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Geothermal Aspects of Radioactive Waste Disposal into the Subsurface

IRSHAD R. MUFTI¹

Amoco Production Company Research Center Tulsa, Oklahoma 7/4102

A method is presented for calculating the temperature increase that results from heat generation by radioactive wastes placed in a subsurface cavity. Cavities of spherical, cylin-drical, and other shapes are considered. The method takes into account the disintegration energy released by the various isotopes present in a waste and is applicable to wastes of all types, irrespective of their origin, age, or radioactive composition. This has been achieved by grouping the isotopes according to their decay characteristics. The procedure described is particularly useful when dealing with high-level wastes from power reactors. After a detailed examination of all the isotopes identified in significant proportions in a 90-day-old waste, it is demonstrated that when such a waste becomes 1 year old there are only 5 isotope groups that are mainly responsible for the heat generation. The number of groups reduces to 4 when the waste becomes 2 years old. In the theoretical treatment of the problem dimensionless parameters are used. These parameters make it possible to present the results in a few diagrams, which can be easily used for a rapid determination of the temperature rise for a subsurface cavity containing radioactive waste. Since these diagrams do not depend on specific values of cavity dimensions, thermal constants of subsurface materials, or decay characteristics of the radioactive isotopes, they are applicable to a wide variety of situations that may be encountered in practice. By comparing the temperature rise from a subsurface heat source of constant strength with another that decays exponentially with time, the necessary conditions are determined for considering that the radioactive waste is a constant source of heat without introducing serious errors. As an application of the theory, the temperature field from a spherical cavity in rock salt containing a 2-year-old reactor waste has been investigated in detail. The space and time variation of the temperature, the maximum temperatures at various points, the rate of variation, the extent of penetration of the field, and the influence of this penetration on major geologic structures such as a salt dome are discussed.

Radioactive wastes pose a serious threat of pollution to man's environment. These wastes are being produced in increasing quantities by nuclear power plants. Other applications of nuclear energy, such as nuclear-explosion seismology, further contribute toward the production of unwanted radioactive materials.

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To avoid a possible contamination of ground waters, which may result from subsurface waste disposal, most of the radioactive wastes are being stored in metallic tanks at or near the surface.

Many types of rock have been considered for burying the radioactive wastes. Rock salt, because of its imperviousness to water, seems to be ideal for this. However, because radioactive wastes release a considerable amount of heat and because rock salt becomes very plastic at high temperatures, any attempt to dispose of wastes into a salt cavity must be preceded by a thorough investigation of the resulting temperature field in and around the cavity and its propagation into surrounding media in terms of space and time.

The present paper describes a theoretical study of this field. The work was carried out in connection with a research project of the Geological Survey of West Germany for investigating the feasibility of waste disposal in abandoned salt mines; therefore a major portion of the discussion and the practical examples that follow are in relation to rock salt. The theoretical treatment is, however, quite

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¹ Formerly at the Geological Survey of West Germany, Hanover, Germany.

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RADIOACTIVE WASTE DISPOSAL

The rise in temperature resulting from deposit of radioactive waste in a salt cavity is a complicated function; it depends on factors such as geometry of the cavity, thermal characteristics of the waste and of the surrounding medium, and radioactive composition of the waste.

Several authors have studied this problem: Goldenberg [1951], Birch [1958], Schlechter and Gloyna [1959], Kotewale and Ganguly [1960], Brandes [1964], and Black and Dickey [1966]. Most of them have tackled the problem by assuming that the waste is a constant source of heat and have restricted their investigation to the temperatures at the center and the surface of the cavity. Other investigators have taken into account the variation in the rate of heat generation in the waste, presenting their results in the form of complicated relations that can give temperature values only at a few points; such results require tedious numerical calculations each time a change occurs in the size of the cavity, the value of the thermal constants, or the radioactive composition.

In the present paper, a method is presented that permits a ready calculation of the temperature field resulting from a waste having an arbitrary radioactive composition. The method provides for the values of the disintegration energy of the various isotopes present in the waste and of their decay constants. By using dimensionless parameters, the results obtained are applicable to the wide variety of situations that may be encountered.

A complete list of symbols used is given as an appendix.

RADIOACTIVE WASTE AS A SOURCE OF HEAT

The energy released by disintegration from a high-level radioactive waste containing fission products is so great that the waste keeps boiling through self-heating. After cooling for some time, all the fission products but those that are very short-lived may still be present in significant amounts. Fission products are produced in varying proportions, and their energy of disintegration varies considerably.

A fission product in the waste will make a significant contribution to heat generation only if it meets the following conditions:

2. Its half-life is not very short. Most of the short-lived isotopes would have already disintegrated at the time of subsurface disposal of the waste. The only exception to this is the short-lived daughter products resulting from the disintegration of long-lived parents. The energy released by them must be taken into account.

1. Its fission yield is not very low.

3. The energy of disintegration is not very small.

4. The half-life is not very long. The rate of disintegration and hence of heat release for a long half-life product remains negligiblysmall even when its yield is significant.

The fission yields, the half-lives, and the disintegration energies of the various fission products have been reported many times (see for example Watson [1961]). As the values vary over very wide ranges, the probability that a fission product will meet the four conditions mentioned above is very small. As an example, the half-life of ¹⁴⁴Ce is 0.78 year, whereas that of ¹⁵⁵Eu is 1.6 years; both have comparable amounts of disintegration energy, but the yield of ¹⁴⁴Ce is about 200 times higher. Therefore, in a mixture of fission products in which both are present, the thermal contribution from ¹⁵⁵Eu can be neglected.

By a similar argument, it can be ascertained [Mufti, 1966] that all the products whose fission yield is less than 0.1% collectively release a negligibly small amount of energy as compared to the energy released by products whose yield is more than 0.1%. Consequently, the isotopes that need to be considered must have a yield of at least 0.1%. Of these, the isotopes that are very short lived will not be present in significant proportions if the waste has been allowed to cool for a period of 1 year or longer, and therefore they also need not be considered. The very few isotopes that still remain are further reduced in number on account of condition 3 mentioned above.

Let us consider an actual example of a 90day-old waste (Table 1) reported by *Bruce* [1960]. We call this waste 'X'. When this waste becomes, let us say, 1 and 2 years old, the activities of the various isotopes present in it can be easily calculated by using the wellknown decay equations (see, for example,

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TABLE 1. Important Fission Products Identified in Significant Proportions in a 90-Day-Old Reactor Waste

be expressed as

$$e_{zr} = 1.221 \times 10^{-3} \text{ cal/sec}$$

$$e_{nb} = 1.130 \times 10^{-3} \text{ cal/sec}$$

Since e_{sr} and e_{nb} are almost equal, we can take an average value of 1.176×10^{-3} cal/sec and write down

$$E_{zr+nb} = (3.25 \times 1.176) \times 10^{-3} C_{zr}$$

(cal cm⁻³ sec⁻¹)

We further note that the half-life of ⁶¹Y is almost equal to that of ⁶⁵Zr, and that the energy/ curie released by ⁶¹Y is 7.251 \times 10⁻⁴ cal/sec. Therefore, for purposes of energy calculations, 1 curie of ⁶¹Y is equivalent to 0.1897 curie of ⁶⁵Zr. Consequently, by assuming an average half-life of 60.2 days for both ⁶⁵Zr and ⁶¹Y, both isotopes can be handled together. Similarly ⁶⁵Sr and ¹³⁷Cs, which have almost equal halflives, can be handled together, along with their daughter products ⁶⁰Y and ¹³⁷Ba. The energy released by them per curie is

$$e_{tr+y} = 1.317 \times 10^{-3} \text{ cal/sec}$$

 $e_{ct+bc} = 1.205 \times 10^{-3} \text{ cal/sec}$

Therefore, by assuming an average half-life of 29 years, we can consider that 1 curie of ^{137}Cs is equivalent to 0.915 curie of ^{69}Sr . By classifying the fission products in this manner, we finally end up with a very short list of the isotopes that need to be considered in the waste X when it becomes 1 year old. The results are presented in Table 3.

