

concealed much of the fecal material. As many as 622 seeds were recorded in a single fox scat, indicating that foxes consume sufficient seeds of autumn olive to be important in seed dispersal. Our findings are in agreement with Yearsley and Samuel (1980), who found that fox scats deposited on surface mines during October were composed almost entirely of autumn olive seeds. Using radio telemetry they also located gray foxes on mine habitat planted to autumn olive during 18 and 25 percent of the daytime and nighttime attempts, respectively.

TABLE 1. Autumn olive seeds removed from fox scats collected from the Ollis Creek surface mine from November 1979 to April 1980.

Months 1979-1980	No. of Scats	Mean No. of Seeds/Scat/Month	Standard Deviation	Range in No. of Seeds/Scat
November	35	218.0	±144	0 to 583
December	41	196.0	±118	0 to 447
January	20	174.0	±180	0 to 622
February	3	22.0	± 29	0 to 056
March	15	182.0	±128	0 to 363
April	3	0.3	± 1	0 to 001

Because both red and gray foxes have large home ranges (Ables, 1969; Sheldon, 1950), seed-laden scats may potentially be deposited several miles from the point of origin. However, Yearsley and Samuel (1980) calculated home range sizes of 75 to 125 hectares for gray foxes utilizing surface mines in West Virginia and concluded that use of the mines was highest in fall and lowest in summer. Our findings suggest that autumn olive seed dispersal by foxes

would tend to be relatively localized. Some of the other animals that consume autumn olive berries contribute to the dispersal of seeds.

Autumn olive shrubs begin fruit production the second growing season after planting, and dispersal of the plant to other portions of the mine by animals could begin at that time. Our study did not test seed-laden fox scats for germination potential under natural conditions. However, large numbers of seedling autumn olive shrubs that were offspring of the original plantations have been observed on the Ollis Creek mine, indicating that autumn olive seeds did successfully germinate on surface mines. Consequently establishment of even small numbers of autumn olive shrubs on coal surface mines may significantly increase the availability of food and cover and greatly improve the quality of habitat for wildlife.

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THERMAL RUNAWAY IN COAXIAL CHEMICAL REACTORS

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ABSTRACT

Thermal runaway in coaxial cylindrical chemical reactors is examined theoretically. The equation governing the temperature distribution is linearized and solved in the steady state case and the conditions required for the solution to become infinite (thermal explosion) are ascertained. A Green's function method is used to obtain thermal runaway conditions in coaxial tubular reactors of finite length. Results and limitations of the calculations are discussed.

INTRODUCTION

It is common practice in chemical kinetics to assume that the temperature of the reaction mixture in a vessel is equal to the temperature of the thermostat in which the vessel is immersed. This, of course, cannot possibly be true for reactions taking place at a finite rate and having a non-zero value for ΔE , since a temperature gradient is necessary to cause heat to flow from the reaction mixture to the thermostat (in the case of exothermic reactions) or vice versa. Benson has explored this phenomenon in some

detail for the case in which the reaction vessel is spherical and the temperature differences are small enough that the rate of the reaction can be regarded as unaffected by them (Benson 1954 and Benson, 1960). We then carried out an analysis of temperature gradients in cells of spherical, slab, infinite cylindrical, and finite cylindrical geometries for reactions for which the temperature dependence of the rate must be taken into account (Wilson 1958). In these analyses it is assumed that heat transfer takes place by conduction only—that convection can be neglected. The general differential equation governing temperature gradients in such reacting systems is given in Hirschfelder, Curtiss, and Bird (1954). Benson (1954) and Benson and Axworthy (1957) found this approach to be in good agreement with data on ozone and azomethane decompositions.

Work on thermal explosions in chemical reactors pre-dates these studies on temperature gradients, and is summarized in Benson's book (1960). The first worker to address this problem was Semenov (1935). Frank-Kamenetskii (1955) and Rice (1940) examined the differential equation governing the temperature in reacting mixtures to determine the conditions under which the solutions to the steady-state problem fail to exist and one has an explosion; they examined spherical and infinitely long cylindrical cells. The Russian work has been summarized by Kondratiev (1964).

