Heat Transfer and Modelling Studies for the Analysis of Waste Storage Facilities

by

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Dedication

To the fond memory of my beloved father.

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CHAPTER 1

INTRODUCTION

Reprocessing of spent nuclear fuel to reclaim depleted uranium and plutonium through chemical separation leads to generation of high-level radioactive wastes which are thermally hot, besides being highly radioactive. Elevated temperature is one of the major problems encountered in the storage of these wastes. More than 80 million gallons of high-level radioactive wastes have been accumulated in the U.S., part of which have been converted to solids by calcination, pending final conversion to stable and nonleachable waste forms. These thermally hot high-level liquid wastes and converted calcines are typically stored in an interim manner (usually less than five years) to reduce the heat generation in order to make them amenable for the final conversion to stable waste forms. These wastes are currently stored in relatively complex and therefore expensive facilities and as the nuclear industry grows, the volume and hence the total cost of waste management sharply increases. So a search for safe and less expensive methods of waste storage is essential.

At present, the cooling of these wastes - necessary because of the heat generated by the decay of radioactive fission products in the wastes - is accomplished by forced circulation in secondary heat-exchangers cooled externally (for liquid wastes) and by convective air cooling (for the solid calcines). Present storage facilities consist of a central container or containers surrounded completely by an outer vault, which is buried in soil at varying depths. A significant reduction in cost of future storage facilities would be achieved if the internal cooling systems and the outer containment vault could be omitted. For such a simplified waste storage facility, all of the heat generated would be removed by conduction through the soil surrounding the single waste container or cluster of individually buried waste storage containers. Because of the fission product volatility and the limits of the container material, most nuclear wastes are not permitted to exceed a specific maximum in-storage temperature. For the liquid wastes, the maximum temperature limit is imposed by the container structural stability and the boiling limit of the alkaline supernatents in tank, so as not to uncover the salt layer lying underneath which has a much higher heat generation rate. This could result in temperature build-up and thereby threaten the liner integrity. As for the calcines, the temperature limits are dictated by the allowable calcine sintering temperature and collapse of the calcine bed during such abnormal increases in bin temperatures.

The objective of this research is to develop heat transfer models for simplified geometries and evolve solutions for the temperatures in the storage containers. Previous studies by Dickey et al.[1] had assumed a solid cylinder configuration for such underground storage and had evaluated temperature profiles in the storage units as well as the surrounding soil using finite transforms. The geometry of their model had been a circular solid cylinder having finite depth. The scope of this research work is an extension of their model with annular configuration, to see what effect the configuration has on the resulting temperature profile, introducing air in the annulus. Also pursued are the dependence of other physical parameters, such as thermal conductivities (of the waste, soil surrounding the storages), heat generation rates of the waste, and the proportioning of the waste storage bins on the resulting temperature profile. The model predicts the temperature distribution in the tank and the adjoining soil asauming steady state conditions and idealized material having thermal properties independent of position or temperature. Thus, the result must not be regarded as a definitive deacription of the true thermal characteristics of any operational system and the reaults muat only be regarded as a reference system to compare analyais of real cases.

Interim Storage of High-Level Wastes:

Problems involved in interim storage of high-level wastes are due to the decay heat evolved by the wastes, solids present in the waste, the corrosiveness of the waste (particularly if stored in acidic form), radiolysis effects due to high radioactivity and, to a much lesser extent, to the potential for volatilization of the constituents. Concentrated and stored high-level wastes will boil because of the radioactive decay heat generated in them, causing several problema that must be accounted for in the design and operation of such storage systems.

In the early years of operation of the reprocessing plants, particularly at the Hanford Reservation, the concrete tanks built to store these solutions had single walled carbon steel liners, which were susceptible to acid attack. The hot acidic solutions were therefore neutralized with alkali, which increased their volumes and formed a precipitate, the so called 'sludge' containing the bulk of the radioactive materials. The thermally hot, alkaline, highly radioactive mixture of liquid-plus-sludge increased stress corrosion cracking of steel and over the years several of these liners have developed leaks.

To minimize leaks and to reduce volume of the wastes further, a program of evaporating the liquid to salt cake was undertaken. Preliminary separation of most of the radioactive strontium and cesium from hot solutions was necessary to prevent over heating of salt cakes. Thus, the high-level wastes at present consist of liquid, sludge, and salt cake capsules of radioactive strontium and cesium salts and the hot terminal liquor.

Most of the high-level waste in the U.S. is the result of DOE (Department of Energy) activities in the area of national defense and is stored at the Savannah River Plant (SRP), South Carolina, Idaho National Engineering Laboratory (INEL), Idaho Falls, and the Hanford Reservation (HANF). At the Savannah River Plant and the Hanford Reservation, the acidic wastes are neutralized, dehydrated and then stored as damp crystalline salts, sludge and the supernatent liquid. Radioactive cesium and strontium have been separated from the high heat producing wastes at the Hanford Reservation and are encapsulated and stored in a water cooled basin. The volume, radioactivity and heat generation of high-level wastes accumulated through the year 1981 in the U.S. including the current inventories of high-level waste in storage at the end of 1981 are shown in Table 1.1.

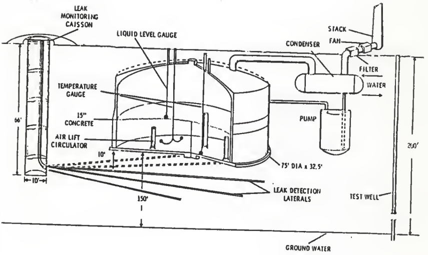
Liquid Wastes:

Typically, these high-level wastes, which are thermally hot, are stored in underground tanks which are 76 ft in diameter, 36 ft high, double walled in construction, and stress relieved. Figure 1.1 shows typical construction details of these storage tanks which are housed in concrete vaults. A number of such tanks are laid out in what are known

Table 1.1. Current Inventories of HLW in Storage by Site as of December 31, 1981.

Site	Volume (10^3 m^3)						Radioactivity (MCi)	Heat Generation rate (KW)
	Liquid	Sludge	Salt Cake	Calcine	Sr, Cs Capsules	Total		
Defense								
Savannah River Plant	66.3	11.8	27.6	--	--	105.7	982	2.90
Idaho Chemical Processing Plant	9.8	--	--	2.2	--	12.0	63.5	0.21
Hanford	39.0	49.0	95.0	--	0.0017	183	531	1.64
Subtotal	115.1	60.8	122.6	2.2	0.0017	300.7	1,577	4.75
Commercial								
Nuclear Fuel Services								
Alkaline waste	2.1	0.047	--	--	--	2.147	36.4	0.106
Acid waste	0.045	--	--	--	--	0.045	3.0	0.009
Subtotal	2.145	0.047	--	--	--	2.192	38.4	0.115
Grand Total	117.2	60.8	122.5	2.2	0.0017	302.8	1,616.3	4.87

SOURCE: Ref. 23, Table 2.3



SOURCE: Ref.22, Figure II.1-43.

Fig. 1.1. Typical Control and Safety Features for Boiling High-level Wastes.

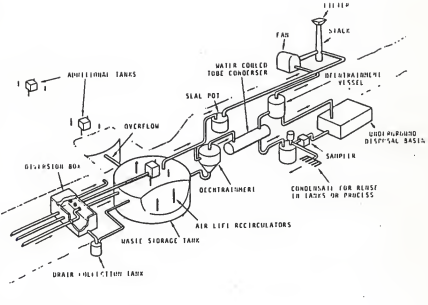
as 'tankfarms' where individual tanks are spaced 120 feet apart center-to-center, primarily because of heat transfer and safety considerations. The typical tankfarm accessories and associated cooling equipment are shown in Fig. 1.2.

Thermal Histories and Operational Experiences of Tankfarms:

The experience in the operation of these tanks has shown that a few of them (especially at the Hanford Reservation) had leaked soon after being placed into service. The ones that had failed st Hanford were carbon steel construction and single walled in design. Subsequent tanks that were constructed at Hanford, SRP, and INEL are of improved design, double walled and stress relieved. Nevertheless, the operational experience gained at Hanford had initiated efforts in detailed modelling studies to study the heat transfer aspects, evaluation of the thermal histories of the tanks that had failed in service and also the radiolytic effects of the aging waste over the extended periods of storage.