Table 3 shows that the energy released by the first 5 isotope groups amounts to 7.951×10^{-4} cal/(cm³ sec), whereas the total energy released from all the remaining isotopes amounts to 7.077×10^{-7} cal/(cm³ sec), i.e., only 0.09% of the former amount. This leads to the interesting conclusion that when a mixture of fission products, such as waste X, is 1 year old, an accurate estimate of the heat generation can be made by considering only 5 isotope groups.

It is easy to verify that when the waste becomes 2 years old, the contribution from ⁵⁰Zr, ⁵⁰Nb, and ⁵¹Y becomes negligible, and the number of isotope groups responsible for the generation of heat is reduced to 4. This simpli-

Half-life, Concentration, Isotope years $\mu c/cm^3$ 89Sr 0.148 $1.6 imes10^4$ 1.0×10^{4} 2.3×10^{4} 2.3×10^{5} 2.7×10^{5} 7.2×10^{4} 3.5×10^{4} 3.1×10^{3} 90Sr 28.091Y 0.159 ⁹⁵Zr 0.178 106Ru 1.0 103Ru 0.11 ¹²⁹Te 0.09 137Cs 2.0×10^{4} 5.2×10^{3} 9.8×10^{4} 5.5×10^{3} **30.0**. 140Ba 0.035 141Ce 0.088 143Pr 0.038 144Ce 0.78 $6.9 imes10^{5}$ 147Nd 1.6×10^{3} 9.0×10^{4} 0.032 147Pm 2.6151Sm 93.0 5.6×10^2

Evans [1955]). The results are presented in Table 2, which also includes the rate of heat generation by various isotopes in calories per cubic centimeter. The energy data for those products that have short-lived daughter products includes the energy released by the latter. In the case of ⁹⁰Sr, ¹³⁷Cs, and ¹⁴⁴Ce, which have very short-lived daughter products and are therefore in equilibrium with the corresponding parent products, the total disintegration energy could be obtained by simple addition. Such a procedure is not justified for ⁰⁵Zr (half-life: 63 days), whose daughter product has a comparable half-life of 35 days. The procedure for evaluating the release of energy for this case is explained below.

When the waste has cooled for 1 year or more, one can assume that

$$C_{nb}/C_{zr} = T_{zr}/(T_{zr} - T_{nb}) = 2.25$$
 (1)

where C_{zr} and C_{nb} denote the activities in curies of ⁶⁵Zr and ⁶⁵Nb per cubic centimeter of the waste at a given time, and T_{zr} and T_{nb} denote their half-lives. Then the energy contributed per cubic centimeter by ⁶⁵Zr and its daughter product can be expressed as

$$E_{ar+nb} = C_{ar}e_{ar} + 2.25C_{ar}e_{nb}$$

 $E_{sr+nb} = C_{sr}(e_{sr} + 2.25e_{nb})$

where e_{sr} and e_{nb} denote the energy/curie released by ⁶⁸Zr and ⁶⁸Nb, respectively, and can

.

8 71 17 Isotope ¹⁰Sr ¹⁰Sr ¹⁰Y ¹⁰⁴Ru ¹⁰⁴Ru ¹⁰⁷Cs ¹⁰⁴Ce ¹⁰⁴Pm ¹⁰⁴Sm

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*%/r +-

**Sr +

¹⁰⁶Ru + ¹¹¹Ce +

¹⁴⁷Pm

103 Ru -

12 9m Te

140Ba

141C0

10Nb

143Pr

*ºSr

Group

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$$= 1.221 \times 10^{-3}$$
 cal/sec

 $= 1.130 \times 10^{-3}$ cal/sec

 $a e_{n\delta}$ are almost equal, we can take value of 1.176×10^{-3} cal/sec and

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$$(cal cm^{-3} sec^{-1})$$

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 TABLE 2.
 Generation of Heat from the Waste X

| otope | Daughter Product | Isotope Concentration | | H C | Heat Generation from Waste X | |
|------------------|---------------------|------------------------------------|-------------------------------------|-----------------------------|--|---|
| | | After 1 Year, c/cm ³ | After 2 Years, c/cm ³ | tion per Curie,* cal/sec | After 1 Year, cal/(cm ³ sec) | After 2 Years, cal/(cm ³ sec) |
| ⁹⁹ Sr | | 4.76×10^{-4} | 4.48×10^{-6} | 6.897×10^{-4} | 3.28×10^{-7} | 3.09×10^{-9} |
| ⁹⁰ Sr | 90Y | 2.26×10^{-2} | 2.20×10^{-2} | 1.317×10^{-3} | 2.97×10^{-5} | 2.90×10^{-5} |
| nΥ | ••• | 8.74×10^{-3} | 1.13×10^{-4} | 7.251×10^{-4} | 6.34×10^{-6} | 8.19×10^{-8} |
| 5Zr | 95Nb | $1.46 	imes 10^{-2}$ | 2.97×10^{-4} | 3.822×10^{-3} | 5.56×10^{-5} | $1.14 	imes 10^{-6}$ |
| 06Ru | 106Rh | 4.28×10^{-2} | 2.14×10^{-2} | 1.859×10^{-3} | 7.96×10^{-5} | 3.98×10^{-5} |
| ^{37}Cs | 137Ba | 1.97×10^{-2} | 1.92×10^{-2} | 1.205×10^{-3} | 2.37×10^{-5} | 2.32×10^{-5} |
| 44Ce | 144Pr | 3.54×10^{-1} | 1.46×10^{-1} | 1.671×10^{-3} | 5.92×10^{-4} | $2.44 	imes 10^{-4}$ |
| 47Pm | | 7.36×10^{-2} | $5.65 	imes 10^{-2}$ | 1.048×10^{-4} | 7.71×10^{-6} | 5.92×10^{-6} |
| ⁵¹ Sm | ••• | $5.56	imes10^{-4}$ | $5.52	imes10^{-4}$ | $6.231	imes10^{-6}$ | $3.47 	imes 10^{-8}$ | $3.44	imes10^{-8}$ |

* The data given here include the energy released by the corresponding daughter products.

fication permits the investigation of the geothermal field resulting from the subsurface disposal of radioactive waste, taking into account the amounts of energy released by the various isotopes present in the waste.

TEMPERATURE RISE DUE TO SUBSURFACE CAVITIES FILLED WITH RADIOACTIVE WASTE

Spherical Cavities

We shall consider the simple theoretical model of a spherical cavity filled with radioactive waste. The spherical cavity is assumed to be located in an infinite, homogeneous, and isotropic medium at a uniform initial temperature. We further assume that the thermal properties of the sphere are the same as those of the surrounding medium. To start with, we

TABLE 3. Heat Generation from Various Isotope Groups When the Waste X Becomes 1 Year Old

| Isotope | Heat Generation, cal/(cm ³ sec) |
|--|---|
| ⁹⁵ Zr + ⁹⁵ Nb + ⁹¹ Y | 6.25×10^{-5} |
| ⁹⁰ Sr + ⁹⁰ Y + ¹³⁷ Cs + ¹³⁷ Ba | 5.33×10^{-5} |
| 106Ru + 106Rh | 7.96×10^{-5} |
| ¹⁴⁴ Ce + ¹⁴⁴ Pr | 5.92×10^{-4} |
| 147Pm | 7.71×10^{-6} |
| 89Sr | 3.28×10^{-7} |
| 103Ru + 103Rh | 2.65×10^{-7} |
| 129mTe + 129Te | 2.46×10^{-8} |
| 140Ba | negligible |
| ¹⁴ Ce | 9.01 × 10 ⁻⁸ |
| 147Nb | negligible |
| 143Pr | negligible |

shall consider the case when the cavity generates heat at a constant rate.

We shall base our treatment on dimensionless parameters. Such an approach, besides being more generally applicable, will enable us to investigate more complicated cases, such as an exponentially decaying heat source and a mixture of such sources decaying at different rates.

Let us introduce the following notation:

- temperature, °C. T
- time, sec. t.

