary differential equations, each with numerical coefficients of order 10, involved in its computation.

The accuracy of the prediction (51) is illustrated in Fig. 2(a), which includes comparisons with results of numerical integrations of the initial problem (11)–(14) for $Ra = 100$, 200, and 300, shown as points along the curves. The numerical procedure employed a pseudo-transient method in seeking convergence to a steady solution, which prevented the branch of unstable solutions found beyond the first turning point from being accessible in the numerical integrations, so that the numerical results are limited to the lower branch of solutions extending from $Da = 0$ to $Da = Da_c$. Over the range that these numerical results are obtained, they differ negligibly from the results of the expansion, as may be seen by comparing the points with the thin curves in the figure. The corrections at the first non-vanishing order to the F-K result for $Da_c$ are seen to amount to less than ten percent even at $Ra = 300$. The range of the Rayleigh number over which the expansion remains valid thus is very large.

An unexpected peculiarity of the curve of $\delta_3$ as a function of $\phi_o$, shown in Fig. 1, is the attainment of a maximum, followed by a progression to negative values, beyond the turning-point value marked by the vertical line. These negative values imply that the motion of the fluid inside the sphere under those conditions, instead of decreasing the peak temperature by increasing the rate of heat loss to the boundary, tends on the average to insulate the central fluid from conductive heat loss, decreasing temperature gradients to the sides and bottom there, thereby increasing the peak temperature. Although reminiscent of the related effect in planar geometry mentioned earlier, the fluid-dynamic cause of this is quite different. Because the flow computed here in the region beyond the vertical line in the figure is unstable, however, this peculiarity would not be observed experimentally.

5. Concluding remarks

The present analysis has described effects of buoyancy on the slowly reacting mode of combustion in spherical vessels. The motion induced for small values of the Rayleigh number is an axisymmetric vortex termed here the Frank-Kamenetskii vortex—that also exhibits symmetry about the equatorial plane, with positive vorticity generated in the central region by the finite-rate chemical heating but also with a region of negative vorticity near the wall, where the viscous retardation of the motion by the wall reverses the sign of the vorticity. At the leading order in the Rayleigh number, developed here, the zero-vorticity surface is perfectly spherical. At much higher Rayleigh numbers, this downward gas motion in the vicinity of the walls will generate a boundary layer there, growing with distance from the top, distorting the zero-vorticity sphere, so that the vortex tends to produce temperature stratification in the core, with hotter fluid in the upper portion of the sphere, as has been seen in numerical computations. The resulting modifications of the F-K vortex may be expected to affect the criticality conditions for ignition, which motivates pursuit of associated boundary-layer analyses in the future.

Especially noteworthy of the present results is how remarkably small the fluid-flow influences associated with the Frank-Kamenetskii vortex are on the critical ignition Damköhler number (the value beyond which the slow-reaction types of solutions derived here no longer exist). Despite the gas motion, the spatial variations average out in such a way that the overall effects remain small. This is not
an asymptotic phenomenon; rather, it is a purely numerical influence of the small numerical coefficients arising in the expansions. For example, as may be seen in Fig. 1, even though the nondimensional flow velocities are roughly of order $10^{-2}$, the nondimensional modification of the temperature gradient is only on the order of $10^{-3}$, resulting in the change in the critical ignition Damköhler number, a second-order quantity, being only of order $10^{-6}$, seen in the figure. Furthermore, while the leading-order corrections to the flow field of the Frank-Kamenetskiě vortex do depend on the Prandtl number, even at the second order calculated here, the corrections to the critical ignition Damköhler number do not. This underscores the fact that the Rayleigh number, not the Grashof number, is the appropriate parameter of expansion.

The present analysis pertains only to the classical one-step Arrhenius heat-release description underlying the F-K theory. Extensions of the early F-K theory, however, incorporating realistic chemistry in descriptions of hydrogen-oxygen systems, have been shown to predict explosion conditions in spherical vessels in excellent agreement with experiments [14], including critical pressures along the so-called third explosion limit [15]. Possible influences of buoyant fluid motion on the resulting critical ignition conditions have not been addressed. Corrections accounting for buoyancy effects could be incorporated in these analyses following the general procedure used here, accounting for the significant differences in the chemical-kinetic descriptions. In the absence of such studies, it remains unclear how significant the influences of the buoyant fluid motion will be on ignition in systems with real chemical kinetics, although it may be conjectured that the influence on the third limit, which retains a thermal-explosion character, may be comparable with what has been found here, while for the second limit, the branched-chain explosion, the influence may be larger.

In addition to the future worthwhile investigations of different fluid-mechanic and chemical-kinetic aspects of problems of this kind, as indicated above, it would also be of interest to address different geometrical configurations, other than the sphere. For example, the problem that has been analyzed by the present expansion procedure has not yet been considered in cylindrical geometry. Related ignition experiments have, in fact, been performed with gases contained in long cylindrical tubes, and an analysis of the present type may well be even simpler in cylindrical coordinates. Concerning the idealized reactive slab bounded by two isothermal infinite horizontal walls, which has been considered in a number of previous theoretical analyses [4–7], the resulting F-K vortices could be investigated for different convective cells, including also rectangular and hexagonal shapes. These are just some of the many possible future investigations along these lines that could be revealing.

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References