All of the high-level liquid wastes in storage in the U.S. are in the form of liquid, sludges, salt cakes and crystals in a variety of underground tanks, (with the exception of INEL where a granular calcine is stored in stainless steel bins in underground concrete vaults). After cooling for six months, a composite waste would have the average chemical composition shown in Table 1.2 and average radionuclide composition as shown in Table 1.3. Both the chemical and radionuclide composition changes as the waste ages.[2] The major changes that take place during such interim storage are:

i) Radiolytic decomposition of the waste - The major effect of the radiolytic decomposition is the slow reduction in the



SOURCE: Ref.22, 1.39, Figure II.1.42.

Fig. 1.2. Tankfarm Accessories and Associated Equipment For High-level Wastes.

Table 1.2. Average Chemical Composition of Fresh High-Level Waste.[b]

Constituent	Concentration, Molar
$NaNO_3$	3.3
$NaNO_2$	<0.2
$NaAl(OH)_4$	0.5
NaOH	1
Na_2CO_3	0.1
Na_2SO_4	0.3
$Fe(OH)_3$	0.07
MnO_2	0.02
$Hg(OH)_2$	0.002
Other solids	0.13[a]

[a]Assuming an average molecular weight of 60.

[b]Figures typical for SRP tank wastes, Ref. 2, Table 1.

Table 1.3. Average Radionuclide Composition of Fresh High-Level Waste.[a]

Radionuclide	Activity, Ci/gal.	Radionuclide	Activity, Ci/gal.
$^{144}Ce-^{144}Pr$	68	^{241}Am	1×10^{-3}
^{95}Zr	60	^{99}Tc	5×10^{-4}
^{91}Y	47	^{239}Pu	3×10^{-4}
^{89}Sr	36	^{154}Eu	1×10^{-4}
^{95}Nb	15	^{92}Zr	1×10^{-4}
^{141}Ce	12	^{240}Pu	6×10^{-5}
^{147}Pm	12	^{135}Cs	4×10^{-5}
^{103}Ru	10	$^{126}Sn-^{126}Sb$	1×10^{-5}
$^{106}Ru-^{106}Rh$	4	^{79}Se	1×10^{-5}
^{90}Sr	3	^{233}U	2×10^{-6}
^{137}Cs	3	^{129}I	1×10^{-6}
^{129}Te	2	^{238}U	6×10^{-7}
^{127}Te	2	^{107}Pd	5×10^{-7}
^{134}Ce	1	^{237}Np	4×10^{-7}
^{151}Sm	8×10^{-2}	^{152}Eu	2×10^{-7}
^{238}Pu	1×10^{-2}	^{242}Pu	6×10^{-8}
^{241}Pu	2×10^{-3}	^{158}Tb	6×10^{-8}
^{244}Cm	1×10^{-3}	^{235}U	3×10^{-8}

Note: After reprocessing fuel that has been cooled six months after discharge from reactor.

[a]Figures typical for SRP high-level wastes, Ref. 2, Table 2.

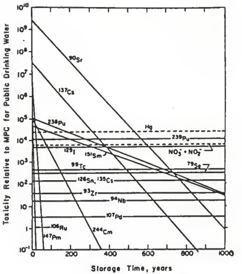
$NaNO_3$ composition with an equivalent increase in $NaNO_2$ concentration. After 5 to 10 years, the $NaNO_2$ concentration approaches the residual $NaNO_3$ concentration.

ii) A slow reduction in NaOH concentration due to reactions with CO_2 absorbed from air, forming Na_2CO_3. Then NaOH is periodically added to the wastes, to maintain its concentration.

iii) Decay of radionuclides - Fig. 1.3 shows the decay of major nuclides.

iv) Natural partitioning of the wastes into sludge and soluble fractions - The sludge scavenges most of the radionuclides from the solution as it settles to the bottom of the tank. The sludge is composed primarily of oxides and hydroxides of manganese, iron and to a lesser extent aluminum. The sludge contains essentially all of the fission products, originally in the irradiated fuel, except cesium and essentially all the actinides. The sludge also contains a significant quantity of mercury from the catalyzed nitric acid dissolution of the heavy metal fuel. The primary radioactive component of the dissolved salt solution after aging is cesium.

The salt solution is transferred to a continuous evaporator for de-watering and the concentrates from the evaporator are then transferred to a cooled water tank where suspended salt settles. Cooling causes additional salts to crystallize. The supernate from the cooled water tank is then returned to the evaporator for further concentration. The process is repeated until the portion of waste has been converted to a damp salt cake. The salt produced by evaporation of



Decay of radionuclides.

SOURCE: Ref.2, Fig.1.

Fig. 1.3. Typical Decay of Major Nuclides.

the aged supernate consists primarily of $NaNO_3$, NaOH, Na_2CO_3 and $NaAl(OH)_4$. The radionuclide concentration in the salt is approximately three times that in the original salt solution.

Model Studies for Heat-Transfer in the Liquid Waste Tanks:

The model studies undertaken for liquid waste storages were necessitated by the leakages that took place at the Hanford Reservation, where the original storage tanks were of carbon steel material. These models calculate various aspects of the heat transfer problem including the thermal histories involved in the start-up tanks containing boiling solutions.[3] As the need at that time was for production of plutonium, sufficient cooling time was not available for these initial batches of spent fuel reprocessed at Hanford and hence resulted in the hottest waste solutions. Subsequent models evaluated combined thermal and load stresses so as to determine the amount of solution that can be held in the tank safely.[4] Finally, temperatures have been calculated in tanks that had lost their liquid cover, and contours of temperature profiles around tank leaks to assess the impact of such leaks in the soil and in the immediate tank vicinity.[5] Their investigations had been detailed and elaborate which is outside our scope. Only important conclusions of their studies are reported here.

1) The heat transfer calculations were made for a 76 ft diameter tank in a tank system with 120 ft center-to-center spacing. For this system heat is lost to the atmosphere and to the water table, through a conduction layer of soil in between.
2) The normal contents inside the tank consist of a boiling supernatent that is reflexed by a condenser which held the tank

temperature essentially constant, close to the boiling point of the alkaline supernates.

3) The bottom of the tank contains a layer of sludge and salt cake of different heat generation rates, the sludge layer having generation rates usually higher than that of the salt layer and much greater than the supernates. The typical values of the heat-transfer parameters used in the calculations are given below:

Cake thickness, (ft): 4 to 24
Soil thickness (above the tank top) (ft): 7 - 11
Heat Generation Rate (Btu/hr ft^3): 0.07 - 2.83
Soil thermal conductivity (Btu/hr ft °F): 0.15 - 0.65
Vapor thermal conductivity (Btu/hr ft °F): 200
Film coefficients (Btu/hr ft^2 °F): 0.001 to 6.00

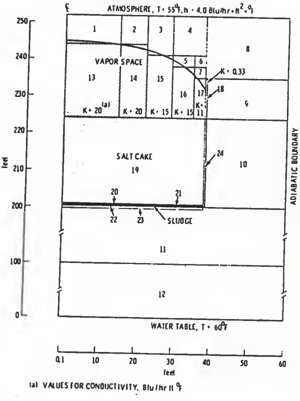
4) In all the cases, most of the conducted heat flows from the top of the tank and to the atmosphere. With the tank temperature held constant by the boiling supernate, (refluxed by a condenser) more heat is dissipated into the surrounding soil that is wet, than is dissipated into the dry soil. The temperature distribution throughout the soil tank system is nearly independent of the moisture content of the soil - as if all the soil in the system has the same thermal conductivity. The high salt content and alkalinity results in a sludge layer which contains most of the heat generating fission products such as strontium, cerium, zirconium-niobium, and ruthenium. Cesium and a small fraction of ruthenium remain in the supernate. The heat generated in alkaline waste tanks may vary over a wide range depending upon the volume reduction achieved through self concentration.

5) For this system (Fig. 1.4), about 85% of the generated heat flows to the atmosphere and the remaining to the water table at a depth of 200 ft. About 53% of the heat flowing to the atmosphere passes through the soil layer directly above the tank.