С

- distance from the cavity center, cm. r, R_{\star} cavity radius, cm.
- initial temperature of the medium, °C.
- T_{0} , heat capacity of the medium per unit c. volume, cal cm⁻³ °C⁻¹.
- Κ, thermal conductivity, cal cm⁻¹ sec⁻¹ °C⁻¹.
- k, diffusivity, $cm^2 sec^{-1}$.
- A_i , rate of heat generation, cal cm⁻³ sec⁻¹.

The boundary-value problem for the constant heat source can then be stated as

$$\frac{\partial T}{\partial t} = K \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} \right) + A_i$$

$$0 \le r \le R \quad t \ge 0$$
(2)

$$c\frac{\partial T}{\partial t} = K\left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r}\frac{\partial T}{\partial r}\right) \quad r > R \quad t > 0 \quad (3)$$

$$T(r, 0) = T_0 \tag{4}$$

$$\lim_{r \to \infty} T(r, t) = T_0 \tag{5}$$

$$T(R + 0, t) = T(R - 0, t)$$
 (6)

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$$\left. \frac{\partial T}{\partial r} \right|_{r=R+0} = \left. \frac{\partial T}{\partial r} \right|_{r=R-0}$$

$$T(0, t)$$
 is finite for $t > 0$

Birch [1958] has solved this problem by using the method of Laplace transformation. If we introduce the following new variables,

$$\tau = kt/R^2 \tag{9}$$

 $\rho = r/R$ (10)

 $U = T - T_0 \circ C$ (11)

$$A_0 = A_i R^2 / K \,^{\circ}\mathrm{C} \tag{12}$$

the equations given by Birch can be written in the form

$$\frac{U(\rho, \tau)}{A_0} = \tau \left[1 + \frac{4\sqrt{\tau}}{\rho} \left(i^3 \operatorname{erfc} \frac{1+\rho}{2\sqrt{\tau}} - i^3 \operatorname{erfc} \frac{1-\rho}{2\sqrt{\tau}} \right) + \frac{2}{\rho} \left(i^2 \operatorname{erfc} \frac{1+\rho}{2\sqrt{\tau}} - i^2 \operatorname{erfc} \frac{1-\rho}{2\sqrt{\tau}} \right) \right] \quad 0 \le \rho < 1 \quad (13)$$
and

$$\frac{U(\rho, \tau)}{A_0} = \frac{2\tau}{\rho} \left[2\sqrt{\tau} \left(i^3 \operatorname{erfc} \frac{\rho+1}{2\sqrt{\tau}} - i^3 \operatorname{erfc} \frac{\rho-1}{2\sqrt{\tau}} \right) + i^2 \operatorname{erfc} \frac{\rho+1}{2\sqrt{\tau}} + i^2 \operatorname{erfc} \frac{\rho-1}{2\sqrt{\tau}} \right] \qquad \rho > 1 \qquad (14)$$

where

*

 i^n erfc $x = \int_{1}^{\infty} i^{n-1}$ erfc $\xi d\xi$ $n=1,\,2,\,3,\,\cdots,$

e

and

$$\operatorname{rfc} x = 1 - \operatorname{erf} x \qquad (16)$$

The temperature rise for the steady state can be very easily obtained by setting $\partial T/\partial t =$ 0 in equations 2 and 3 before performing the integrations. The results expressed in terms of A_0 and ρ are

$$U(\rho)/A_0 = \frac{1}{2}(1-\rho^2/3)$$
 $0 \le \rho < 1$ (17) and

$$U(\rho)/A_0 = \frac{1}{3}\rho \qquad \rho > 1$$
 (18)

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(7)

(8)

Note that all quantities appearing in equations 13, 14, 17, and 18, including the ratio U/A_0 , are dimensionless.

Hitherto we have considered the case of a constant spherical heat source. We shall now assume that this source consists of radioactive waste containing only one type of isotopes. If λ is the decay constant, the heat generation can be expressed as

$$A(t) = A_i e^{-\lambda t} \tag{19}$$

 A_{i} denotes the heat generation from the waste at an initial time t = 0. This is the time when the waste is deposited into the ground. By using the dimensionless time scale τ , relation 19 can be rewritten as

 $A(\tau) = A_i e^{-\lambda R^2 t/k} = A_i e^{-\lambda r}$ (20)where

$$\lambda = R^2 \lambda / k \tag{21}$$

is also a dimensionless parameter. The temperature for this case can be obtained with the help of Duhamel's theorem [Churchill, 1958] from relations 13 and 14. One gets as result

$$U(\rho, \tau) = A_0 V(\rho, \tau)$$

- $A_0 \bar{\lambda} \int_0^\tau e^{-\lambda(\tau-\mu)} V(\rho, \mu) d\mu$ (22)

where V is the value of U in equation 13 or 14 for $A_0 = 1$.

The integral appearing in relation 22 can be written in the form

 $\int_{a}^{r} e^{-\mathfrak{X}(r-\mu)} V(\rho, \mu) \ d\mu$

$$= V(\rho, l\tau) \int_0^\tau e^{-\chi(\tau-\mu)} d\mu$$

$$\int_0^{\infty} e^{-\chi(\tau-\mu)} V(\rho, \mu) \ d\mu$$

$$=\frac{V(\rho, \tau 1)}{\lambda}(1-e^{-\lambda \tau}) \qquad 0 < l < 1$$

Therefore

(15)

$$\lim_{\to\infty}\int_0^\tau V(\rho,\,\mu)e^{-\chi(\tau-\mu)}\,d\mu\,=\,\frac{V(\rho,\,\infty)}{\chi}$$

where $V(\rho, \infty)$ corresponds to the temperature in the steady state. Whence

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$$= \int_{0}^{\tau} V(\rho, \tau)$$

$$= \int_{0}^{\tau} e^{-\bar{\lambda}(\tau-\mu)} V(\rho, \mu) d\mu \qquad (22)$$

The value of U in equation 13 or

al appearing in relation 22 can be form

$$(a, l\tau) \int_0^\tau e^{-\chi(\tau-\mu)} d\mu$$

$$\frac{c_{\star}, \tau 1}{\lambda} (1 - e^{-\lambda \tau}) \qquad 0 < l < 1$$

$$(\rho, \mu)e^{-\tilde{\chi}(r-\mu)} d\mu = \frac{V(\rho, \infty)}{\tilde{\chi}}$$

:) corresponds to the temperature state. Whence

$$\lim U(\rho, \tau) = 0$$

and

 $T = T_0$

i.e., after a very long time, the disintegration energy released from the waste is dissipated away into the surrounding medium, and the medium has everywhere its original temperature T_0 .

Relation 22 shows that by considering a decaying source of heat to be constant and of a strength equal to that at $\tau = 0$, the values of temperature rise will be overestimated by the amount

$$F = A_0 \lambda \int_0^\tau e^{-\lambda(\tau-\mu)} V(\rho, \mu) d\mu$$

$$F < A_0 \lambda V(\rho, \tau) \int_0^\tau e^{-\lambda(\tau-\mu)} d\mu \qquad (23)$$

$$= A_0 V(\rho, \tau) (1 - e^{-\lambda\tau})$$

Now the temperature rise for a constant heat generation A_0 is given by

$$U = A_0 V$$

Therefore

$$F/U < 1 - e^{-\lambda \tau} = 1 - e^{-\lambda \tau}$$
 (24)

Note that the expression on the right does not include R or A_0 , i.e., the percentage error depends only on t and λ and is independent of radioactive concentration of the waste or cavity size. The error will remain less than 10% if

$$1-e^{-\lambda t}\leq 0.1$$

i.e., when

$$t \leq 0.152 T_{1/2} \approx T_{1/2}/6$$

where $T_{1/2}$ denotes half-life of the isotopes constituting the waste. Therefore if we accept 95% of the temperature rise corresponding to A(t) $= A_t$ (which is the rate of heat generation of the waste at t = 0) as the probable value, the error introduced thereby will remain $\leq 5\%$ as long as $t \leq T_{1/2}/6$. A similar procedure shows that 90% of the value of the temperatures obtained by setting $A(t) = A_t$ involves an error of $\leq 10\%$ when $t \leq T_{1/2}/3$. Thus, for an old waste that mostly contains 96 Sr and 137 Cs (both have an almost equal half-life of about 29 years), the subsurface heating results will be reasonably accurate for the first 10 years.

