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Prepared for the U.S. Department of Energy
Office of Environmental Restoration and
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Westinghouse Hanford Company Richland, Washington

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ABSTRACT

Analytical solutions available in Carslaw and Jaeger's Conduction of Heat in Solids *for continuous point sources and for continuous finite sources are used to demonstrate that placement of thermocouples on a fine enough grid to detect a hot spot is impracticable for existing waste tanks but fortunately not necessary. Graphs covering ranges of diffusivities, times, temperatures and heat generation rates are included.*

THERMOCOUPLE PLACEMENT AND HOT SPOTS IN RADIOACTIVE WASTE TANKS

THERMOCOUPLE SPACING

The initial intention was to determine thermocouple spacing necessary to detect a hot spot in waste stored in tanks, should one develop, in time to counter any possible unsafe situation.

The avenue of investigation that proved fruitful was examining continuous point sources in an infinite medium, a classical tack that yields closed-form solutions. The evaluation of such solutions does not require computers but can benefit greatly from them.

CONTINUOUS POINT SOURCE

These results are based on calculations using an equation on page 261 of Reference 2. That equation for temperature, v, can be written

$$v = (Q/[4*\pi*\varrho*c*D*r])*\mathrm{erfc}[r/\mathrm{sqrt}(4Dt)] \tag{1}$$

where

Q = heat rate of point source, Btu/hY
π = 3.1415927
ϱ = density of the medium, lbs/ft^3
c = specific heat, Btu/lb,°F
D = diffusivity, ft^2/h
r = radius, ft
t = time, hours from the start of the heat source.

Figure 1 shows v/Q versus diffusivity at various times at a distance of 0.1 ft from the heat source. It is seen that v/Q decreases as diffusivity increases and that it approaches a steady-state limit asymptotically as time increases. The diffusivities cover the range for soils given by Carslaw and Jaeger, which is also applicable for the radioactive wastes in the ferrocyanide tanks (Reference 2).

To examine the effect of distance, it is convenient to form the ratio of the temperature at radius r to that at a radius of 0.1 ft, which is given by the following equation:

$$v(r=r)/v(r=0.1) = (0.1/r)*\{\mathrm{erfc}[r/\mathrm{sqrt}(4Dt)] \\ /\mathrm{erfc}[0.1/\mathrm{sqrt}(4Dt)]\}. \tag{2}$$

Figures 2 through 7 show v(r=r)/v(r=0.1) versus time (the logarithms of those variables are plotted to fit the information into a convenient space) at various diffusivities for radii from 1 to 6 ft from the heat source. It is seen that the temperature ratio increases with time but starts at a small fraction. It is also seen that at small times, the effect of diffusivity is palpable, but at long times diffusivity exerts only a small influence.

These figures may be used to assess the spacing necessary for thermocouples to detect hot spots. For example, if the average volumetric heat production rate in the waste were q and a particular location happened to accumulate a concentration of radioactivity that produced a volumetric heat rate of nq, where $n > 1$, then a small volume, dV, around a point in that region (which we can say encompasses all of the accumulation) can be said to be represented by a "hot spot" heat rate $Q = (n-1)qdV$ Btu/h at that point. Entering into Figure 1 the time and diffusivity of interest enables one to read the ratio v/Q from the ordinate. Then, by multiplying that ratio by Q one can arrive at the temperature rise, v, over and above the background temperature (at the volumetric heat rate q) in that region, at a distance of 0.1 ft from the source. The effect of distance, at the time and diffusivity of interest, is then assessed by tracing the temperature ratio, v(r=r)/v(r=0.1), which decreases very rapidly as r increases, seen in Figures 2 through 7.

Such estimates indicate that the concentration factor, n, would have to be unreasonably large before the hot spot temperature would reach a value sufficient to initiate a chemical reaction in the wastes. The estimates also show that the spacing of thermocouples would have to be impracticably close together to detect such a temperature rise in a reasonable time.

FINITE HEAT SOURCE

The trouble with point sources is that they tend to produce temperatures that increase without limit as the distance from the point source decreases toward zero.

Finite sources do not have that limitation.

Consequently, the results have been calculated for a finite volume of radioactive waste, in the form of a right cylinder buried in the tank wastes, in which the heat generation rate can exceed that of the average rate in the surrounding wastes.

Carslaw and Jaeger (Reference 2), on page 266, Eq. 1, show that for radius $0 < r < a$ and height $-b < z < b$, the maximum temperature rise, v, in a cylinder with its center at the origin (in an infinite medium initially at zero temperature) is given by Eq 3:

$$v = \frac{A}{\rho c}\int_0^t [1-e^{-a^2/4Du}]\,\mathrm{erf}\frac{b}{\sqrt{(4Du)}}\,du \tag{3}$$

where

ρ = density
c = specific heat
A = heat generation rate per unit time per unit volume
D = diffusivity
t = time
u = the integration variable (with units of time).