6) If the total heat generation rate from the fission producta is assumed constant, variation in the cake thicknesa has only a small effect on the temperature in the system. Because moat of the resistance to heat flow is in the soil, the thermal conductivity of the soil above the tank strongly affects the maximum temperature.

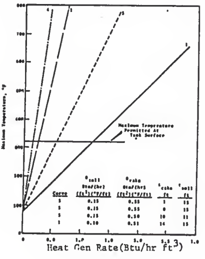
7) The cake thermal conductivity is of secondary importance, and the convective heat transfer coefficients and the tank height almoat have no effect on the temperatures. Stratification of the heat generating fission products has little effect on the maximum cake temperature.

8) Figure 1.5 shows the maximum cake temperature shown as a function of volumetric heat generation with cake thickness as a parameter. The temperature increasea linearly with volumetric heat generation rate and increases with cake thicknesa because of the total heat content of sludge inventory in the tank. Using the 320 °F at the tank surface as the cut-off limit, the quantity of the heat generating material that can be stored safely, drops very rapidly as the cake thickness increases. Figure 1.6 shows the same information as Figure 1.5, but the total heat generation is used as the parameter, which is more meaningful in such a comparison. The effect of the cake



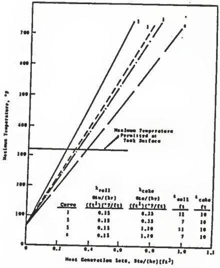
SOURCE: Ref.5, Fig.3.

Fig. 1.4. Schematic Diagram for the Vapor Space Model- Heat Transfer From High-level Waste Tanks.



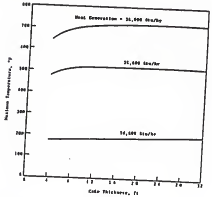
SOURCE: Ref.4, Fig.6.

Fig. 1.5. Maximum Temperature in Buried Waste Storage Tanks- Effect of Volumetric Heat Generation Rate.



SOURCE: Ref.4, Fig.8.

Fig. 1.7. Maximum Temperature in Buried Waste Storage Tanks- Effect of Soil Cover Depth.



SOURCE: Ref.4, Fig.7.

Fig. 1.6. Heat Dissipation from Buried Waste Storage Tanks- Effect of Cake Thickness.

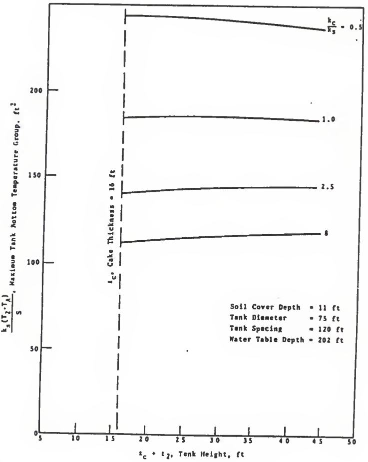
thickness on maximum temperature is shown to be small when the total heat generation rate (i.e., the quantity of fission products in the tank) is held constant.

9) The effect of soil cover depth is shown in Figure 1.7. The temperature is decreased only by 14.94% by removing 4 ft of the total 11 ft of dry soil cover on the roof, (using K_{soil} = 0.15, k_c = 0.33 (Btu/hr ft °F) and a volumetric heat generation rate = 0.4 (Btu/hr ft^3)). However, this might be just adequate to keep the tank in service, without exceeding temperature limits. In other words, this specifies the upper limit on the maximum volumetric heat generation rates of the tank contents that can be held safely. One factor which must be taken into account before removing the soil cover is the loss of the radiation shielding which may make the configuration unsafe for operations. It may be necessary to leave a minimum depth of soil from the shielding point of view.

10) Within limits, the height-to-diameter ratio of the tanks has no effect on the maximum temperature. This effect is shown in Fig. 1.8. The higher the tank, the more is the heat that is lost through the sides in the vapor space, but this is compensated by higher flux paths to the surface.

Methods for Maintaining Low Tank Temperatures:

Several steps were suggested for lowering the high temperatures that were experienced in the first set liquid waste storage tanks at Hanford. They also serve as guidelines for the safety and future design of similar storage tanks.



SOURCE: Ref.4, Fig.10.

Fig. 1.8. Maximum Temperature at the Bottom of Waste Storage Tank-- Effect of Tank Hight.

The remedial suggestions for lowering the in-service maximum temperatures are the following:

* Removal of soil from above the tank.
* Spraying dry sludge with fresh water.
* Passing cooling air through the vapor space.
* Wetting the soil above and beside the tank.

Removing soil from above the tank would reduce the temperature drastically, since temperature is directly proportional to the overlaying soil thickness. The amount of soil which could be at best removed is limited again by the loss of shielding considerations. Spraying the dry sludge with fresh water and maintaining a liquid layer would bring the surface of the cake to the boiling point of water and in practice the water would simply flash into steam. If the water did not penetrate the cake, the temperature at the bottom of the cake would be a function of the cake thermal conductivity and cake thickness. Passing cooling air through the vapor space at the practical flow rates would not be very effective when compared with spraying the vapor space with water, as above. Wetting the soil above the tank would reduce the temperature as long as the water remained in the soil. The thermal conductivity of the wet soil is about 4 or 5 times that of the dry soil and the storage temperature is inversely proportional to the soil thermal conductivity. Vaporization of the water in soil would remove heat.

CHAPTER 2

SOLIDIFICATION OF HIGH-LEVEL LIQUID WASTES

Solid forms of waste are preferred over liquid forms from a long-term waste management point of view. The end products from the in-tank solidification (ITS) discussed earlier do not qualify strictly as a 'solid waste' as defined by the 'Federal Register'.[6] United States policy is to convert high-level liquid radioactive wastes to 'dry-solid' which is 'chemically, thermally and radiolytically stable to the extent that the equilibrium pressure in the container will not exceed the safe operating pressure of the container (canister) during the period from canning through a minimum of 90 days after receipt (transfer of physical custody) at the Federal repository. The primary function of the in-tank solidification is volume reduction and production of a partially mobile salt cake, which requires a further solidification step in order for the product to meet the definition. Whereas, calcination is recognized as a solidification step and the end products from auch a step could qualify as a waste form for permanent disposal. Calcined solids are more stable than the liquids, and the volume is reduced by a factor of 10 (approximately). By solidifying the liquid waste an additional barrier against leakage and hence loss of radioactivity is gained. Solidified wastes ensure continued confinement for long periods of time (usually of the order of 100 years) when compared with similar liquid waste storage. Solidification is generally accepted aa a necessary step in the disposal of high-level wastes and is required by federal regulation.[7] Fluidized-bed calcination is but one of the several processes[8] that could be used to solidify wastes; however, it ia the only calcination process in production operation today in the U.S.

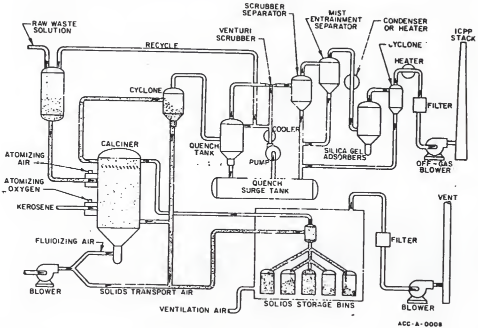
The first engineering-scale operational facility for the solidification of liquid radioactive wastes was the waste calcination facility (WCF) which began calcining wastes from the Idaho Chemical Processing Plant in 1963. The WCF product is a granular free-flowing powder which is stored on-site, in vented stainless steel bins. A batch calcine process, called the POTCAL process was developed at Oak Ridge National Laborstory (ORNL) specifically for the commercial HLWs and demonstrated at PNL (Pacific Northwest Laboratory) on a fully radioactive basis in the WSEP (Waste Solidification Engineering Prototypes). Other calcines, (such as spray calcination, rotary-kiln calcination, etc.) can also stop at a calcine, although that is not the purpose for which these were developed (i.e., for vitrification). The calcines produced by all these processes have many properties in common, differing mainly in particle size and bulk density. Some common properties of calcines which affect the process and other considerations include, thermal conductivity, volatiles content, and leachability. The first two properties are of great importance from heat-transfer and storage points of view. The thermal conductivity of the calcine is generally two to three times lower than that of consolidated products. Thus, because of high-heat generation of the high-level wastes, the storage of calcines require small diameter bins or other special heat removal features especially for fresh wastes.[9]

Description of the INEL Calcining Facility:

High-level liquid wastes are solidified by fluidized-bed calcination process at Idaho National Engineering Laborstory (INEL). Originally developed for acidic aluminium nitrate waste solutions, the process has been used also to solidify zirconium fluoride and stainless steel sulfate waste solutions.[10] High-level wastes at INEL result from complete dissolution rather than leaching of the fuel. The wastes are acidic solutions, resulting from reprocessing aluminium, zirconium, and stainless steel clad fuels. This is primarily meant to reduce precipitate formation in the tank and prevents high sodium concentrations from neutralizing the waste with NaOH. Wastes of high sodium content can not be calcined without further treatment.