Finally we consider the case of a waste containing *n* different types of isotopes. Let $A_{i,i}$ denote the generation of heat (cal cm⁻³ sec⁻¹) due to the isotope of type *s* at time t = 0. Then the heat generating function will be given by

$$A(\tau) = \sum_{s=1}^{n} A_{is} e^{-\lambda_{s} R^{s} t/k} = \sum_{s=1}^{n} A_{is} e^{-\lambda_{s} \tau}$$
(25)

and the expression for temperature will take the form

$$U(\rho, \tau) = \sum_{s=1}^{n} A_{0s} \left[V(\rho, \tau) - \lambda_{s} \right]$$
$$\cdot \int_{0}^{\tau} e^{-\lambda_{s}(\tau-\mu)} V(\rho, \mu) d\mu \left] \qquad (26)$$

where

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$$A_{0s} = A_{is}R^2/k$$

Relation 23 must now be replaced by

$$P_{\text{mixture}} = \sum_{s=1}^{n} A_{0s} \lambda_{s} \int_{0}^{\tau} e^{-\lambda_{s}(\tau-\mu)} V(\rho, \mu) d\mu
 P_{\text{mixture}} < V(\rho, \tau) \sum_{s=1}^{n} A_{0s}(1-e^{-\lambda_{s}\tau})$$

$$= V(\rho, \tau) \sum_{s=1}^{n} A_{0s}
 - V(\rho, \tau) \sum_{s=1}^{n} A_{0s} e^{-\lambda_{s}\tau}$$

Therefore

$$U > V(\rho, \tau) \sum_{s=1}^{n} A_{0s} e^{-X_s \tau}$$

But on account of relation 26 we have

$$U < \sum_{s=1}^{n} A_{0s} V(\rho, \tau)$$

Therefore we can write

$$V(\rho, \tau) \sum_{s=1}^{n} A_{0s} > U > V(\rho, \tau) \sum_{s=1}^{n} A_{0s} e^{-X_{s}\tau}$$

whence

$$U \approx \frac{V(\rho, \tau)}{2} \sum_{s=1}^{n} A_{0s} (1 + e^{-x_s \tau}) \qquad (28)$$

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This approximate relation underestimates the temperatures for short times, and overestimates them for long times.

Cylindrical Cavities

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We shall now consider a heat source having the form of a cylinder. It seems natural to investigate this case by using cylindrical coordinates. The corresponding heat equations can be written as

$$\frac{\partial T}{\partial t} = K \left[\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial z^2} \right] + A_i \quad (29)$$

$$0 \le r < R, \quad -b \le z \le b, \quad t > 0$$

$$c \frac{\partial T}{\partial t} = K \left[\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial z^2} \right] \quad (30)$$

$$|z| > b, \quad 0 < r < \infty, \quad t > 0$$

$$(|z| < b, \quad r > R, \quad t > 0)$$

where b denotes the half-length of the cylinder. These equations are independent of the direction of r and are valid for an isotropic and homogeneous medium. The method of Laplace transformation is not suitable for solving this problem because the equations derived by transformation are again partial differential equations. An expression for determining temperature rise for this case can, however, be obtained by employing the method of instantaneous heat sources [Carlslaw and Jaeger, 1959, p. 253]. This method deserves special consideration because a radioactive isotope is a physical example of an instantaneous source of heat.

The heat equation

Â M

$$\frac{\partial^2 U}{\partial x^2} + \frac{\partial^2 U}{\partial y^2} + \frac{\partial^2 U}{\partial z^2} = \frac{1}{k} \frac{\partial U}{\partial t} \qquad (31)$$

is satisfied by the function

$$U = \frac{A}{8(\pi kt)^{3/2}} \exp \left\{-\left[(x - x')^2 + (y - y')^2 + (z - z')^2\right]/4kt\right\}$$
(32)

Relation 32 can be interpreted as the temperature rise at a point (x, y, z) at time t due to an instantaneous point source located at (x', y', z') which releases Ac units of heat at t = 0. A is called the strength of the source, and c and k, respectively, denote the heat capacity per unit volume and diffusivity of the medium. If the source, instead of being instan-

taneous, continuously acts from t = 0 to t = t, being variable with time, the temperature rise from it will be given by

$$U = \frac{1}{8(\pi k)^{3/2}} \int_{0}^{t} \frac{A(t')}{(t-t')^{3/2}} \exp\left\{-\left[(x-x')^{2} + (y-y')^{2} + (z-z')^{2}\right]/4k(t-t')\right\} dt'$$
(33)

Let this source be represented by a radioactive substance confined around the point (x', y', z')in a volume element dx' dy' dz' and releasing heat at the rate of $A_i e^{-\lambda t'}$ cal cm⁻³ sec⁻¹. We further assume that the thermal properties of the radioactive substance are the same as those of the surrounding medium. Then equation 33 will be replaced by

$$U = \frac{A_{i}}{8c(\pi k)^{3/2}} \int_{0}^{t} \frac{e^{-\lambda t'}}{(t - t')^{3/2}}$$

$$\cdot \exp\left\{-\left[(x - x')^{2} + (y - y')^{2}\right]\right\}$$
(34)

$$+ (z - z')^{2}]/4k(t - t') \} dx' dy' dz' dt'$$

where A_i and λ are as previously defined.

When the radioactive material is uniformly distributed inside a cylinder, the resulting rise in temperature at any point (r, θ, z) and at time t can be written as

$$U = \frac{A_{i}e^{-\lambda t}}{8c(\pi k)^{3/2}} \int_{0}^{t} \int_{0}^{2\pi} \int_{-b}^{b} \int_{0}^{R} \frac{e^{\lambda \mu}}{\mu^{3/2}}$$

$$\cdot \exp \{-[r^{2} + r'^{2} - 2rr' \cos(\theta - \theta') (35) + (z - z')^{2}]/4k\mu\}r' dr' d\theta' dz' d\mu$$
where

$$x = r \cos \theta$$
 $x' = r' \cos \theta'$ (36)

$$y = r \sin \theta$$
 $y' = r' \sin \theta'$ (37)

and

$$= t - t' \tag{38}$$

ш Relation 35 can also be written as

$$U = \frac{A_{i}e^{-\lambda t}}{8c(\pi k)^{3/2}} \int_{0}^{t} \frac{e^{\lambda \mu}}{\mu^{3/2}} d\mu \int_{-b}^{b} \\ \cdot \exp\left[-(z - z')^{2}/4k\mu\right] dz' \\ \cdot \int_{0}^{R} \exp\left[-(r^{2} + r'^{2})/4k\mu\right]r' dr' \\ \cdot \int_{0}^{2\pi} \exp\left[2rr' \cos\left(\theta - \theta'\right)/4k\mu\right] d\theta'$$

 $\int_{-b}^{b} \exp\left[-\frac{1}{2}\right]$ $= (\pi k \mu)$ $\int_0^{2\pi} \exp\left[2\right]$ $\int_{0}^{R} \exp\left[-\frac{1}{2}\right]$ = 2RkWe finally $U = \frac{A_i R_i}{M_i}$ $\cdot \int_0^\infty e^{-k\mu}$ Once again parameter $\rho = r/R$ $\tau = kt/l$ Expressed takes the

We now i

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n differen be replace

 $U = \sum_{s=1}^{n}$

busly acts from t = 0 to t = t, with time, the temperature rise wen by

$$\frac{A(t')}{(t-t')^{3/2}} \exp \left\{-\left[(x-x')^{2}+(z-z')^{2}\right]/4k(t-t')\right\} dt'$$
(33)