The multiplier of du in the integrand starts out at unity when u = zero but diminishes to zero as u increases; therefore, the result of the integration approaches an asymptote as t increases.

Table 1 shows how the maximum temperature rise above the background varies with the size of the right cylindrical volume between volumes of 1/8 gal and 8 gal, for a concentration of activity twice that of the average (based on a volumetric heat generation rate of 2.264 Btu/h ft^3, which corresponds to 20,000 Btu/h being generated in a 2-ft layer of waste in a tank 75 ft in diameter.

Table 1.

Right cylinder		Diffusivity (ft^2/h)	Maximum temperature rise (°F)
radius (ft)	volume (gal)		
0.1385	0.125	0.005	0.2009
		0.010	0.1003
		0.015	0.06729
		0.020	0.05069
		0.025	0.01669
0.2771	1.000	0.005	0.7854
		0.010	0.3968
		0.015	0.2646
		0.020	0.2009
		0.025	0.1613
0.5542	8.000	0.005	3.1187
		0.010	1.5678
		0.015	1.0483
		0.020	0.7854
		0.025	0.6300

At a diffusivity of 0.017674 ft^2/h, which corresponds to a thermal conductivity of 0.5 Btu/h,ft^2,(°F/ft), and density times specific heat of 28.29 Btu/°F,ft^3, the maximum temperature rise, at twice the average heat generation rate, would be 0.23627 °F in a 1-gal volume. Therefore, at ten times the average heat generation rate, the maximum temperature rise would be 9(0.23627) = 2.126 °F. Even for an 8-gal overheated zone, it is clear that, using a lower diffusivity of 0.015 ft^2/h, at ten times the concentration, the maximum temperature rise would not exceed 9(1.0483) = 9.4347 °F.

It is unlikely that concentrations much higher than one times the average are extant in the ferrocyanide tanks because of the nature of the materials, the way they were laid down in the tanks (Reference 1), and extensive experience over many years of operations. Therefore, hot spots from unusual accumulations of radioactivity are not a reasonable basis for analysis of safety conditions in ferrocyanide tanks.

The integral in Eq 1 is simplified, and the result is more universally applicable if the integration variable is changed to $x = 4Du/a^2$ because the integrand then is dimensionless and the equation becomes

$$\left(\frac{4D}{a^2}\right)\left(\frac{\rho c}{A}\right)v = \int_0^{\frac{4Dt}{a^2}} [1-e^{-\frac{1}{x}}]\,\mathrm{erf}\left(\frac{1}{\sqrt{x}}\right)dx \qquad (4)$$

and the integral has a constant value that is understood to be 2.63. The appendix gives a FORTRAN program for evaluating that integral.

Table 2 shows how the value of the integral changes as the tolerance is tightened.

Table 2.

Tolerance	Value of integral	Percent change	$4Dt/a^2$
1E-4	2.60274		5.8E3
1E-5	2.6214	0.717	4.23E4
1E-6	2.62726	0.2235	1.949E5
1E-7	2.62997	0.1032	9.033E5

This means that the temperature is given by

$$v = 2.63\left(\frac{a^2}{4D}\right)\left(\frac{A}{\rho c}\right) \qquad (5)$$

and the results in Table 1 can be reproduced from that relationship.

Notice that the temperature is directly proportional to the square of the radius of the right cylinder and to the volumetric heat generation rate but is inversely proportional to the diffusivity and to the volumetric specific heat of the infinite diffusion medium.

Suppose there is a large volume, V, of waste that is generating a total heat rate Q, so that the average volumetric heat rate is q' Btu/h,ft^3 = Q/V.

Further suppose that within V there is a smaller volume, v, of waste that has a volumetric heat generation rate n times the average rate, or nq'. Then a heat balance on the system would yield

$$nq'v + q(V-v) = Q \tag{6}$$

where q is the volumetric heat generation rate outside the small volume v. Dividing through by V, it is seen that q is given by

$$\begin{aligned} q &= \{ Q/V - nq'(v/V) \}/\{ 1 - v/V \} \\ &= q'[1-n(v/V)]/[1-v/V] \end{aligned} \tag{7}$$

Table 3 shows how the ratio q/q' varies with v/V for various values of the heat concentration factor, n.

Table 3.

Ratio: volumetric heat rate outside concentrated region divided by average volumetric heat rate

	q/q'						
v/V \ n =	2	3	4	5	10	20	40
0.01	0.9899	0.9798	0.9697	0.9596	0.9091	0.8081	0.60606
0.02	0.9796	0.9592	0.9388	0.9184	0.8163	0.6122	0.2041
0.04	0.9583	0.9167	0.875	0.8333	0.6250	0.2083	-------
0.08	0.9130	0.8261	0.7391	0.6522	0.2174	------	-------
0.16	0.8095	0.6190	0.4286	0.2381	------	------	-------
0.32	0.5294	0.0588	------	------	------	------	-------

Obviously, when n = 1, q/q' = 1 for all values of v/V. The first row in Table 3 shows that, when v/V is 0.01, the background temperature is not strongly affected even when n = 10. Comparing the first row with the column at n = 2, it is seen that there is not much change in q/q' out to n = 10 or down to v/V = 0.08 (which corresponds to V/v = 12.5).