Calcination Process Detail:

The process flow-sheet of a typical calcination process is shown in Fig. 2.1. Wastes are atomized into a fluidized-bed heated by in-bed combustion, and operating at a bed temperature in the range of 500-600°C. A recycle stream of gas scrubbing solution representing 20-30% of the total feed rate is added to the calciner feed stream. Inlet fluidizing velocities, based only on the fluidizing air flow through the empty cross-sectional area of the calciner vessel, at 0.18 to 0.36 m/sec are generally used and a freeboard of about 2.3 m, supplemented by a louvered baffle, is provided for de-entrainment of solids from off-gases within the calciner vessel. The bed height is maintained at a constant level above the feed spray nozzle by continuously withdrawing the bed material. Evaporation occurs on the surface of the particles and the result is a product consisting of granular bed materials and powdered calcine, both of which are removed from the calciner. The in-bed combustion system provides sufficient heat to calcine up to 400 liters/hr of high-level wastes, in a fluidized-bed maintained at a constant bed temperature of 500°C. A schematic diagram of the calciner vessel showing in-bed combustion is



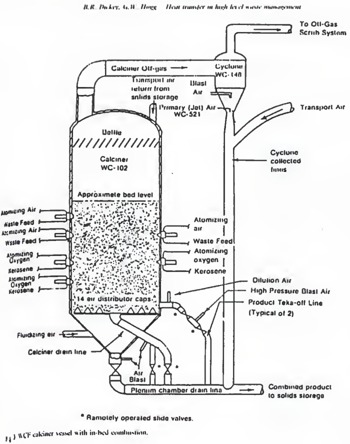
SOURCE: Ref.16, Fig.1.

Fig. 2.1. Schematic Flow Sheet of the Waste Calcining Facility, INEL.

given in Fig. 2.2. During operation, kerosine is sprayed through external atomizing nozzles into a hot fluidized bed where the fuel burns, releasing heat and producing CO_2 and water vapor and a small quantity of CO. The product is a mixture of granular bed materials and powdery solids from the off-gas system in primary cycle; the product is removed from the bed pneumatically.

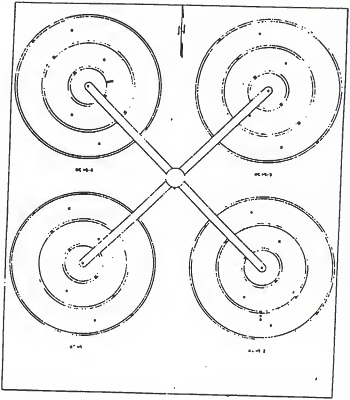
INEL Calcine Storages:

Early defense waste calcined in WCF consisted essentially of a mixture of aluminium nitrate and fission products. The first set of bins was designed to maintain the temperature of the calcined solids below 400°C (752°F), the sintering temperature, to prevent possible evolution of volatile fission products.[11] A plan of the first set of bins at INEL, Idaho is shown in Fig. 2.3. An isometric drawing of the first and second set of annular bins is shown in Fig. 2.4. The first set of bins were designed rather conservatively, taking into account the fresh nature of the wastes, assuming a volumetric heat generation rate of 1200 W/m^3 (142.75 Btu/hr ft^3). Calcined solids were transferred pneumatically from the calciner to a cyclone located over the top of the bins; calcine falls by gravity to each of the concentric bins. The bins are located inside an underground concrete vault. Total capacity of the first bin set, which was filled in 1964, was approximately 200 m^3 (7400 ft^3). The second and third calcined solids storage facility consisted of 3.7 meter diameter cylindrical bins, also enclosed in reinforced concrete vaults. The second and third sets of calcined storage differ primarily in the height of the bins, capacity being 900 m^3. The fourth set of bins is similar to the second or third set but the capacity is only 500 m^3. But beginning with the fifth set, and future sets of bins



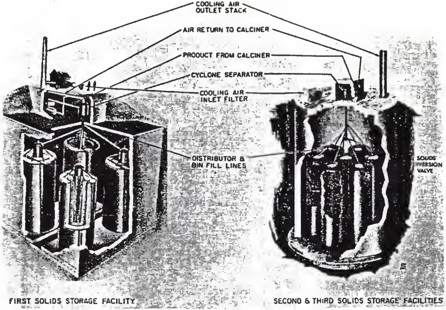
SOURCE: Ref:11, Fig.1.

Fig. 2.2 Schematic of the In-Bed Calciner Vessel.



SOURCE: Ref.14, Fig. VI-14.

Fig. 2.3. First Set of Calcine Storage Bins- Plan View.



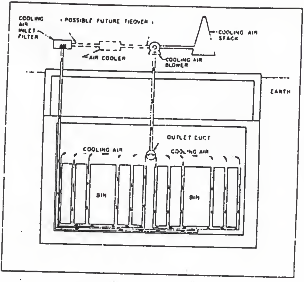
SOURCE: Ref:10, Fig.3.

Fig. 2.4 Cut-a-Way Views of the High-level Solids Storage Facilities.

will be similar in design to the first set, i.e., annular configuration with natural circulation/forced circulation cooling because of projected increase in heat-generation rates.

Geometry of the Calcine Storages:

The calcines storage facility at INEL consists of vertical stainless steel bins within an underground concrete vault. Connections are available to permit ultimate disposal or further treatment if that is desired. Three solids storage facilities were placed in operation prior to 1976 and the fourth was designed and constructed in 1976. The first set consisted of four bins, each set containing a central bin 0.915 m in diameter by 7.3 m high, and two progressively larger concentric bins, each 0.61 m thick by 6.1 m high with an annular space between bins for convective cooling.[12] These four sets of bins have a total capacity of 220 m^3 (770 ft^3) and are surrounded by a rectangular reinforced concrete vault. The bins are cooled by ambient air, which flows through the pre-filters down an inlet duct, to the bottom of the vault. Air then flows upward through the vault by natural convection and out of the vault through a 15 m (50 ft) tall cooling stack (Fig. 2.5). A forced circulation system was installed in the first storage facility but has not been needed. The cooling air can be shut off and high efficiency particulate filters can be installed should radioactivity be detected. The second set of bins consists of seven 3.66 m (12 ft) cylindrical bins, 12.8 m high (with a central bin and six on the periphery), again housed in a reinforced concrete outer vault. The capacity of the second set amounted to 900 m^3 (31800 ft^3) of solids. The bins are cooled by natural convection and designed for a maximum surface temperature of 290°C (554°F).



SOURCE: Ref.14, Fig. IX-2.

Fig. 2.5. Cooling Air Closed Loop System for Solids Cooling.