We address here the problem of calculating the conditions for thermal runaway in cylindrical reactors having a cooling tube located coaxially in the reactor and maintained at the temperature of the thermostat surrounding the reactor. Such geometry permits the construction of cylindrical reactors of larger capacity than would be possible if a coaxial cooling tube is not used and the reaction may undergo thermal runaway. We consider only heat transfer by conduction.

ANALYSIS

The differential equation governing the temperature in a reacting mixture in which a second-order reaction is occurring and heat transfer is by conduction only is given by

$$c \frac{\partial T}{\partial t} = KV^2T - \Delta E \cdot A \exp\left(\frac{E_a}{RT}\right) c_1 c_2 \quad (1)$$

where

c = specific heat of reaction mixture

T = temperature

t = time

K = thermal conductivity

ΔE = energy change of the reaction per mole of reactant 1 consumed

$$A \exp\left(\frac{E_a}{RT}\right) c_1 c_2 = -\partial c_1 / \partial t$$

A = rate constant pre-exponential factor, $\text{cm}^3 \text{mole}^{-1} \text{sec}^{-1}$

E_a = activation energy of reaction

c_1 = concentration of reactant 1, moles/ cm^3

c_2 = concentration of reactant 2

T_0 = temperature of thermostat

We define a new dependent variable,

$$\tau = T - T_0$$

which converts Eq. (1) into

$$c \frac{\partial \tau}{\partial t} = KV^2\tau - \Delta E \cdot A c_1 c_2 \exp\left[\frac{-E_a}{RT_0(1 + \tau/T_0)}\right] \quad (2)$$

We then expand the exponential in ascending powers of τ , and keep terms only

through first order in τ ; this yields

$$c \frac{\partial \tau}{\partial t} = KV^2\tau - \Delta E \cdot A c_1 c_2 \exp\left(\frac{-E_a}{RT_0}\right) \frac{E_a}{RT_0^2} \tau$$

$$- \Delta E A c_1 c_2 \exp\left(\frac{-E_a}{RT_0}\right) \quad (3)$$

Let

$$\alpha = -\Delta E A c_1 c_2 \exp\left(\frac{-E_a}{RT_0}\right) K^{-1}$$

$$\beta = -\Delta E A c_1 c_2 \exp\left(\frac{-E_a}{RT_0}\right) \frac{E_a}{RT_0^2} K$$

and assume that c_1 and c_2 do not change significantly during the time period of interest. We shall also look for the steady state solution, for which $\partial \tau / \partial t = 0$; this gives

$$0 = \nabla^2 \tau + \beta \tau + \alpha \quad (4)$$

We consider an annular reactor of inner radius r_1 , outer radius r_2 , and length ℓ , so that $r_1 \leq r \leq r_2$ and $-\ell/2 \leq z \leq \ell/2$. Our boundary conditions are

$$\tau(r_1, z) = \tau(r_2, z) = \tau(r, -\ell/2) = \tau(r, \ell/2) = 0 \quad (5)$$

We digress here briefly to consider the case where ℓ is sufficiently large that we can treat our reactor as infinitely long. Eq. (4) then becomes

$$0 = \frac{1}{r} \frac{d}{dr} r \frac{d\tau}{dr} + \beta \tau + \alpha \quad (6)$$

The substitution $\beta r + \alpha = \beta R$ then yields

$$0 = \frac{1}{r} \frac{d}{dr} r \frac{dR}{dr} + \beta R \quad (7)$$

and we have as boundary conditions

$$R(r_1) = \alpha/\beta \quad (8)$$

$$R(r_2) = \alpha/\beta$$

Eq. (7) can be rewritten as

$$r^2 \frac{d^2 R}{dr^2} + r \frac{dR}{dr} + \beta r^2 R = 0 \quad (9)$$

which is Bessel's equation of order zero, with solution

$$R(r) = g J_0(\beta^{1/2} r) + h Y_0(\beta^{1/2} r) \quad (10)$$

where J_0 and Y_0 are Bessel functions of the first and second kind, as described in various standard texts (10). This solution must satisfy the boundary conditions, Eqs. (8), which requirement yields