Consider a waste layer 5 ft thick generating a total of 10,000 Btu/h in a 75-ft diameter tank that would correspond to an average volumetric heat generation rate of 0.4527 Btu/h,ft^3 = q'. If the temperature could be allowed to rise to 350 °F without concern (because that would be 430 - 350 = 80 °F below the spontaneous reaction temperature under the most favorable conditions) then the temperature in a small volume of waste, with a heat generation rate of nq', could be allowed to rise from about 135 °F to 350 °F, or 215 °F at the maximum point in the small right-cylindrical volume assumed within the total volume of heat-generating waste.

Substituting values into Eq 5 results in the following (with $D = 0.015$ ft^2/h, $\varrho * c = 28.29$ $Btu/ft^3, °F$):

$$nq' = 28.29(215)4(0.015)/(2.63*a^2) = 138.76/a^2 \qquad (8)$$

and, using $a^2 = 231.21(v/V)^{0.666667}$,

$$n = 1.3257/(v/V)^{0.666667}. \qquad (9)$$

Table 4 shows how n varies with v/V.

Table 4. Allowable Concentration Factor, n, Versus Fractional Volume at n Times the Average Volumetric Heat Rate and the Corresponding Volume, v, and Radius, a, of the Overheated Volume.

Fractional volume	n	Volume, v, ft^3	Right cylindrical radius (ft)
0.01	28.56	220.9	3.276
0.02	17.99	441.8	4.127
0.04	11.33	883.6	5.200
0.08	7.14	1767.2	6.552
0.16	4.50	3534.4	8.255
0.32	2.83	7068.7	10.400

Although the geometry of the basis of Eq 4 does not exactly fit the situation in the waste tanks, particularly as the radius of the right cylinder, a, gets larger, the results in Table 4 are useful as indicators of important conclusions.

Carrying the investigation a step further shows how time becomes a consideration. Time is buried in the parameter $4Dt/a^2$, which is listed in Table 2, and which is called x. Taking $D = 0.015$ ft^2/h, it is seen that t, in hours, is given by $16.667a^2x$, with a in feet. Therefore, we construct the table below as an extension of Table 2.

Table 5.

Value of integral (Eq 3)	Value of x $(=4Dt/a^2)$	t/a^2 $(D=0.015ft^2/h)$	t, hours (a=3.276ft)
2.6027	5.8E3	0.0967E6	1.037E6
2.6214	4.23E4	0.705E6	7.566E6
2.62726	1.949E5	3.248E6	34.858E6
2.62997	9.033E5	15.055E6	161.573E6

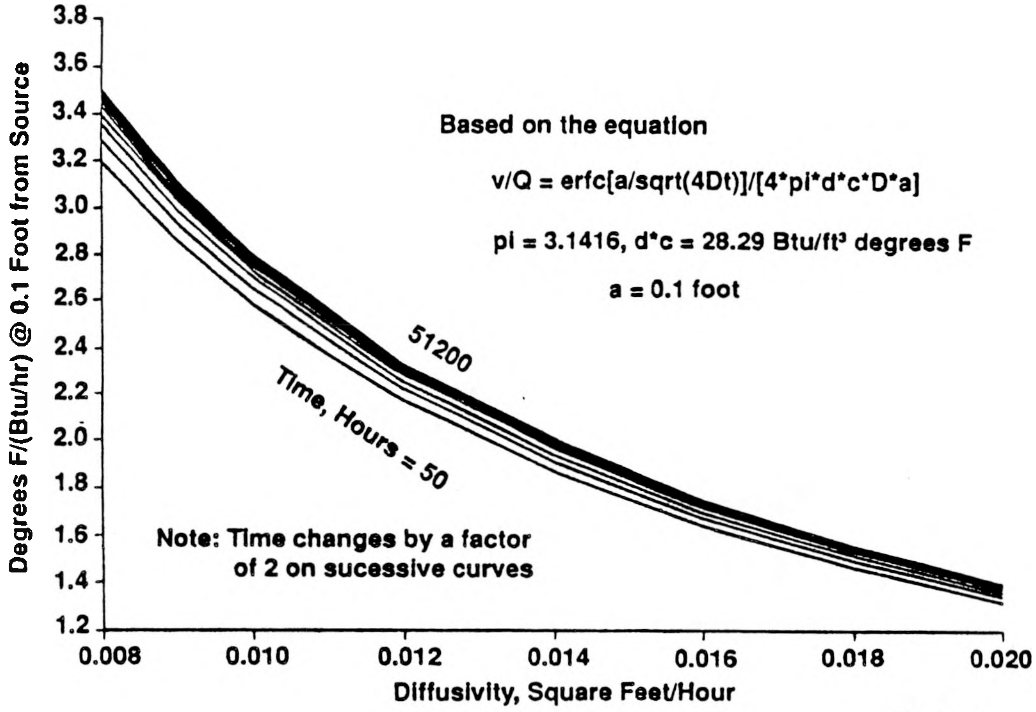
Because 30 years, the half-life of ^{137}Cs, corresponds to 0.263E6 h, it is clear that the decay of the radioactivity has to be taken into account in the integrand of Eq 5 by introducing the factor $\exp(-\lambda a^2 x/4D)$, where λ = the decay constant for $^{137}Cs = 2.6357E-6\ h^{-1}$. With a = 3.276 ft and D = 0.015 ft^2/h, the constant 2.63, with an error of 1E-7, becomes 2.547, with an error of 1E-7. Such a small correction has an unimportant effect on any conclusions here. Figure 8 shows how the value of the integral is affected by the decay of the heat source with time, using dimensionless variables.