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$$\frac{e^{-\lambda t'}}{(t-t')^{3/2}}$$

$$x')^{2} + (y-y')^{2} \qquad (34)$$

$$t-t') dx' dy' dz' dt'$$

re as previously defined. Detive material is uniformly a cylinder, the resulting rise any point (r, θ, z) and at ten as

$$\int_{0}^{2\pi} \int_{-b}^{b} \int_{0}^{R} \frac{e^{\lambda \mu}}{\mu^{3/2}}$$

$$\int_{0}^{2\pi} 2rr' \cos \left(\theta - \theta'\right) (35)$$

$$(\mu) r' dr' d\theta' dz' d\mu$$

$$x' = r' \cos \theta' \qquad (36)$$
$$y' = r' \sin \theta' \qquad (37)$$

$$= t - t' \tag{38}$$

so be written as `

$$\frac{\lambda \mu}{z^{3/2}} d\mu \int_{-b}^{b} dz'$$
$$- z')^{2}/4k\mu dz'$$
$$- (r^{2} + r'^{2})/4k\mu r' dr'$$

$$2rr'\cos(\theta - \theta')/4k\mu] d\theta'$$

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We now make use of the following relations [Carlslaw and Jaeger, 1959, p. 259]:

$$\int_{-b}^{b} \exp\left[-(z-z')^{2}/4k\mu\right] dz'$$

$$= (\pi k\mu)^{1/2} \left[\operatorname{erf} \frac{z+b}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{z-b}{(4k\mu)^{1/2}} \right] (39)$$

$$\int_{0}^{2\pi} \exp\left[2rr' \cos\left(\theta-\theta'\right)/4k\mu\right] d\theta'$$

$$= 2\pi I_{0} \left(\frac{rr'}{2k}\right) \qquad (40)$$

$$\exp\left[-(r^{2}+r'^{2})/4k\mu\right]I_{0}\left(\frac{rr'}{2k\mu}\right)r'\,dr'$$
$$=2Rk\mu\int_{-\infty}^{\infty}e^{-k\mu\alpha}J_{0}(r\alpha)J_{1}(R\alpha)\,d\alpha\qquad(41)$$

$$= 2Rk\mu \int_0^{\infty} e^{-k\mu\alpha^2} J_0(r\alpha) J_1(R\alpha) \ d\alpha$$

Ve finally get

$$U = \frac{A_{i}Re^{-\lambda t}}{2c}$$

$$\cdot \int_{0}^{t} \left[e^{\lambda \mu} \left(\operatorname{erf} \frac{z+b}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{z-b}{(4k\mu)^{1/2}} \right) \right]$$

$$\cdot \int_{0}^{\infty} e^{-k\mu \alpha^{*}} J_{0}(r\alpha) J_{1}(R\alpha) d\alpha d\alpha d\mu \qquad (42)$$

Once again we introduce a set of dimensionless parameters:

 $\rho = r/R$ $\zeta = z/R$ h = b/R $\tau = kt/R^2$ $A_0 = A_i R^2/K$ $\lambda = \lambda R^2/k$ Expressed in these parameters, equation 42 takes the form

$$U = \frac{A_0 e^{-\lambda \tau}}{2}$$

$$\cdot \int_0^{\tau} e^{\lambda \mu} \left[\left(\operatorname{erf} \frac{\zeta + h}{2\sqrt{\mu}} - \operatorname{erf} \frac{\zeta - h}{2\sqrt{\mu}} \right) \right]$$

$$\cdot \int_0^{\infty} e^{-\mu \alpha^*} J_0(\rho \alpha) J_1(\alpha) \, d\alpha d\alpha d\mu$$
(43)

When we are considering a mixture containing n different types of isotopes, relation 43 must be replaced by

$$U = \sum_{n=1}^{n} \frac{A_{0n} e^{-\chi_{n}\tau}}{2}$$

$$\cdot \int_{0}^{\tau} e^{\chi_{n}\mu} \left[\left(\operatorname{erf} \frac{h+\zeta}{2\sqrt{\mu}} - \operatorname{erf} \frac{h-\zeta}{2\sqrt{\mu}} \right) \right]$$

$$\cdot \int_{0}^{\infty} e^{-\mu\alpha} J_{0}(\rho\alpha) J_{1}(\alpha) d\alpha d\alpha d\alpha$$

Relation 44 gives the temperature rise in and outside a cylinder of finite length filled with a waste of arbitrary radioactive composition.

From the symmetry of the problem it follows that the highest temperature is reached at the point ($\rho = 0, \zeta = 0$); that is,

$$U|_{\substack{\rho=0\\\varsigma=0}} = \sum_{s=1}^{n} A_{0s} e^{-\chi_{s\tau}} \int_{0}^{\tau} e^{\chi_{s\mu}} \operatorname{erf} \frac{h}{2\sqrt{\mu}}$$
$$\cdot \int_{0}^{\infty} e^{-\mu\alpha^{s}} J_{1}(\alpha) \ d\alpha \ d\mu \qquad (45)$$

By using the relation

$$\int_0^\infty e^{-\mu\alpha^*} J_1(\alpha) \ d\alpha = 1 - e^{-1/(4\mu)} \qquad (46)$$

equation 45 can be expressed in a simpler form

$$U|_{\substack{\rho=0\\ f=0}} = \sum_{s=1}^{n} A_{0s} e^{-\chi_{s}\tau}$$
$$\cdot \int_{0}^{\tau} \operatorname{erf} \frac{h}{2\sqrt{\mu}} e^{\chi_{s}\mu} (1 - e^{-1/(4\mu)}) d\mu \qquad (47)$$

And the highest temperature in the surrounding medium will be reached at the point ($\rho = 1, \zeta = 0$).

When the length of the cylinder is much greater than its radius, it can be treated as an infinite cylinder. The temperature rise for an infinite cylinder can be obtained from relation 44 by setting $h = \infty$:

$$U = \sum_{s=1}^{n} A_{0s} e^{-\chi_{s\tau}} \int_{0}^{\tau} e^{\chi_{s\mu}}$$
$$\cdot \int_{0}^{\infty} e^{-\mu\alpha^{s}} J_{0}(\rho\alpha) J_{1}(\alpha) \ d\alpha \ d\mu \qquad (48)$$

The temperatures everywhere along the axis of an infinite cylinder and also at all points that are equidistant from the axis are the same. The axial temperature is given by

$$U = \sum_{s=1}^{n} A_{0s} e^{-\chi_{s}\tau} \int_{0}^{\tau} e^{\chi_{s}\mu} \int_{0}^{\infty} e^{-\mu\alpha^{2}} J_{1}(\alpha) \, d\alpha \, d\mu$$
$$U = \sum_{s=1}^{n} A_{0s} e^{-\chi_{s}\tau} \int_{0}^{\tau} e^{\chi_{s}\mu} (1 - e^{-1/4\mu}) \, d\mu \quad (49)$$

Cavities of Other Shapes

(44)

In actual practice, a cavity that may be considered for waste disposal may not always correspond to a perfect sphere or a regular cylinder. It could be, for example, a hollow

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space in an abandoned salt mine from which salt has been excavated, or a naturally developed solution cavity within a body of limestone meeting the conditions of environmental safety. It could also be one or more porous layers on a synclinal structure or a system of layers artificially created in the subsurface such as those resulting from hydraulic fracturing. Therefore it would be of interest to consider those cavities that can neither be approximated to a sphere nor to a cylinder.