$$\frac{g}{\beta} = g J_0(\beta^{1/2} r_1) + h Y_0(\beta^{1/2} r_1) \quad (11)$$

$$\frac{h}{\beta} = g J_0(\beta^{1/2} r_2) + h Y_0(\beta^{1/2} r_2)$$

The solutions to this set of equations are

$$g = \frac{\alpha}{\beta} \begin{vmatrix} 1 & Y_0(\beta^{1/2} r_1) \\ 1 & Y_0(\beta^{1/2} r_2) \end{vmatrix} \begin{vmatrix} J_0(\beta^{1/2} r_1) & Y_0(\beta^{1/2} r_1) \\ J_0(\beta^{1/2} r_2) & Y_0(\beta^{1/2} r_2) \end{vmatrix}^{-1}$$

$$h = \frac{\alpha}{\beta} \begin{vmatrix} J_0(\beta^{1/2} r_1) & 1 \\ J_0(\beta^{1/2} r_1) & 1 \end{vmatrix} \begin{vmatrix} J_0(\beta^{1/2} r_1) & Y_0(\beta^{1/2} r_1) \\ J_0(\beta^{1/2} r_2) & Y_0(\beta^{1/2} r_2) \end{vmatrix}^{-1} \quad (12)$$

These coefficients determine $R(r)$, from which we can calculate $T(r)$ by the relationship

$$T(r) = T_0 - (\alpha/\beta) + R(r) \quad (13)$$

Let us next examine the condition for thermal runaway and explosion. Evidently under such circumstances there can be no steady-state solution to Eq. (3), so the expressions for g and/or h must become singular. This requires that the denominators in Eq. (12) vanish, which yields

$$\begin{vmatrix} J_0(\beta^{1/2} r_1) & Y_0(\beta^{1/2} r_1) \\ J_0(\beta^{1/2} r_2) & Y_0(\beta^{1/2} r_2) \end{vmatrix} = 0 \quad (14)$$

as the condition for explosion to occur. For fixed β and r_2 we take the largest value of $r_1 < r_2$ which satisfies Eq. (14) as giving us the smallest volume per unit length of a reactor of outer radius r_2 which would exhibit thermal runaway under the reaction conditions specified by the parameters making up β .

We return now to consider reactors of finite length ℓ . A Green's function technique is well-adapted to establish the condition for thermal runaway. The partial differential equation, Eq. (4), becomes

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \tau}{\partial r} \right) + \frac{\partial^2 \tau}{\partial z^2} + \beta \tau = -\alpha \quad (15)$$

We shall construct the bilinear expansion of the Green's function for this equation in terms of the eigenfunctions of the homogeneous equation, ϕ_λ , satisfying and boundary conditions identical to those given in Eqs. (5).

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \phi_\lambda}{\partial r} \right) + \frac{\partial^2 \phi_\lambda}{\partial z^2} + \beta \tau = -\alpha \quad (16)$$

and boundary conditions identical to those given in Eqs. (5).

This equation can be solved in the usual way by separation of variables; one assumes that $\phi_\lambda = R(r)Z(z)$ and substitutes in Eq. (16) to get, after division by RZ ,

$$\frac{1}{rR} \frac{d}{dr} (rR) + \frac{1}{z} \frac{d^2 Z}{dz^2} + \lambda = 0 \quad (17)$$

This splits into an equation for Z ,

$$Z'' + \lambda_2 Z = 0 \quad (18)$$

and one for R ,

$$\frac{d}{dr} \left(r \frac{dR}{dr} \right) + (\lambda - \lambda_2) r R = 0 \quad (19)$$

The symmetry of the problem dictates that Z be an even function of z , which yields

$$Z = B \cos \sqrt{\lambda_2} z \quad (20)$$

The boundary conditions require that

$$\cos(\sqrt{\lambda_2} \ell/2) = 0 \quad (21)$$

which yields

$$\lambda_2 = [(2n-1)\pi/\ell]^2, \quad n = 1, 2, \dots \quad (22)$$

The radial equation rearranges to

$$r^2 \frac{d^2 R}{dr^2} + r \frac{dR}{dr} + (\lambda - \lambda_2) r^2 R = 0 \quad (23)$$