REFERENCES

1. Chemical Engineering Progress, Vol 53 No 2, February 1957, p 3F and 95F, *Volume Reduction of Radioactive Wastes By Carrier Precipitation*, by R. E. Burns and M. J. Stedwell (Hanford Laboratories Operation and Chemical Processing Dept., respectively, during General Electric Company tenure).

2. Carslaw, H. S. and J. C. Jaeger, 1959, *Conduction of Heat in Solids*, 2nd Ed., Oxford University Press.

Heat Diffusion: Continuous Point Source Temperature Rise per Heat Rate versus Diffusivity at Various Times



Based on the equation

v/Q = erfc[a/sqrt(4Dt)]/[4*pi*d*c*D*a]

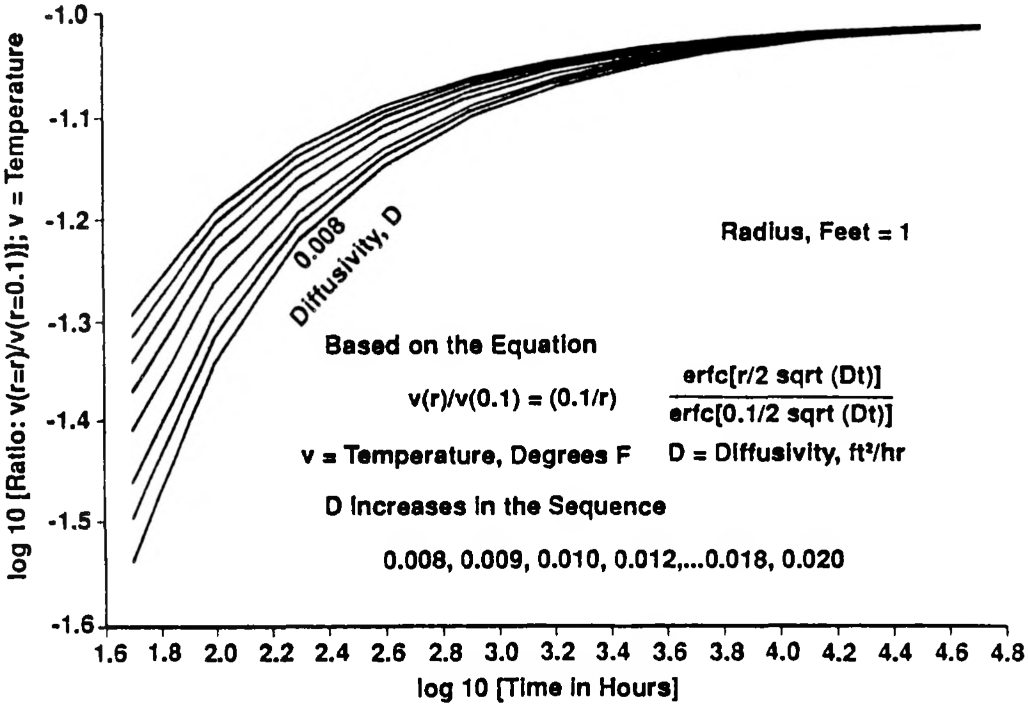
pi = 3.1416, d*c = 28.29 Btu/ft³ degrees F

a = 0.1 foot

Note: Time changes by a factor of 2 on sucessive curves

Figure 1

Temperature Ratios for a Continuous Point Source in an Infinite Conductor versus Time at Various Diffusivities for a Given Radius



Radius, Feet = 1

Based on the Equation

$$v(r)/v(0.1) = (0.1/r)\ \frac{\text{erfc}[r/2\ \text{sqrt}\ (Dt)]}{\text{erfc}[0.1/2\ \text{sqrt}\ (Dt)]}$$

v = Temperature, Degrees F   D = Diffusivity, ft²/hr

D Increases in the Sequence

0.008, 0.009, 0.010, 0.012,...0.018, 0.020

Figure 2

Temperature Ratios for a Continuous Point Source in an Infinite Conductor versus Time at Various Diffusivities for a Given Radius