Parallelopiped. The temperature field for a parallelopiped ($-a \le x \le a, -b \le y \le b, -d \le z \le d$) can be easily derived from equation 34 by integration. The resulting expression is

$$U = \sum_{s=1}^{n} \frac{A_{1s} e^{-\lambda_{s}t}}{8c(\pi k)^{3/2}} \int_{0}^{t} \int_{-a}^{a} \int_{-b}^{b} \int_{-d}^{d} \frac{e^{\lambda_{s}\mu}}{\mu^{3/2}}$$
$$\cdot \exp \left\{ -[(x - x')^{2} + (y - y')^{2} + (z - z')^{2}]/4k\mu \right\} dz' dy' dx' d\mu$$

where

$$u = t - t'$$

. It can be written in the form

$$U = \sum_{i=1}^{n} \frac{A_{i,e} e^{-\lambda_{i}t}}{8c(\pi k)^{3/2}} \int_{0}^{t} \frac{e^{\lambda_{i}\mu}}{y^{3/2}} d\mu$$

$$\cdot \int_{-a}^{a} \exp\left[-(x - x')^{2}/4k\mu\right] dx'$$

$$\cdot \int_{-b}^{b} \exp\left[-(y - y')^{2}/4k\mu\right] dy'$$

$$\cdot \int_{-d}^{d} \exp\left[-(z - z')^{2}/4k\mu\right] dz'$$

$$U = \sum_{i=1}^{n} \frac{A_{i,e} e^{-\lambda_{i}t}}{8c} \int_{0}^{t} e^{\lambda_{i}\mu} \left[\left(\operatorname{erf} \frac{x + a}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{y - b}{(4k\mu)^{1/2}} \right) \right] \left(\operatorname{erf} \frac{z + d}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{z - d}{(4k\mu)^{1/2}} \right) \right] d\mu \quad (50)$$

By setting a = b = d in equation 50, one gets temperatures for a cavity of cubical shape. The temperatures at the center of the cube are given by

$$U = \sum_{s=1}^{n} \frac{A_{is} e^{-\lambda_{i}t}}{c} \int_{0}^{t} e^{\lambda_{i}\mu} \left(\operatorname{erf} \frac{a}{(4k\mu)^{1/2}} \right)^{3} d\mu$$
(51)

The maximum temperature that the surrounding medium will attain will be at the center of each side of the cube, for example, at the point (x = a, y = 0, z = 0); it can be expressed as

$$U_{\max} = \sum_{i=1}^{n} \frac{A_{is} e^{-\lambda_i t}}{2c} \int_0^t e^{\lambda_i \mu} \\ \cdot \operatorname{erf} \frac{2a}{(4k\mu)^{1/2}} \left(\operatorname{erf} \frac{a}{(4k\mu)^{1/2}} \right)^2 d\mu \qquad (52)$$

The minimum temperature at the surface of the cube will be at the corners, such as the point (x = a, y = a, z = a):

$$U_{\min} = \sum_{i=1}^{n} \frac{A_{ii} e^{-\lambda_{i} t}}{8c} \int_{0}^{t} e^{\lambda_{i} \mu} \left(\operatorname{erf} \frac{2a}{(4k\mu)^{1/2}} \right)^{3} d\mu$$
(53)

Thus the temperature field close to a cubical cavity ranges between a number of maximums and minimums.

A layer of infinite extension and uniform thickness. A layer of infinite extension and uniform thickness can be regarded as a rectangular parallelopiped with any two of its opposite faces of infinite extension. The temperatures for such layers can therefore be readily obtained from equation 50 by taking the limit as a and b approach infinity. We get as a result

$$U = \sum_{s=1}^{n} \frac{A_{is} e^{-\lambda_{s}t}}{2c}$$
$$\cdot \int_{0}^{t} e^{\lambda_{s}\mu} \left[\operatorname{erf} \frac{z+d}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{z-d}{(4k\mu)^{1/2}} \right] d\mu \quad (54)$$

A system of parallel layers of infinite extension. An interesting possibility for disposing of waste into the subsurface is disposal by means of hydraulic fracturing. Let the number of layers be denoted by m. Let the thickness of each layer be denoted by 2d and the separation interval between the layers by f. If each layer is filled with radioactive waste, then the zones of heat generation will be as follows: The temperat be given by

n

$$U_{p} = \sum_{s=1}^{n} \frac{1}{1-s}$$
$$\int_{0}^{t} e^{\lambda_{st}}$$
$$- \operatorname{erf} \frac{z}{1-s}$$
Therefore for can write
$$U = \sum_{s=1}^{n} \frac{A_{st}}{1-s}$$
$$\cdot \sum_{p=1}^{m} \left[\operatorname{erf} \frac{z}{1-s} \right]$$

The derivation assumption the waste a layers are find being filled a heat generat ferent times t_0, t_1, \ldots, t_0 $t_0 < t_1 <$ The express case can be $U = \sum_{r=1}^{n} \sum_{p=1}^{n}$

- erf $\frac{z}{z}$

ares at the center of the cube are

$$\frac{-\lambda_{\star,\iota}}{2} \int_0^{\iota} e^{\lambda_{\star,\mu}} \left(\operatorname{erf} \frac{a}{(4k\mu)^{1/2}} \right)^3 d\mu$$
(51)

temperature that the surroundil attain will be at the center of e cube, for example, at the point z = 0; it can be expressed as

$$\frac{e^{-\lambda_{\star t}}}{2c} \int_0^t e^{\lambda_{\star \mu}}$$

$$\frac{a}{d^{1/2}} \left(\operatorname{erf} \frac{a}{(4k\mu)^{1/2}} \right)^2 d\mu \qquad (52)$$

temperature at the surface of De at the corners, such as the = a, z = a:

$$\frac{e^{-\lambda_{\star}t}}{8c} \int_0^t e^{\lambda_{\star}\mu} \left(\operatorname{erf} \frac{2a}{(4k\mu)^{1/2}} \right)^3 d\mu$$
(53)

erature field close to a cubical between a number of maximums.

infinite extension and uniform ayer of infinite extension and ess can be regarded as a recacopiped with any two of its opinfinite extension. The temperaayers can therefore be readily equation 50 by taking the limit oproach infinity. We get as a

$$\frac{d}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{z-d}{(4k\mu)^{1/2}} d\mu \quad (54)$$

parallel layers of infinite exeresting possibility for disposo the subsurface is disposal by the fracturing. Let the number ated by m. Let the thickness of noted by 2d and the separation the layers by f. If each layer moactive waste, then the zones on will be as follows:



The temperature rise due to the *p*th layer will be given by

$$U_{p} = \sum_{s=1}^{n} \frac{A_{is}e^{-\lambda_{s}t}}{2c} \\ \cdot \int_{0}^{t} e^{\lambda_{s}\mu} \left[\operatorname{erf} \frac{z - (2p - 3)d - (p - 1)j}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{z - (2p - 1)d - (p - 1)f}{(4k\mu)^{1/2}} \right] d\mu$$

Therefore for the entire system of layers, we can write

$$U = \sum_{s=1}^{n} \frac{A_{is} e^{-\lambda_{s}t}}{2c} \int_{0}^{t} e^{\lambda_{s}\mu}$$

$$\cdot \sum_{p=1}^{m} \left[\operatorname{erf} \frac{z - (2p - 3)d - (p - 1)f}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{z - (2p - 1)d - (p - 1)f}{(4k\mu)^{1/2}} \right] d\mu \quad (55)$$

The derivation of relation 55 is based on the assumption that all the layers are filled with the waste at the same time. If the various layers are filled at different times, each layer being filled subsequent to the one below it, the heat generation in each layer will start at different times. Let these times be denoted by $t_0, t_1, \ldots, t_{p-1}, \ldots, t_{m-1}$ for the first, second, \ldots , mth layer, such that

$$t_0 < t_1 < t_2 < \cdots < t_{p-1} < \cdots < t_{m-1}$$

The expression for temperature rise for this case can be written as

$$U = \sum_{s=1}^{n} \sum_{p=1}^{m} \frac{A_{is}}{2c} \exp\left[-\lambda_{s}(t-t_{p-1})\right]$$
$$\cdot \int_{t-p}^{t} e^{\lambda_{s}\mu} x \left[\operatorname{erf} \frac{z-(2p-3)d-(p-1)f}{(4k\mu)^{1/2}} - \operatorname{erf} \frac{z-(2p-1)d-(p-1)f}{(4k\mu)^{1/2}} \right] d\mu \quad (56)$$

This expression reduces to equation 54 by setting $t_0 = 0$ and m = 1. One should also note that t appearing in relation 56 must be greater than t_{m-1} . The situation $t < t_{m-1}$ implies that the mth layer either does not exist or has not yet been filled with the waste.

Cavities of irregular shape. All cavities of irregular shape can be treated by considering them as consisting of a number of parallelopipeds and superposing the temperature fields resulting from each.

Throughout our discussion, we have treated the waste as a solid. Therefore, strictly speaking, the theory presented above is applicable only to those wastes that have been reduced by necessary treatment to solid form either before or immediately after their disposal into the subsurface.