Calcine Cooling:

The first set of bins was designed for cooling by forced air circulation however, the decay heat was removed using only natural convection by circulation of cooling air in the annular spaces between the bins. Additional heat was removed by thermal radiative transfer between the bin wall and concrete vault. The first set was designed on the basis of a calcine volumetric heat generation rate of 1200 W/m^3 (142.75 Btu/hr ft^3). Calcine placed in bin set-1 had an average heat generation rate of only 300 W/m^3 (35.68 Btu/hr ft^3). The highest measured calcine center line temperature in bin set-1 was 185°C. Bin set-2 contains both alumina and zirconia calcine with an average heat generation rate of 250 W/m^3 (29.74 Btu/hr ft^3). Cooling air was shut off for a period of twelve months beginning in 1969. Heat removal was then primarily by convection to the vault air and bin walls to vault walls. Heat was eventually dissipated by conduction to the surrounding soil and to the water table. The maximum measured calcine temperature during this test in 1969 was 695°C (1283°F).[13]

The bin set-3 contains calcine with an average heat generation rate of 50 W/m^3 (5.95 Btu/hr ft^3) and the maximum calcine temperature measured has been only 300°C (572°F). The expected heat generation rate of the calcines in the fourth and fifth set were to be 400-500 W/m^3. Maximum calcine temperature in these sets were to be limited to 650°C to prevent sintering of calcines from sodium-zirconium feed. The properties of the calcines significant to storage consideration are given in Table 2.1 and the measured bin/wall temperatures are given in Table 2.2.

Table 2.1. Properties of Calcines in Storage at INEL.

Thermal conductivity (W/m-°C)		0.19 - 0.35
Storage bulk density (g/cc)		Zr 1.55 - 1.7
Sintering Temperatures (°C):		
zirconium fluoride	700	
alumina calcine	1200	
Zr-sodium calcine	650	

Table 2.2. Storage Experience with Calcine Solids at INEL

	Calcine Solids Bin Set 1	2	3
Calcine Heat Generation (W/m^3)	300	250	50
Measured Maximum Temperature (°C)	185	695	300
Minimum Measured Temperature (°C)	-	85	55

Note:

1) Core sample from 2 bins in set-2 taken in 1978 revealed no sintering.

2) Cooling air in bin set-2 was shut off for 12 months beginning in 1969. Maximum measured calcine temperature was 695°C and bin wall temperature was 290°C.

Design of the Existing Solids Storage Bins:

The existing bins, especially the first set of bins at INEL, have been designed taking into account the high rates of heat generation of the first cycle wastes reprocessed and the lack of initial aging of these wastes. Hence, the design was conservative. The design was also due to conservative data on fission product volatility of the calcines stored at high temperatures and also the sintering temperature ranges of the varieties of calcines involved. The following were the design basis assumptions with which the first set of bins were originally constructed.[14]

i) The bins were designed to maintain all of the stored material below the calcination temperature, i.e., 400°C (750°F) and to prevent, if possible, the evolution of radioactive fission products. At temperatures above 400°C unconverted aluminum nitrate and mercuric oxide in the product would decompose, with the evolution of oxides of nitrogen, oxygen, and mercury. These gases might entrain active dust in passing through the stored solids. At still higher temperatures, radioactive fission products may volatilize.

ii) Storage of the WCF (Waste Calcining Facility) solid products is complicated by the poor heat transfer characteristics of the granular calcine formed by the fluidized-bed calcination process.

iii) Attenuation of the gamma and beta radiation from decay of the contained fission products generates heat within the stored particles of alumina.

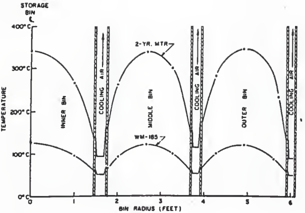
iv) In the narrow bin type of storage developed, excessive solids temperature are avoided by limiting the distance between cooling surfaces to 24 inches with the exception of the central cylinder which has a diameter of 3 ft.

v) The maximum temperature will occur in the center of the bin and depends upon the heat generation rate, the thermal conductivity of alumina, the bin thickness, and the heat removal rate from the bin walls. Each of the four WCF bins consists of 36 inch diameter standpipe nested in two concentric 24 inch wide bins. Heat is removed from the bin surface by air flowing in the cooling air channels between the bins. Either natural circulation or forced circulation can be used to move the cooling air. A 9000 scfm blower is installed to provide the forced circulation. It was anticipated that the forced circulation may be necessary only to hold the center line temperature below 400°C in the case of wastes less than three years out of the reactor. Natural circulation will provide sufficient cooling for older wastes.

Calculated temperature profiles with forced circulation are shown in Fig. 2.6 for both two year cooled solids (MTR Type fuel) and solids from WM-185 tank (aged defense wastes). Figure 2.7 shows similar temperature profiles for natural convective cooling for both the wastes.

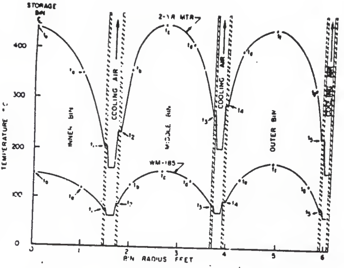
<u>Activity and Heat Generation by Fission Products</u>:

Except for ruthenium, which is volatile in the calciner, the fission products will appear in the calcine products at the same ratio of aluminium to the fission products that existed in the initial wastes



SOURCE: Ref.14, Fig. V-1.

Fig. 2.6. Temperature Profile in the First Set of Calcine Bins- Calculated Distribution for Forced Circulation Cooling.



SOURCE: Ref.14, Fig. V-4.

Fig. 2.7. Temperature Profile In the First Set of Calcine Bins- Calculated Distribution for Natural Circulation Cooling.

processed. Using this basis and an estimated aluminium content in the solids to be 91% allows the fission product activities of the tank-185 wastes and the theoretical fresh wastes to be converted to curies per lb of solids. It was assumed that 100% of ruthenium would appear in the product. The actual activities of the solids would be lower than the calculated activities, the extent depending upon the degree of ruthenium volatilization.

The rate of heat generation by decay of fission products in the solids is an important factor determining the design of the solid storage bins. The heat generation rates for the solids have been calculated using fission product decay energies taken from the literature and assuming that all the radiation is dissipated within the solids. However, heat generation rates calculated on this basis will be somewhat higher than the actual. The error decreases as the bulk of the solids increases. The assumption that 100% of ruthenium appears in the solids would also contribute to the difference between calculated rates and actual rates. The heat generation rates from the first cycle waste solutions are shown in Fig. 2.8. The same data applied to the theoretical MTR type of wastes are shown in Fig. 2.9. The procedure involved in estimation of the heat generation rates is the conversion of the waste activity to an activity per lb of solids and conversion of the same to heat generation per lb of solids using decay energies available from the literature. The heat generation rates then can be converted to volumetric heat generation rates using the average bulk density (59.3 lb/ft^3) of the formed calcines.[16]

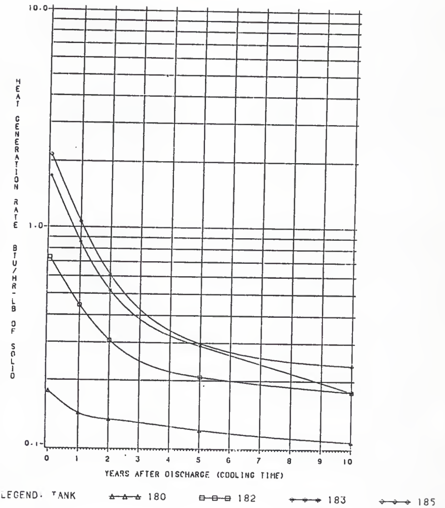


Fig. 2.8 Heat Generation Rate of Calcined Solids in Tank - 180,182,183 & 185.

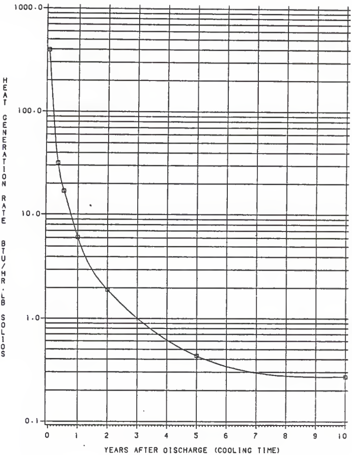


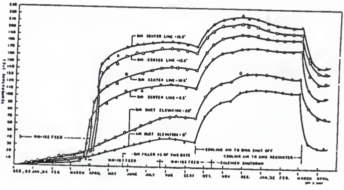
Fig. 2.9 Heat Generation Rate of Calcined Solids- Theoretical MTR Wastes.