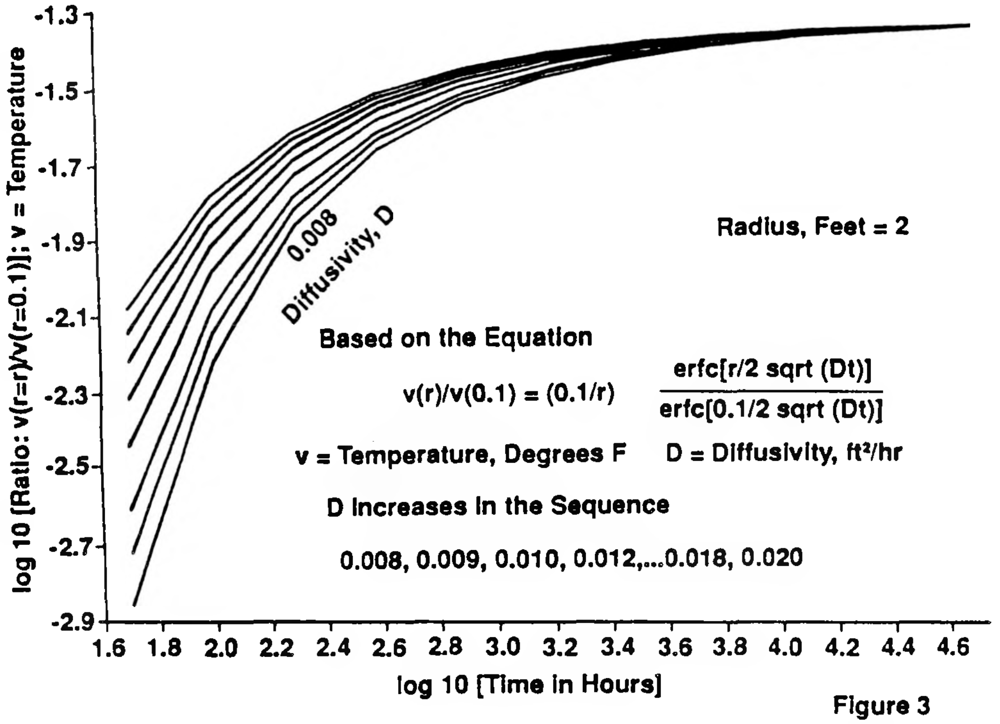


Figure 3

Temperature Ratios for a Continuous Point Source in an Infinite Conductor versus Time at Various Diffusivities for a Given Radius

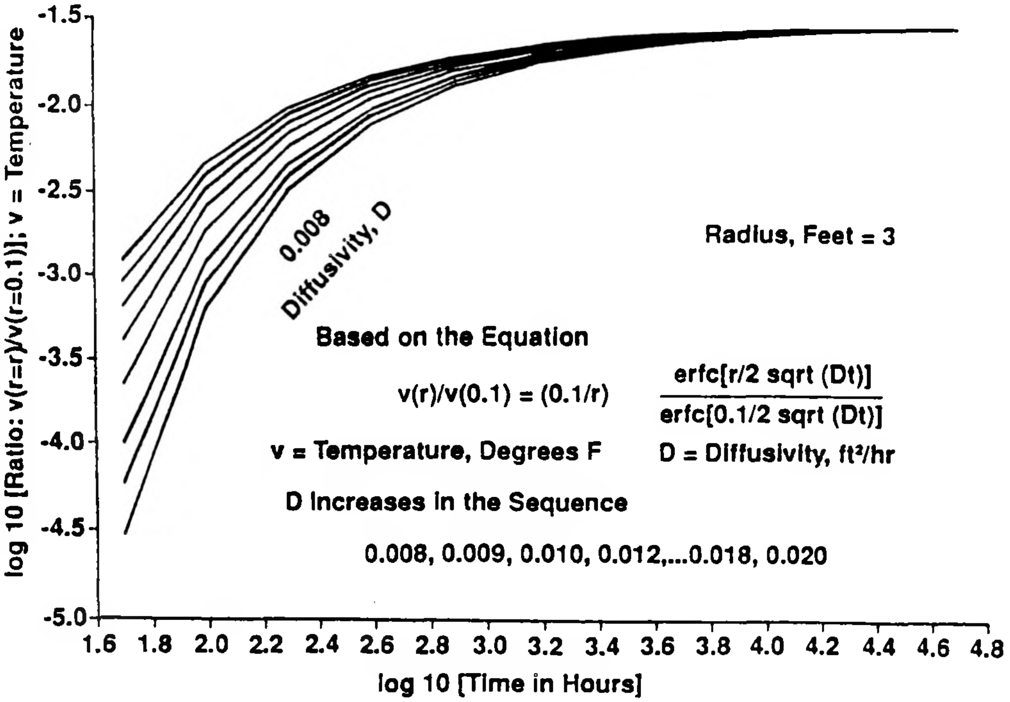


Figure 4

Temperature Ratios for a Continuous Point Source in an Infinite Conductor versus Time at Various Diffusivities for a Given Radius