NUMERICAL EVALUATION OF TEMPERATURES AND RESULTS

We shall give below an account of the numerical evaluation of temperatures based on the theory presented in the above section on temperature rise. The calculations were carried out by using an IBM digital computer. As we mentioned in the beginning, most of the numerical work refers to rock salt.

Spherical Heat Sources

The temperatures for this case were obtained by using relations 13, 14, and 22. For computing i^2 erfc x and i^3 erfc x appearing in relations 13 and 14, the following formulas [Carlslaw and Jaeger, 1959, p. 484] were employed:

$$i \operatorname{erfc} x = \frac{1}{\sqrt{\pi}} \exp(-x^2) - x \operatorname{erfc} x \quad (57)$$

$$2ni^{n} \operatorname{erfc} x = i^{n-1} \operatorname{erfc} x - 2xi^{n-1} \operatorname{erfc} x \quad (58)$$
$$(n = 2, 3, \cdots)$$

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The function erfc x was computed by using the following series [Jahnke et al., 1960]:

erfc
$$x = 1 - \frac{2}{\pi} \sum_{n=0}^{\infty} \frac{(-1)^n x^{2n+1}}{(2n+1)n!}$$
 $x \le 2.5$
erfc $x = \frac{e^{-x^2}}{\sqrt{\pi x}} \left[1 - \frac{1}{2x^2} + \frac{1 \cdot 3}{(2x^2)^2} - \frac{1 \cdot 3 \cdot 5}{(2x^2)^3} + \cdots \right]$ (59)
(2.5 < x < 7.0)
erfc $x = 0$ $(x \ge 7.0)$

The temperatures were calculated for different sets of values of ρ , λ , and τ . In each case, A_0 was taken as unity. Temperatures for any other value of A_0 can be obtained by simple multiplication.

For determining a suitable range within which the practical values of λ would lie, the following ranges for λ , k, and R were adopted (both values of k are from *Birch*, 1959):

- $\lambda_{\max} = 5.06/\text{year}$ (corresponding half-life, 50 days).
- $\lambda_{\min} = 0.023/\text{year}$ (corresponding half-life, 30 years).

 $k_{\text{max}} = 10.5 \times 10^{5} \text{ cm/year}$ (diffusivity of salt at 0°C).

 $k_{\min} = 30.0 \times 10^4$ cm/year (diffusivity of salt at 400°C).

 $R_{\rm max} = 1000$ cm.

 $R_{\min} = 50 \text{ cm.}$

Corresponding to these ranges of λ , k, and R, relation 21 yields

$$\lambda_{\rm max} = 16.88 \qquad \lambda_{\rm min} = 5.5 \times 10^{-1}$$

Therefore λ should range from 5.5 \times 10⁻⁵ to 16.88. Preliminary calculations revealed that the data corresponding to $\lambda < 4.0 \times 10^{-4}$ differed very little from data obtained by assuming a constant rate of heat generation, and therefore the smallest value of λ for which calculations were done is 4.0 $\times 10^{-4}$.

The results for $\rho = 1$, i.e., r = R, are presented in the form of a diagram (Figure 1), showing U/A_0 as a function of τ for different values of λ . The curve corresponding to $\lambda = 0$ (infinite halflife) yields temperatures for a constant rate of heat generation. Figure 1 can be used for a rapid determination of temperatures at the surface of a cavity containing waste of arbitrary radioactive composition. The procedure is quite straightforward and consists of the following steps:

1. From a knowledge of the decay constants and concentrations of various isotopes that are present in significant proportions in the waste, calculate the values of λ and A_0 .

2. Corresponding to each value of λ computed, read off U/A_0 from the diagram for a given value of τ .

3. Multiply each value read by the corresponding value of A_0 and add the results. That gives temperature rise at a given time.



Fig. 1. Temperature rise at the surface of an exponentially decaying spherical heat source as a function of τ .

4. Repeat steps to obtain the ten of time.

It can be seen f very large values be encountered in pretty close to the the temperature in that the difference cases increases rawe are interested is surface waste diconstant rate of errors.

Since the range of λ arising in proon the curves protemperatures mus To avoid this different that give the tenfunction of λ for presented in Figgrams refers to a look quite similar say Figure 5, moture in the surr of 3 times the cartemperature for



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 $\frac{\log 1}{\log k}$ to these ranges of λ , k, and R, $\frac{\log 1}{\log k}$

.88
$$\lambda_{\min} = 5.5 \times 10^{-5}$$

mould range from 5.5×10^{-5} to mary calculations revealed that sounding to $\lambda < 4.0 \times 10^{-4}$ difinform data obtained by assuming of heat generation, and therefore value of λ for which calculations $\lambda \times 10^{-4}$.

The procedure is quite straightconsists of the following steps: $r \rho = 1$, i.e., r = R, are presented a diagram (Figure 1), showing thion of τ for different values of λ . Asponding to $\lambda = 0$ (infinite halfconstant rate of constant rate of a constant rate of constant rate of a constant rate of constant rate of the surface constant rate of arbitrary radioacconstant rate of the surface straightconsists of the following steps:

mowledge of the decay constants from of various isotopes that are fricant proportions in the waste, frues of λ and A_{0} .

and add the results. That gives at a given time.



ing spherical heat source as a

4. Repeat steps 1 to 3 for several values of τ to obtain the temperature rise as a function of time.

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It can be seen from Figure 1 that, except for very large values of λ (which are not likely to be encountered in practice), all the curves run pretty close to the curve for $\lambda = 0$ as long as the temperature maximum is not reached; after that the difference in temperatures in the two cases increases rapidly. This means that when we are interested in the long-term effects of subsurface waste disposal, the assumption of a constant rate of heat generation leads to large errors.

Since the range of λ is very large, the values of λ arising in practical cases will not likely fall on the curves presented in Figure 1, and the temperatures must be obtained by interpolation. To avoid this difficility, diagrams were prepared that give the temperature rise as a continuous function of λ for a set of values of τ . They are presented in Figures 2 to 7. Each of these diagrams refers to a certain value of ρ . All of them look quite similar. Let us examine one of them, say Figure 5, more closely. It gives the temperature in the surrounding medium at a distance of 3 times the cavity radius. To each value of the temperature for a given λ , two values of τ correspond. The smaller value gives the time at which the medium will first reach a given temperature during the process of heating, and the larger value gives the time when it returns to this temperature during cooling. In this manner one knows how long various points of the surrounding medium will remain at temperatures higher than a given temperature. Since salt becomes very plastic at high temperatures, this information is of great importance for investigating possible plastic flow of the salt when subjected to radioactive heating. To each value of λ , there is only one τ curve that is touched by the envelope tangentially. This curve gives the time at which the maximum temperature will be reached.

Cylindrical Heat Sources

The temperature field for an exponentially decaying cylindrical heat source of finite length is given by relation 43. On account of equation 16, the evaluation of the error function terms in 43 was done using equation 59, explained previously. For computing the integral

$$\psi(\mu) = \int_0^\infty e^{-\mu\alpha^*} J_0(\rho\alpha) J_1(\alpha) \ d\alpha$$

which also appears in equation 43, the follow-



Fig. 2. Temperature rise at the center of an exponentially decaying spherical heat source as a function of X. The numbers on the curves denote corresponding values of τ .





ing relations for $\psi(0)$ [Magnus and Oberhettinger, 1948] were used:

$$\int_{0}^{\infty} J_{0}(\rho\alpha) J_{1}(\alpha) \ d\alpha = \begin{cases} 1.0 & \text{for } \rho < 1 \\ 0.5 & \text{for } \rho = 1 \\ 0.0 & \text{for } \rho > 1 \end{cases}$$

It is obvious from relation 43 that the temperatures for a cylindrical source depend not only on τ , λ , and ρ but also on two additional parameters h and ζ . The temperature field from a cylindrical source is therefore much more complicated. Preliminary calculations, however, revealed that for practical purposes a cylinder whose length is 5 times or more its diameter can be treated as an infinite cylinder. In such case, the temperatures depend only on τ , λ , and ρ . In other words, the number of practical cases for cylindrical heat sources is quite limited. Several diagrams for the determination of temperature field in and outside an exponentially decaying cylindrical heat source were computed [Multi, 1966]. They can be used in the same manner as those presented for the spherical sources, and they yield similar information.