Performance Evaluation of the Bin Set-1:

The first processing campaign of converting the defense waste solutions into solid calcines at INEL was carried through during 1963-64 and calcines were actually placed in the first set of bins beginning in 1964. The field test and the actual temperature profiles measured during this storage at the end of the first processing campaign gave the feedback data and performance testing of the first set of bins (bin designated WC-115-2). The outcome of the field test and the temperature data indicated overdesign and conservative estimates with regard to maximum temperatures. The following were the gist of the field test results.[15]

i) The heat generation rates of the wastes calcined during the run ranged from 0.28 to 0.85 Btu/hr per pound of solids in storage (16.8 to 51.0 Btu/hr ft^3). At the end of the campaign the total heat generation rate was 222,800 Btu/hr, of which 65% was due to calcined solids from WM-187 feed solution. The temperature at the center line of various chambers (vertical mid points of bins) were dependent upon the heat generation rates of solids in the specific chamber and were also the highest.

ii) Temperature profiles at various elevations in the middle chamber (central bin, 0.91 m dia) of bin WC-115-2 are shown in Fig. 2.10. The profiles are typical of all the chambers, except for the magnitudes of temperatures and the physical time periods at which respective elevations were filled-up. The bottom half of the chamber contained solids from WM-185 (five year aged wastes) operations and the top half from WM-187 (three year aged wastes) operations.



SOURCE: Ref.14, Fig.10.

Fig. 2.10. Center Line Temperature of Solids in the Middle Chamber of WC-115-2, INEL.

iii) The temperature during Nov., 1964 (about 225°C, at 15.5 ft elevation) was the highest observed during and following the campaign.

Removal of Heat from the Bins and Interpretation of Results:

The solid storage bins were designed to accommodate solids with a heat liberation rate of 2.5 Btu/hr. lb (equivalent to a volumetric heat generation rate of 142.75 Btu/hr ft^3). Forced convection by means of a cooling air blower was considered necessary to prevent the solids temperature from exceeding 400°C, which was in turn considered as the design maximum upper limit from volatility and sintering considerations. Because the actual heat generated by the calcines averaged only 0.52 Btu/hr. lb (29.692 Btu/hr ft^3) the operation of the blower (forced circulation) was considered unnecessary. The bin, in effect, was cooled by natural convection of fresh air, as the air rose past the bin surfaces, with the bin vented through a 50 ft stack. Even with this mode of cooling the actual temperatures measured in various chambers of the bins were about 185°C (365°F). By September, 1964, it became apparent that a maximum temperature had been reached and the declining temperatures were following the decrease in ambient temperature resulting from the seasonal changes.

In mid-September, 1964, air flow rates and temperature differentials through the cooling air system were measured for estimating the extent of convective heat transfer taking place with the natural circulation mode. From those data, conservative estimates indicated that only 34% of the heat generated was being removed by the cooling air stream and the rest of the heat was apparently being rejected by radiant and conductive means to the soil. A second field test was undertaken by shutting off the cooling air valves to obtain equilibrium temperatures with no inflow of cooling air. The maximum temperature approached 225°C after two months (which was also the maximum temperature measured during these tests) and started to decline with the natural decay of radionuclides. After an additional 3 months (March, 1965), the cooling air inlet valves were reopened and temperatures reached a new steady state at 145 - 150°C (300°F). Outside concrete wall temperatures at the soil - concrete interface midway up the wall surface reached a pseudoequilibrium value of 45°C (113°F). Though a rigorous mathematical analysis was not undertaken, it was apparent from the field test that considerable heat was being transferred (perhaps by a combination of radiative, and convective means) and eventually by conduction to the soil surrounding the vaults.

Mathematical Modelling Studies for Simplified Geometries:

Following the field test a mathematical modelling study was undertaken by D. E. Black and B. R. Dickey, Idaho Nuclear Corporation,[1] to analyze the heat transfer, particularly heat conduction through the soil, and to pursue the inherent mode of heat rejection through conduction. The merits of this system over the existing arrangement are:

i) Takes advantage of the inherent mode of cooling through heat conduction which is direct and failure proof.

ii) Avoidance of equipment for air circulation and the related air cleaning accessories.

iii) The ruthenium volatilities in the exhaust was a serious problem with fluidized bed calcination process.[16] Should a bin leak and spill radioactive solids into the cooling air stream, the

> sctivity relesse would be dangerous and is, in fsct, one of the design basis accidents that is considered in the safety snalysis.[14] In the existing arrangement, though the calcine feeder pipes and the distributor cyclone are isolated from the primary system from the cslciner vessel and the bins provided with its own ventilation stsck, in sn accident scenario the bins are required to be connected to the plant ventilation, while shutting off the bin cooling. But accidental rupture of feeder pipes and/or failure of the bins, especially due to attrition are all possible occurrences. The problem gets amplified in a bin configuration which is not only large in size but geometrically complex. By totally burying the calcine bins ss individusl units and by making the integrity of such simpler geometries tight, an additionsl protection against leakage can be obtained without the need for sny external cooling.

However, the question of efficient heat removal and the ability of such simplified waste storage in maintaining allowable maximum temperature over long periods of storage remain to be satisfactorily answered. This requires analyticsl solutions and mathematical modelling studies. The model study undertaken by D. E. Black and B. R. Dickey[1] was probsbly the first. To develop a model which would exactly suit the existing geometrically complex bin arrangement was rather difficult. The complexity wss that there was a large number of bins enclosed within a concrete vault surrounded by a convective layer of air surrounded by soil outside. The model must be sble to arrive at analyticsl solutions to vslidate the temperature profiles measured in the field. However, the qualitative assessment of about 65% of heat conducted through the bin walla to the soil does point out the validity of the assumptions in the model, i.e., a viable mathematical modelling can be developed to devise a new syatem of buried aolid cylinders directly in the soil at varying deptha and to simulate the field conditions reaaonably cloaely.

Cesium Migration:

The design of the firat aet of bina at INEL had been baaed upon the maximum temperature limita imposed by the fission product volatility, particularly cesium, which accounts for an appreciable amount of generated heat in the atored calcines. Migration of the same at the storage temperaturea was one of the reasona for the lower design limit on the allowable bin center line temperatures. But aubaequent studies undertaken[17] did reveal that higher temperatures can be tolerated, up to 800°C (1472°F), without any migration of theae fiasion products in auch atorage configuration. The results of these studies indicate that cesium-137 in a storage bin containing radioactive granular solids will migrate only at temperatures above 700°C (1292°F). As the temperature rises above 700°C cesium will move towards the outer and cooler parts of the bin, staying mainly in the 625-750°C temperature band. Movement of cesium in thia manner will result in an actual center line temperature lower than that calculated for a uniformly diatributed aource. In the experiments undertaken for the study of cesium migration using gamma ray spectrometry, migration began alowly below 900°C (1652°F), but waa rapid at 1200°C.

Stability of Alumina Calcine During High Temperature Storage:

Another important reaaon for limiting the maximum temperature was the product was sintered at high temperatures and doubts about the stability of the calcines at such temperatures. But experimental studies undertaken subsequently indicated that zirconia calcine can be maintained in the free flowing granular form for temperatures up to 700°C.[18] In this form it can be removed readily for transport or additional treatment, if that is required. Conversely, it can be converted to a sintered mass without additional chemicals, simply by heating the calcines to 800°C or higher. This may be a more desirable form, if the solids are to be transported to another location. The time required to form the sintered mass decreases, as the temperature increases but volatility of the fluorides becomes significant at temperatures above 1000°C. Cesium volatility also becomes a significant factor above 800°C. The primary advantage of turning calcines from the granular to sintered mass may be its greater physical integrity, but for the calcines in the interim bin type of storages, it may be actually a problem for its eventual retrieval for vitrification. From those angles, safe practice would be to retain its free flowing form and an upper limit of 700°C seems appropriate from sintering considerations.

Modelling Studies at Idaho Nuclear Corporation:

The current trend in nuclear burial facilities is toward design of simplified storage facilities which can be approximately described as solid cylinders buried at varying depths below the surface of soil. The necessity for an outer containment has been progressively omitted in disposal of nuclear wastes except for such interim storages in an aqueous form. With these wastes the doubts about leakages of wastes to environment override other considerations. The current design concept for storage of spent fuel elements (RSSF) follows the simplified design, in spite of the fact that an appreciable amount of heat output is one of the major problems with the dry-type of storage being considered for burial in national facilities.