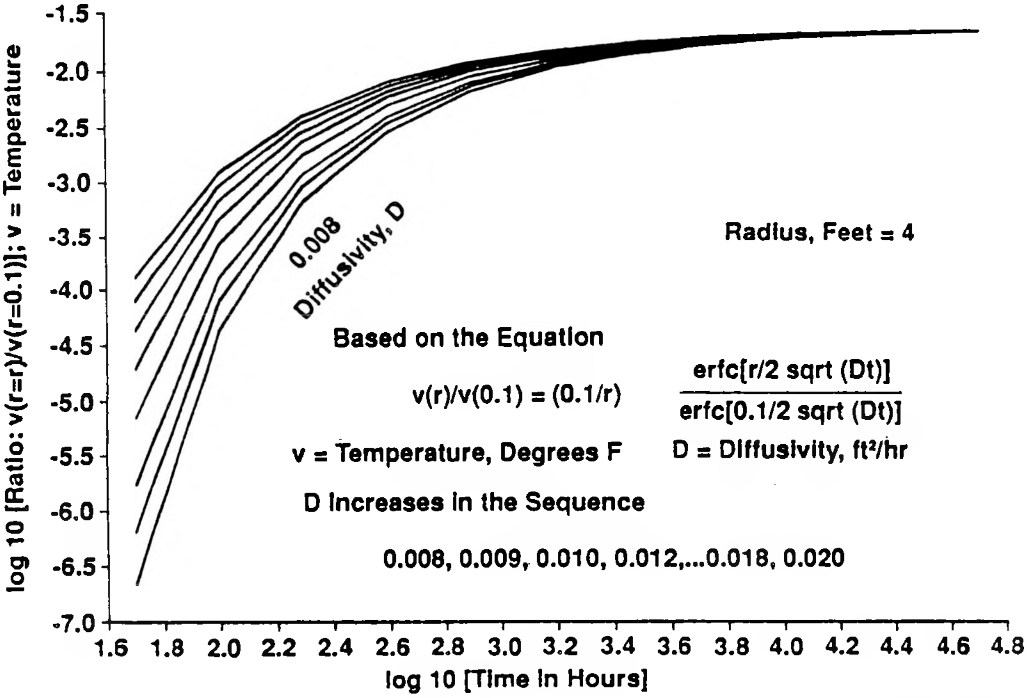


Figure 5

Temperature Ratios for a Continuous Point Source in an Infinite Conductor versus Time at Various Diffusivities for a Given Radius

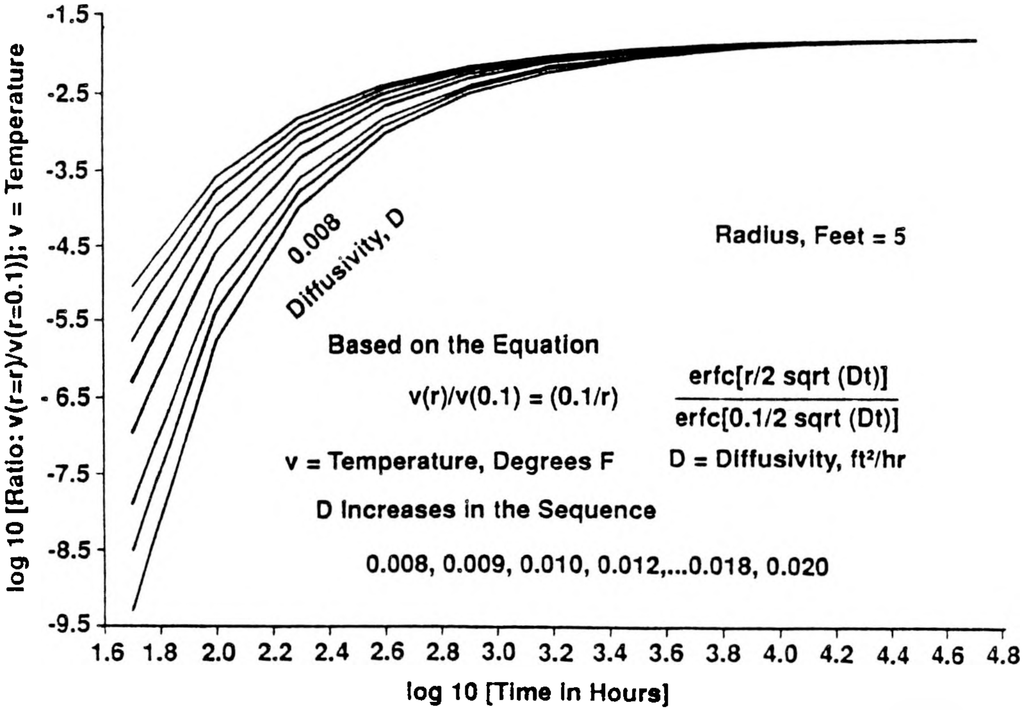


Figure 6

Temperature Ratios for a Continuous Point Source in an Infinite Conductor versus Time at Various Diffusivities for a Given Radius