A Practical Example

As an example, we shall apply the theoretical diagrams of Figures 2 to 7 for calculating the temperatures in and outside a spherical salt cavity filled with the waste X mentioned in the second section. Let us assume the following data:

| Age of the waste | = | 2 years. |
|------------------|----|--|
| T_{0} | = | 0°C. |
| R | == | 2 meters. |
| k | = | $6.4 \times 10^5 \text{ cm}^2/\text{year} =$ |
| | | $0.02 \text{ cm}^2/\text{sec.}$ |
| K | = | 0.0086 cal/cm sec °C. |
| τ | = | $kt/R^2 = 16t$, when t is in |
| | | years. |

The value of A_0 for various isotopes present in the waste can be determined by using Table 2.

The results of the calculations are presented in Figure S, which shows the evaluation of the temperature at the center of the cavity for each of the isotope groups present in the waste. The temperature rise due to the waste X is obtained by summing up the contribution from these isotope groups. Although "Ce is much



contribution is "Sr and ""Cs the activity at of "Ce are ver half-life, however more rapidly. A from all the ""Cs becomes the Figure 9 shift temperature fit The temperature for the tempera





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- = 2 years.
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= 2 meters.

- $= 6.4 \times 10^5 \text{ cm}^2/\text{year} = 0.02 \text{ cm}^2/\text{sec.}$
- = 0.0086 cal/cm sec °C. = kt/R^2 = 16t, when t is in years.

r various isotopes present in etermined by using Table 2. he calculations are presented shows the evaluation of the e center of the cavity for groups present in the waste. See due to the waste X is obtrup the contribution from ps. Although "Ce is much



Fig. 4. Temperature rise at $\rho = 2$ from an exponentially decaying spherical heat source as a function of λ . The numbers on the curves denote corresponding values of τ .

shorter-lived than 60 Sr and 137 Cs, its thermal contribution is much greater even than that of 60 Sr and 137 Cs together. This is because both the activity and the energy of disintegration of 147 Ce are very high. On account of its shorter half-life, however, the effect of 144 Ce dies away more rapidly. At t = 10 years, the contribution from all the isotope groups except 60 Sr and 137 Cs becomes negligible.

Figure 9 shows the complete picture of the temperature field and its variation with time. The temperature curves correspond to various distances from the cavity center. There is a regular shift of the point of maximum temperature away from the center of the cavity. The growth of temperature is much faster than its decay. Thus at the surface of the cavity, the temperature rises from 85° C to a maximum value of 340° C in 69 days, but more than 4 years are required for it to drop to 85° C again. The results for $\rho = 10$ show that the temperatures at a distance of 20 meters from the cavity center are very low. The maximum temperature there is only 8° C and is reached 3 years after the waste is disposed of. These results show that, once the salt medium is







Fig. 6. Temperature rise at $\rho = 4$ from an exponentially decaying spherical heat source as a function of λ . The numbers on the curves denote corresponding values of τ .

heated by the waste, it requires a very long time to cool down (for the case under consideration, it requires many years) and the temperature field penetrates very slowly and only up to a very limited distance into the surrounding medium.

Similar calculations of the temperature field when the waste X at the time of disposal is 1 year old, show [*Mufti*, 1966] that the maximum temperature that would be attained by the surrounding salt medium is 760°C. Since salt melts at this temperature, the theory presented is not valid for those conditions.

Heating of salt beyond its melting point may endanger the structural integrity of the cavity. However, even if the melting of salt were to be restricted to a zone close to the cavity surface, another difficulty is met. The melting may lead





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Fig. 8. Contribution of various isotopes to the temperature rise at the center of a salt cavity of 2-meter radius containing the waste X.

to a firm attachment of the waste to the salt mass when the molten salt solidifies. In fact, experimental studies by *Kappelmeyer* [1966] indicate that a solid body attached to salt by resolidification is extremely hard to detach. Therefore such a procedure of waste disposal should be considered only at those sites where a need for eventual recovery of the waste for emergency or other reasons is not foreseen.

Conclusions

The properties of the temperature field in and outside a subsurface cavity containing radioactive waste were investigated in detail. Cavities of spherical, cylindrical, and other shapes were considered. The results for spherical cavities were presented in several diagrams. These diagrams can be conveniently used for rapid calculations of temperatures for spherical cavities of different sizes containing waste of arbitrary radioactive composition.

As an example, the diagrams were used for calculating the temperature field for a spherical cavity of 2-meter radius located in a salt medium and containing a high-level nuclear reactor waste. Even for a cavity of such small size, the calculations reveal that when the waste is 2 years old at the time of emplacement, the maximum temperature that would be reached at the surface of the cavity is 340°C; when the waste is 1 year old, the resulting temperatures would be sufficient to melt the surrounding salt. The increase of the temperature is much faster than its decay. Therefore the salt medium, once heated by the waste, requires a much longer time to cool down, which for a large cavity may mean several decades or even more. The temperature field extends very slowly and is effective only up to a limited distance into the surrounding medium. This indicates that when several cavities are to be used, the geothermal changes brought about by the waste are not likely to produce significant changes in major geologic structures such as a salt dome if the distance between cavities is larger than the effective penetration for a single

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cavity. On the other hand, a system of cavities located too close together will behave like a single large cavity whose geothermal field will be more persistent and deserves a considerable amount of further theoretical and experimental investigation to ascertain its safety.

APPENDIX: LIST OF SYMBOLS

- A_i is the rate of heat generation at an initial time in cal cm⁻³ sec⁻¹ at t = 0. $A_0 = A_i R^2/K$, °C.
- A(t) is the strength of a time-dependent heat source, cm³ °C.
 - a is the half-length of a parallelopiped, cm.
 - b is the half-length of a cylinder or the half-width of a parallelopiped, cm.
 - C_x is the activity of the radioisotope x, c cm⁻³.
 - c is the heat capacity per unit volume, cal cm⁻³ °C⁻¹.
 - *d* is the half-thickness of a parallelopiped or the half-thickness of a layer, cm.
 - e_x is the energy released per curie by

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- K is thermal conductivity, cal $cm^{-1} sec^{-1}$ °C⁻¹.
- k is thermal diffusivity, $cm^2 sec^{-1}$.
- l is a fraction, 0 < l < 1.
- *m* is the number of layers constituting a system of parallel layers.
- n is the number of types of radioisotopes present in significant proportions in a waste at a given time.
- R is the radius of a sphere or a cylinder, cm.



Fig. 9. Temperature field as a function of time in and outside a salt cavity of 2-meter radius containing the waste X.

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coisotope x, cal sec⁻¹.

_ne error function.

- The complementary error function $1 \operatorname{erf} x$).
- ne separation interval between barallel layers, cm. R.
- $_$ modified Bessel function of the kind and of order n.
- Bessel function of the first kind $\neg f$ order *n*.

ermal conductivity, cal cm⁻¹ sec⁻¹

number of layers constituting stem of parallel layers.

- e number of types of radioisotopes ent in significant proportions in use at a given time.
- radius of a sphere or a cylinder,



salt cavity of 2-meter radius

- RADIOACTIVE WASTE DISPOSAL
- is one of the spherical or cylindrical coordinates, cm.
- s is the Laplace transformation parameter.
- $T_{\frac{1}{2}}^{\frac{1}{2}}$ is the half-life of a radioisotope, sec.
- T is temperature, °C.
- T_{\circ} is temperature at an initial time, t = 0, in °C.
- t is time, sec.
- U is temperature rise = $T T_0$, in °C.
- $u(\rho, s)$ is the Laplace transform of the function $U(\rho, \tau)$.
 - z is one of the cylindrical coordinates, cm.
 - λ is the decay constant, sec⁻¹.
 - $\lambda = R^2 \lambda / k.$
 - $\mu = t t', \text{ sec.}$
 - $\rho = r/R.$
 - $\tau = kt/R^2.$
 - $\zeta = z/R.$

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