This is Bessel's equation of order zero, which has as its general solution

$$R = AJ_0(\gamma^{1/2} r) + BY_0(\gamma^{1/2} r), \quad \gamma = \lambda - \lambda_2 \quad (24)$$

Our boundary conditions require that

$$R(r_1) = R(r_2) = 0$$

so we must have

$$\begin{aligned} 0 &= AJ_0(\gamma^{1/2} r_1) + BY_0(\gamma^{1/2} r_1) \\ 0 &= AJ_0(\gamma^{1/2} r_2) + BY_0(\gamma^{1/2} r_2) \end{aligned} \quad (25)$$

Non-trivial solution of these equations requires that

$$\begin{vmatrix} J_0(\gamma^{1/2} r_1) & Y_0(\gamma^{1/2} r_1) \\ J_0(\gamma^{1/2} r_2) & Y_0(\gamma^{1/2} r_2) \end{vmatrix} = 0 \quad (26)$$

This equation determines the allowed values of $\gamma_1, \gamma_2, \gamma_3, \dots, \gamma_m, \dots$.

The eigenvalues are then given by

$$\lambda = \lambda_{mn} = \gamma_m + [(2n-1)\pi/\ell]^2, \quad (27)$$

$$m = 1, 2, \dots$$

$$n = 1, 2, \dots$$

Henceforth we append subscripts to A and B , and, from Eq. (25), note that

$$B_{mn} = -A_{mn} J_0(\gamma_m^{1/2} r_1) / Y_0(\gamma_m^{1/2} r_1) \quad (28)$$

so

$$\phi_\lambda = \phi_{mn} = A_{mn} \left\{ J_0(\gamma_m^{1/2} r) - [J_0(\gamma_m^{1/2} r_1) / Y_0(\gamma_m^{1/2} r_1)] Y_0(\gamma_m^{1/2} r) \right\} \cos \left[(2n-1)\pi z / \ell \right] \quad (29)$$

We are now in position to construct the Green's function for our problem. The differential equation for the Green's function is

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial G}{\partial r} \right) + \frac{\partial^2 G}{\partial z^2} + \beta G = \delta(r-r') \delta(z-z') \quad (30)$$

where

$$\delta(x) = 0, \quad x \neq 0$$

$$\int_{-q}^q \delta(x) dx = 1, \quad q \neq 0$$

In the usual way (Butkov, 1968) we write G as a series in the eigenfunctions

$$G(r, z | r', z') = \sum_m \sum_n C_{mn}(r', z') \phi_{mn}(r, z) \quad (31)$$

This is substituted into Eq. (30), which, when we note that

$$\frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial \phi_{mn}}{\partial r} \right) + \frac{\partial^2 \phi_{mn}}{\partial z^2} = \lambda_{mn} \phi_{mn} \quad (32)$$

yields

$$\sum_m \sum_n (B - \lambda_{mn}) \phi_{mn}(r, z) C_{mn}(r', z') = \delta(r-r') \delta(z-z') \quad (33)$$

The eigenfunctions ϕ_{mn} can be shown to constitute an orthogonal set in the usual way, as follows.

$$\nabla^2 \phi_i = -\lambda_i \phi_i, \quad i, m, n \quad (34)$$

$$\nabla^2 \phi_j = -\lambda_j \phi_j \quad (35)$$

Multiply Eq. (34) by ϕ_j , Eq. (35) by ϕ_i , and integrate over the volume of the reactor to get

$$\int_0^{2\pi} \int_{r_1}^{r_2} \int_{-l/2}^{l/2} \phi_a \nabla^2 \phi_i dz r dr d\theta = -\lambda_i \iiint \phi_j \phi_i dz r dr d\theta \quad (36)$$

$$\iiint \phi_i \nabla^2 \phi_j dz r dr d\theta = -\lambda_j \iiint \phi_i \phi_j dz r dr d\theta \quad (37)$$