The model developed[1] by D. E. Black and B. R. Dickey for the calcine storages can be approximately described as cylinders buried at varying depths below the surface of the soil. A schematic representation of these storage facilities is shown in Figs. 2.11 and 2.12. The ends of the heat sources were assumed to be perfectly insulated in this study and the heat losses from the ends were neglected without affecting significantly the accuracy of the calculated temperatures. Normally these losses are negligible for small diameter containers. Since there is a discontinuity in the medium at the ends of larger diameter heat sources, boundary conditions should be specified. However, the complexity of mathematics is reduced if a continuous medium is used and accuracy is not significantly affected. The thermal conductivity of the source and the surrounding media significantly affect the maximum temperature in the waste and the temperature distribution in the soil surrounding the container. A homogeneous soil with constant physical properties (with the exception of thermal conductivity dependence on temperature) was assumed in the model.

Mathematical Formulations:

Although the interior of the present containment structure is geometrically complex, a homogeneous heat source was assumed for the model. Since the thermal conductivities in the region $0 < r < R_b$, where R_b is the radius of the bin, are independent of the variable 'Z', the specification of boundary conditions at the top and bottom of the bin

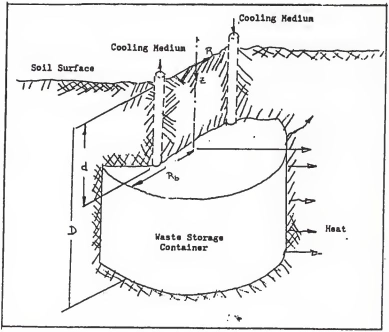


Fig. 2.11 Typical Representation of the Directly Buried Waste Storage Bin for the Model Studies.

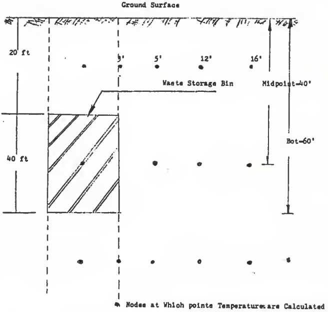


Fig. 2.12 Vertical Section Through the Waste Storage Bin Showing the Nodes.

where a discontinuity normally exists is eliminated. In a steady state mode of heat transfer, and with the assumption that soil surrounding the container is a semi-infinite medium, the boundary value problem for the temperature distribution in the bin and the surrounding medium is given by the Fourier conduction equation:

In the region bounded by the heat source (i.e., in the bin)

$$\frac{\partial^2 T_1}{\partial r^2} + \frac{1}{r}\frac{\partial T_1}{\partial r} + \frac{\partial^2 T_1}{\partial z^2} + \frac{q'''(r,z)}{k_c} = 0, \text{ for } (0 \leq r \leq r_b \text{ and } d < z < D). \quad (2.1)$$

In the region in the soil surrounding the bin,

$$\frac{\partial^2 T_2}{\partial r^2} + \frac{1}{r}\frac{\partial T_2}{\partial r} + \frac{\partial^2 T_2}{\partial z^2} = 0, \text{ for } (r \geq r_b \text{ and any } z). \quad (2.2)$$

The boundary conditions for the model are,

$$T_1 (r,0) = T_2 (r,0) = 0, \quad (2.3)$$

$$\lim_{z\to\infty} T_1 (r,z) = T_2 (r,z) = 0, \quad (2.4)$$

$$\lim_{r\to\infty} T_2 (r,z) = 0, \quad (2.5)$$

$$T_1 (r_b,z) = T_2 (r_b,z), \text{ for } d \leq z \leq D, \quad (2.6)$$

$$k_c \frac{\partial T_1}{\partial r} (r_b,z) = k_s \frac{\partial T_2}{\partial r} (r_b,z), \text{ for } d \leq z \leq D, \quad (2.7)$$

$$T_1 (0,z) = \text{Finite}. \quad (2.8)$$

Continuity of the temperature and heat flux between the regions are given by Eqs. (2.6 and 2.7).

A finite temperature at $r = 0$ is expressed by Eq. (2.8). The remaining boundary conditions state the specific temperatures at the physical boundaries i.e., Eqs. (2.3, 2.4 and 2.5).

The steady state solutions for the temperature distribution in the waste storage bin and the surrounding soil, using Fourier transformation, solving the resultant formulation, evaluation of the arbitrary constants, including the inversion of the transform solutions, (for details of the analytical solution, see Appendix E of Ref. 1) results in the following final solution for the temperatures T_1 (the temperatures inside the heat source, i.e., the calcine bin) and T_2 (the temperatures in the soil):

$$T_1(r,z) = \frac{2L^2q'''}{\pi^3} \sum_{n=1}^{\infty} \frac{[\cos\frac{n\pi d}{L} - \cos\frac{n\pi D}{L}]\, U(X_B,X,k_c,k_s)\, \sin(\frac{n\pi z}{L})}{n^3\,[k_c K_o(X_B) I_1(X_B) + k_s K_1(X_B) I_o(X_B)]}, \quad (2.9)$$

$$T_2(r,z) = \frac{2L^2q'''}{\pi^3} \sum_{n=1}^{\infty} \frac{[\cos\frac{n\pi d}{L} - \cos\frac{n\pi D}{L}][I_1(X_B) K_o(X_B)]\, \sin(\frac{n\pi z}{L})}{n^3[k_c K_o(X_B) I_1(X_B) + k_s K_1(X_B) I_o(X_B)]}, \quad (2.10)$$

where q''' = The heat generation rate, (Btu/hr ft³),

k_c, k_s = The thermal conductivities of the solid waste and soil respectively, (Btu/hr ft °F),

$I_o(X), I_1(X)$ = Modified Bessel functions of the first kind, of zero and first order respectively, evaluated for the argument (X),

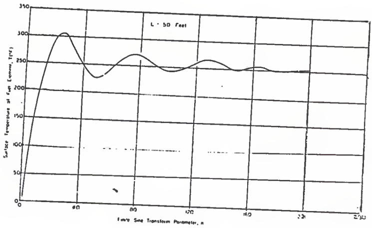
$U(X_B,X,k_c,k_s) = k_c K_o(X_B) I_1(X_B) + k_s\, K_1(X_B) \times [I_o(X_B) - I_o(X)]$

$X = \frac{rn\pi}{L}$,

L = Finite depth of the system, (ft).

The partial differential equation and the boundary conditions commonly termed a BVP (boundary value problem) were solved by transform methods. The boundary conditions of the problem dictate the particular type of transformation which should be employed. Use of an infinite Fourier sine and cosine transform (implying a semi infinite medium in the Z-direction) to eliminate the independent variable 'Z' requires numerical evaluation of a complicated inversion integral containing modified Bessel and trigonometric functions. If the range of the independent variable, 'Z' is limited to $0 < Z < L$, where L is a finite length, evaluation of a complicated inversion integral is not required. Since the temperature at the boundaries in the Z direction are defined, the finite Fourier sine transform is used in the model.[21] The finite length 'L' is the distance at which a further increase in L results in negligible change in the calculated temperatures. The final value of L is normally much greater than the burial depth of the source.

The calculational procedure for deriving the solutions to the equations require that the initial value of L be incremented until the changes in calculated temperature is less than a specified quantity. An arbitrarily large initial value of L can not be used since the number of 'n' required (Bessel routes in the solutions for T1 and T2) increase with increasing L. The final value of L used in a calculation is primarily dependent on the strength of the heat source, the thermal conductivity of the surrounding medium, and specified accuracy of the temperature. The convergence of temperature ss a function of the transform parameter 'n', is analogous to that of a typical Fourier series expansion as shown in Fig. 2.13. For a proper value of L (large enough that the temperature is no longer dependent on L), the temperature oscillstions are small, and the required value of n (≈ 100 for the case shown in Fig. 2.13) is dependent only upon the desired accuracy of the calculated temperature. For values of L which are too small, the oscillations are much more pronounced.



SOURCE: Ref.1, Fig.6.

Fig. 2.13. Convergence of Steady State Temperature in the Storage Bin as a Function of the Parameter 'n'.