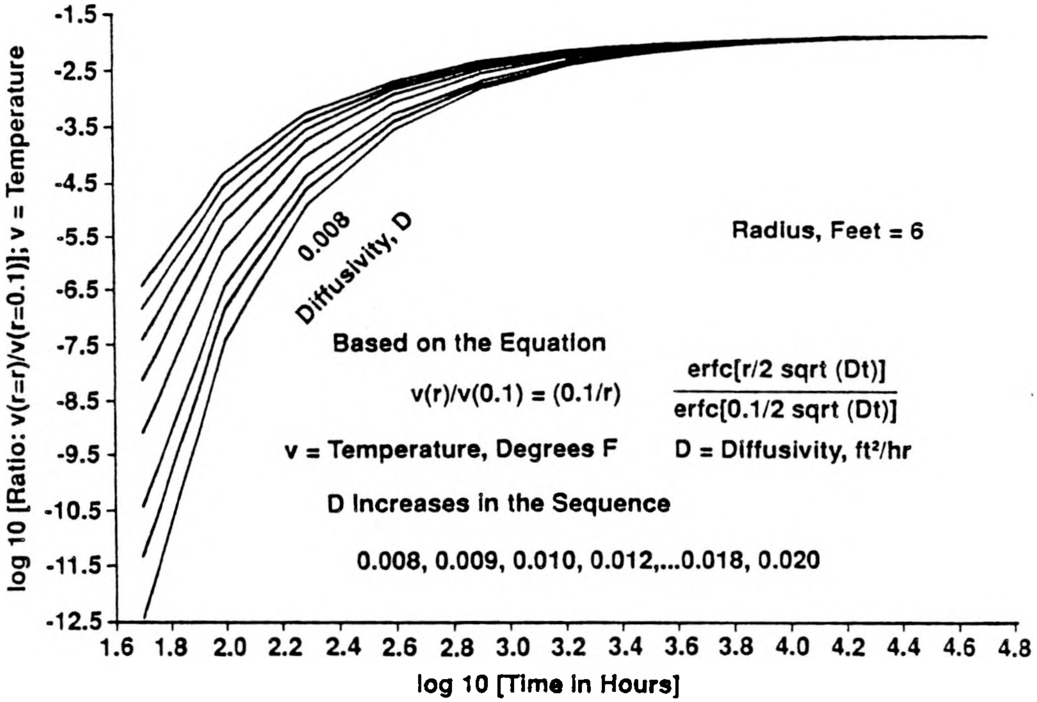


Figure 7

Radioactive Heat Source Integral versus log 10(Z), Z = $(a^2/4D^*\tau)$

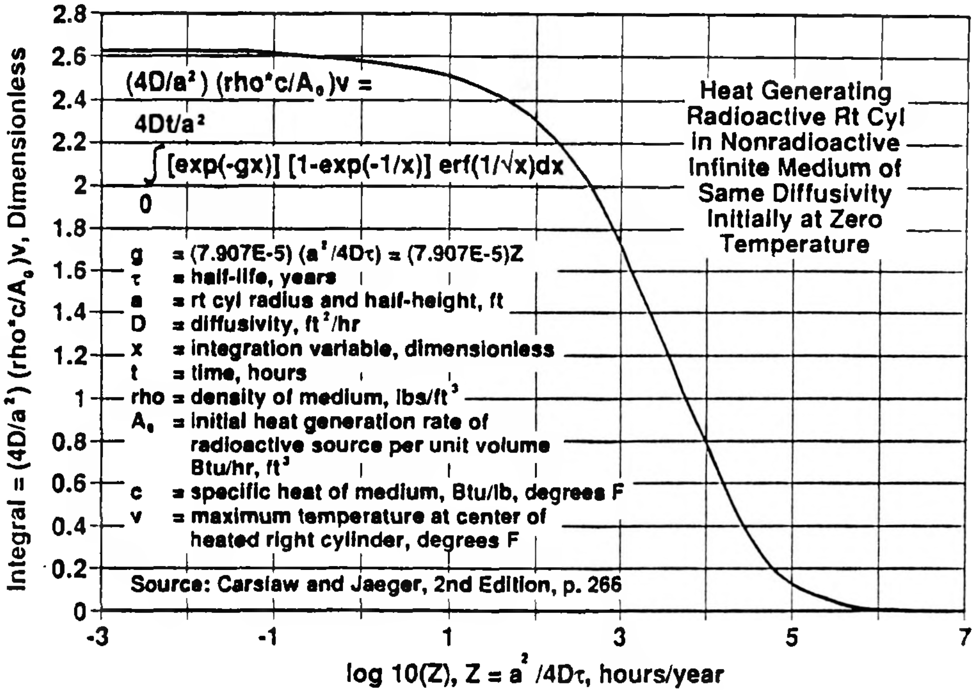


Figure 8

APPENDIX A

```
c     This is r3cylhts.for, a FORTRAN program to calculate the
c     function (ϱ*c/A)*v = (a*a/(4*D))integral from 0 to (4D/a*a)t
c     of
c              [1.-exp(-1./x)]erf(1./sqrt(x))dx
c
c     where a is the radius of a right cylinder of height 2a with
c     center at the origin, ϱ is density, c specific heat, A
c     the volumetric heat generation rate and v is the temperature
c     at the origin as a function of time t.  The function goes to
c     a steady value as t gets large.
c
      OPEN (7,FILE = 'r3cylhts.dat')
      write(6,101)
      write(7,101)
  101 format(1x,'        x                S        ',//)
      S = 1./10.**20.
      x = 1./10.**20.
      dx = 0.0001
    1 continue
      if(x-dx)2,3,3
    2 x1 = x
      y1 = 1.
      yp1 = 1.
      x2 = x1 + dx
      y2 = 1. - exp(-1./x2)
      z2 = 1./sqrt(x2)
        call erfox(z2,w2)
      yp2 = y2*w2
      ypb = 0.5*(yp1 + yp2)
      dS = ypb*dx
      S = S + dS
      x = x2
c     write(6,100) x, S
c     write(7,100) x, S
  100 FORMAT(1x,2e14.6)
      go to 1
    3 x1 = x
      y1 = 1. - exp(-1./x1)
      z1 = 1./sqrt(x1)
      if(z1-3.)4,4,5
    4   call erfox(z1,w1)
      go to 6
    5   call erfcox(z1,v1)
      w1 = 1. - v1
    6 yp1 = y1*w1
      x2 = x1 + dx
      y2 = 1. - exp(-1./x2)
      z2 = 1./sqrt(x2)
      if(z2 - 3.)7,7,8
```

```
    7   call erfox(z2,w2)
      go to 9
    8   call erfcox(z2,v2)
      w2 = 1.-v2
    9 yp2 = y2*w2
      ypb = 0.5*(yp1 + yp2)
      dS = ypb*dx
      S = S + dS
      dT = dS/S
      x = x2
c     write(6,100) x, S
c     write(7,100) x, S
      if(x .ge. .00009 .and. x .le. 0.001) then
        dx = 0.0001