We subtract Eq. (37) from Eq. (36) and note that Green's Theorem,

$$\begin{aligned} \iiint_V \nabla \phi_i \cdot \nabla \phi_j dV &= - \iiint_V \nabla^2 \phi_j \phi_i dV \\ &+ \iint_{S(V)} \nabla \phi_i \cdot d\vec{S} \end{aligned} \quad (38)$$

permits us to write

$$\begin{aligned} &\iiint_V (\phi_j \nabla^2 \phi_i - \phi_i \nabla^2 \phi_j) dz r dr d\theta \\ &= - \iiint_V (\nabla \phi_j \cdot \nabla \phi_i - \nabla \phi_i \cdot \nabla \phi_j) dz r dr d\theta \\ &+ \iint_{S(V)} (\phi_j \nabla \phi_i - \phi_i \nabla \phi_j) \cdot d\vec{S} \end{aligned} \quad (39)$$

The first integral on the right has zero for its integrand; the boundary conditions require that ϕ_i and ϕ_j vanish on $S(V)$, so the surface integral also vanishes, leaving us with the desired result,

$$(\lambda_i - \lambda_j) \iiint_V \phi_i \phi_j dz r dr d\theta = 0 \quad (40)$$

We choose the λ_{mn} such that

$$\iiint_V \phi_{mn}^2(r, z) r dr dz = 1 \quad (41)$$

Next let us return to Eq. (33), multiply both sides of the equation by $\phi_{pq}(r, z)r$, and integrate over the reactor volume. This yields

$$(B - \lambda_{pq}) C_{pq}(r', z') = r \phi_{pq}(r, z) \quad (42)$$

on utilizing the orthonormality of the eigenfunctions and the properties of the δ -function, so

$$C_{pq}(r', z') = \frac{r' \phi_{pq}(r', z')}{B - \lambda_{pq}} \quad (42)$$

and we obtain for $G(r, z | r', z')$

$$G = \sum_m \sum_n \frac{\phi_{mn}(r, z) r' \phi_{mn}(r', z')}{B - \lambda_{mn}} \quad (43)$$

Making use of the solving property of the Green's function yields

$$\tau(r, z) = \int_{r_1}^{r_2} \int_{-l/2}^{l/2} \sum_m \sum_n \frac{\phi_{mn}(r, z) \phi_{mn}(r', z') dz' r' dr'}{B - \lambda_{mn}} \quad (44)$$

Evidently the steady state solution does not exist when $B = \lambda_{mn}$, $m = 1, 2, \dots$; $n = 1, 2, \dots$. The first singularity occurs when $B = \lambda_{11}$, which gives

$$(-\Delta E) A c_1 c_2 \exp\left(\frac{-E_a}{RT_0}\right) \frac{E_a}{kRT_0^2} = \gamma_1 + \left(\frac{\pi}{\ell}\right)^2 \quad (45)$$

where γ_1 is the smallest root of

$$\begin{vmatrix} J_0(\gamma_1^2 r_1) & Y_0(\gamma_1^2 r_1) \\ J_0(\gamma_1^2 r_2) & Y_0(\gamma_1^2 r_2) \end{vmatrix} = 0 \quad (46)$$

Eq. (45) therefore gives the condition for thermal runaway and explosion.

We note that as $l \rightarrow \infty$, $\gamma_1 \rightarrow \beta$, so that Eq. (46) and Eq. (14) become identical in this limit, as expected.

RESULTS AND DISCUSSION

A computer program was written to solve Eq. (14) for r_2 , given the reaction parameters and r_1 ; this gives the smallest outer radius of an infinitely long annular reactor for which explosion should occur. The Bessel functions J_0 and Y_0 were calculated by means of subroutines from IBM's system 360 Scientific Subroutines Package (IBM 1970). Progressively increasing values of r_2 were tested until the determinant changed sign; the increment in r was then decreased and the process repeated. This procedure was continued until the value of r_2 satisfying Eq. (14) had been determined with sufficient accuracy. Calculation of the value of r_2 required about 4 sec. of DEC 1099 computer time.

Table 1 shows the effect of varying the inner radius of the reactor. We see that to a good approximation the thickness of the annular shell is independent of the radius of the inner tube. In Table 2 we see the results of increasing the temperature of the reactor thermostat. As the temperature of the thermostat increases, the increased rate of reaction necessitates progressively smaller values of r_2 if thermal runaway is to be avoided.

The effect of increasing the activation energy is shown in Table 3. Increasing the activation energy decreases the rate of the reaction, leading to increasing values of r_2 , as observed. The effect of varying the ΔE of the reaction is shown in Table 4; as the reaction becomes more exothermic we see the expected decrease in r_2 .

Changing the A factor of the rate constant produces a corresponding change in the rate of heat production, with results as shown in Table 5. Variation in c_1 or c_2 produces exactly the same type of change, as one would expect from Eq. (1). The effect of increasing the thermal conductivity of the reaction mixture is to increase the value of r_2 at which thermal runaway occurs, as one would expect, and as is shown in Table 6.

In conclusion, we note that, within the limitations of our assumptions, Green's functions provide us with a convenient method for determining the conditions for thermal runaway in coaxial reactors, and that the reactor of finite length can be handled with essentially no more difficulty than the reactor of infinite length. We note that limitations on the validity of the results are imposed by the physico-chemical assumptions that (1) the composition of the reaction mixture does not change appreciably during the course of events, and (2) heat transport is by conduction only.

The mathematical approximation made in expanding $\exp(-E_a/RT)$ in ascending powers of $\tau = (T-T_0)$ and keeping terms only through those linear in τ also imposes a constraint. We shall examine the coefficient of τ^2 in the expansion of this exponential. We have

$$\exp(-E_a/RT) = \exp\left[-\frac{E_a}{RT_0}\left(1 - \frac{\tau}{T_0} + \frac{\tau^2}{T_0^2} - \dots\right)\right] \quad (47)$$

$$\approx \exp(-E_a/RT_0)$$

$$\cdot \left[1 + \frac{E_a \tau}{RT_0^2} + \tau^2 \left(\frac{E_a^2}{2R^2 T_0^3} - \frac{E_a}{RT_0^2}\right)\right]$$

The first term in the coefficient of τ^2 is surely much larger than the second, since $E_a \gg RT_0$ for realistic cases, so that our linear approximation is valid if τ/T_0 is substantially less than $\sqrt{2RT_0/E_a}$. Improvement of this approximation requires attack on Eq. (1) without linearization, which apparently can be made only by numerical integration.

Table 1. Effect of Inner Radius on r_2

r_1 (cm)	0.5	1.0	6.0	10.0	20.0
r_2 (cm)	2.4	2.9	7.9	11.9	21.9

In these runs $\Delta E = -174$ kcal/mole, $E_a = 24$ kcal/mole, $A = 7.9 \times 10^{13}$ cm³ mole⁻¹sec⁻¹, $c_1 = c_2 = 1.0 \times 10^{-3}$ mole/cm³, $T = 550^\circ\text{K}$, $K = 55.52$ cal deg⁻¹ cm⁻¹sec⁻¹.

Table 2. Effect of Temperature on r_2

T ($^\circ\text{K}$)	540	600	700
r_2 (cm)	4.3	2.9	2.3

Here $r_1 = 2.0$ cm and the other parameters are as in Table 1.

Table 3. Effect of Activation Energy on r_2

E_a (kcal/mole)	22	24	26
r_2 (cm)	3.0	4.3	7.4

Here $T = 540^\circ\text{K}$, $r_1 = 2.0$ cm, and the other parameters are as in Table 1.

Table 4. Effect of ΔE on r_2

ΔE (kcal/mole)	-74	-174	-274
r_2 (cm)	3.5	3.0	2.8

Here $E_a = 22$ kcal/mole, $T = 540^\circ\text{K}$, $r_1 = 2.0$ cm, and the other parameters are as in Table 1.

Table 5. Effect of A on r_2

A (cm ³ mole ⁻¹ sec ⁻¹)	7.9×10^{12}	7.9×10^{13}	7.9×10^{14}
r_2 (cm)	6.4	3.5	2.5

Here $\Delta E = -74$ kcal/mole and the other parameters are as in Table 4.

Table 6. Effect of Thermal Conductivity on r_2

K (cal deg ⁻¹ cm ⁻¹ sec ⁻¹)	50.0	55.52	60.0
r_2 (cm)	6.2	6.4	6.6

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