      ELSEif(x .ge. .0009 .and. x .le. 0.01) then
        dx = 0.001
      else if(x .ge. .009 .and. x .le. 0.1) then
        dx = 0.01
      else if(x .ge. .09  .and. x .le. 1.) then
        dx = 0.1
      else if (x .ge. 1. .and. x .le. 100.) then
        dx = 1.
      else if (x .ge. 100. .and. x .le. 1000.) then
        dx = 10.
      else if (x .ge. 1000. .and. x .le. 10000.) then
        dx = 100.
      else
      dx = 200.
      end if
      if(dT - 1.E-7)  10,10,1
   10 write(6,99)
      write(7,99)
   99 format(/)
      write(6,100) x, S
      write(7,100) x, S
      write(6,99)
      write(7,99)
      end
```

```
C
C     THIS IS ERFOX.FOR  , A SUBROUTINE TO CALCULATE ERF OF X
C
      SUBROUTINE ERFOX(X,W)
      DIMENSION       S(34), T(34), R(34), A(34), C(34)

      PI=3.1415927
      B = SQRT(PI)
      Y=X
      IF( Y .LE. 0.5) J = 6
      IF( Y .LE. 1. .AND. ( Y .GT. 0.5)) J = 11
      IF( Y .LE. 2. .AND.  ( Y .GT. 1.)) J = 21
      IF( Y .LE. 3. .AND. ( Y  .GT. 2.)) J = 31
      A(1)=0.
      T(1) = (-1.)*( Y**2 )/3.
      S(1) = T(1)
    1 DO 10 I=1,J
        A(I+1)=A(I)+1.
        C(I)=A(I+1)
        R(I) = (-1.)*(Y**2)*(2.*C(I)+1.)/((C(I)+1.)*(2.*C(I)+3.))
        T(I+1) = T(I)*R(I)
   10   S(I+1) = S(I) + T(I+1)
      Z=S(J-1)
         W  = (2./B)*Y*(1.+Z)
      RETURN

    END
C     THIS IS ERFCOX.FOR  , A SUBROUTINE TO CALCULATE ERFC OF X
C
      SUBROUTINE ERFCOX(X,W)
      DIMENSION       S(10), T(10), R(10), A(10), C(10)
            PI=3.1415927
      B = SQRT(PI)
      Y=2.*X*X
      A(1)=0.
      T(1) = -1./Y
      S(1) = T(1)
    1 DO 10 I=1,9
        A(I+1)=A(I)+1.
        C(I)=A(I+1)
        R(I) = (-1.)*(2.*C(I)+1.)/Y
        T(I+1) = T(I)*R(I)
   10   S(I+1) = S(I) + T(I+1)
      Z=S(10)
      W=(1.+Z)*(EXP(-X*X))/(B*X)
      RETURN
      END
```

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