Modification of the Solid Cylinder Geometry by Annular Configuration:

The mathematical model for the temperature profiles in the previous study[1] has been for very simple geometry, namely, a solid cylinder configuration, and determination of the near field effects of the heat source buried in soil. Also, it was the closest geometry suitable for field verification of the model calculations, which were subsequently validated using electrical heating elements buried at various soil depths at INEL site.[1] Though the concept of using annular and finned configurations for these high-heat output wastes is not a new subject, all the analyses done in this direction pertain to interim storage and transport considerations for the high-level waste canisters. The analysis was performed for the reference canisters (which are typically 12 inches or less in diameter and 8 ft - 0 long) at Pacific Northwest Laboratories.[20] Practically no modelling studies were undertaken in the analysis of large waste storage bins such as the ones at INEL for annular configuration. But, the heat transfer is basic and the general principles underlying apply equally well for the annular configuration proposed in this study. An annular waste storage container with an increased outer diameter can hold higher allowable internal heat generation of stored material because of increased surface area for heat dissipation. The annular configuration can store larger volumes of waste per unit height than solid cylinders without exceeding the maximum temperature and therefore could result in a smaller number of such storage facilities required.

But in the comparative analysis of heat-transfer from radioactive containers, there are two additive temperatures to consider. The first is the temperature rise from the surrounding medium to the surface of the container. This is highly dependent on the environment in which the container is stored and is often sufficient if the surface tempersture is the controlling design factor. If the design limitation is at the container center line, the internal temperature rise from the surface to the center line must be added to the external rise to define the design condition. For direct burial in soil the internal rise is secondary; for storage in s water basin it is controlling. This is an important factor in considering the annular geometry that is pursued in this study, because a problem with the annular configuration is thst, the inner annular wall results in the maximum cslcine temperature with a free standing column of sir, that is considered in this model. This of course will not be a problem with the waste directly buried in soil; but will be a problem from transport considerations (such as the calcine transport csnister) which may require convective cooling with a liquid medium.

CHAPTER 3

MATHEMATICAL MODEL FOR THE ANNULAR GEOMETRY

The waste storage configuration pursued in this research work is an improvement over the previous configuration for the simpler geometries of buried heat sources, which assumes a solid geometry surrounded by soil medium. In the current configuration an annular geometry was assumed in which the innermost cylinder holds a column of air. The column of air is surrounded by an annulus which contains the heat source (calcined waste). In turn the heat source annulus is surrounded by soil. A schematic of the annular bin model is shown in Fig. 3.1. The mathematical formulations are derived for such an annular configuration for the three regions, using the Fourier heat conduction equation and using boundary conditions for the regions. The analytical solution gives the temperatures in the three regions, namely, air in the innermost cylinder, calcined waste in the annulus, and soil outside. The results for solid geometry are also calculated by collapsing the innermost radius (region bounded by air) to a very small radius (0.001 inch) which thus reduces approximately to that of a solid geometry. The analytical results arrived at in the earlier study (1) compare favorably (within 50°F, for a maximum temperature of 1694°F) to those obtained for the "collapsed" annular geometry.

Model Assumptions:

The assumptions used in this model for the mathematical formulations of the conduction equations and the boundary conditions are identical to those of the previous study.[1] The principal difference is in the geometry, the bin configuration, and the equation development and the approach to the analytical solutions. The finite Fourier

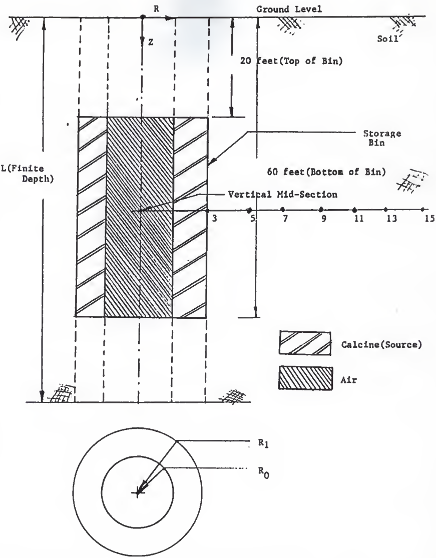


Fig. 3.1. Schematic Diagram for the Annular Geometry of the Bin.

transform, as was used in the previous study was not used in this model in arriving at the analytical solutions. The formulations are arrived at for the three conduction regions by the use of superposition for the inhomogeneous source term involved in the region bounded by the heat source. The partial differential equations are solved using the separable variable technique. The assumptions that are made in development of the formulation and in arriving at the analytical solutions are described below:

1) The temperature calculations are for the steady state mode of heat conduction. No transient solutions are considered in this study.

2) The heat source is a cylindrical annulus with a volumetric heat generation rate of q''' (Btu/hr ft^3) and of length L in the Z direction (depth of the container). This eliminates the need for specification of the boundary conditions at the top and bottom of the bin where a discontinuity normally exists.

3) The rate of heat generation within the homogeneous source is independent of time and the soil surrounding the container is considered as a semi-infinite medium. The heat transfer is ideally represented by the Fourier heat conduction equation and is solved using a procedure common to solution of boundary value problems.

4) For the purpose of this model, the thermal conductivity was assumed independent of temperature. However, for the air in the inner region appropriate values of k_{air} were used in the calculations (i.e., k_{air} at 293 °K = 0.014 (Btu/hr ft°F) and k_{air} at 1400 °K = 0.0414 (Btu/hr ft °F)).

5) The internal complex geometries and the layout of bins in the existing arrangements were reduced to a simplified geometry for the purpose of the model, as was done in the previous study. The bins are assumed to be individual heat sources buried in soil at varying depths.

6) The problem is two dimensional, i.e., in the radial and axial (r and z) directions. The variations in the azimuthal direction was not considered.

7) The effective increase in the source radius due to gamma ray heating outside the physical dimension of the storage container was neglected. The inaccuracy introduced by this assumption in the estimates of temperatures is negligible, especially for large diameter containers. For a container one relaxation length in diameter, or 4 inches for common wastes, approximately 10 percent of the total gamma radiation escapes to the surrounding medium. For a container ten relaxation lengths in diameter (or 40 inches), less than 2 percent of gamma radiation escapes. Since gamma radiation accounts for less than one-half of total heat generation, only one percent of heat dissipation escapes to soil.

8) Heat loss from the ends of the heat source were neglected. Such losses from containers with large length-to-diameter ratios are negligible; as length-to-diameter ratio decreases, i.e., for a large diameter container, heat losses become significant. Thus, the heat flow is one dimensional, i.e., in the radial direction.

The mathematical formulations are made for the three regions and details of the developed equations and the analytical solutions are described in the following section.

For the central region, i.e., $0 \leq r \leq r_o$, the following model was used. The temperature in this region was designated T_1. The Fourier heat conduction equation for this region with no heat source is

$$\frac{\partial^2 T_1}{\partial r^2} + \frac{1}{r}\frac{\partial T_1}{\partial r} + \frac{\partial^2 T_1}{\partial z^2} = 0. \tag{3.1}$$

The boundary conditions used, which are the same as those used by Black and Dickey (1) are

$$T_1(r,0) = 0, \tag{3.2}$$

$$T_1(r,L) = 0, \tag{3.3}$$

$$T_1(0,z) \text{ is finite}, \tag{3.4}$$

$$T_1(r_o,z) = T_2(r_o,z), \tag{3.5}$$

$$k_a \frac{\partial T_1(r,z)}{\partial r} = k_c \frac{\partial T_c(r,z)}{\partial r}. \tag{3.6}$$

The first two boundary conditions describe a perfect insulation condition generated by the soil covering at both top and bottom of the waste. This is somewhat of an unrealistic condition but matches the conditions of Black and Dickey (1).

A separation of variables technique is applied to Eq. (3.1) by assuming a solution of the form

$$T_1(r,z) = R(r)Z(z).$$

Substitution for $T_1(r,z)$ in Eq. (3.1), followed by division by $R(r)Z(z)$ and a slight rearrangement yields

$$\frac{1}{R}\frac{d^2R}{dr^2} + \frac{1}{Rr}\frac{dR}{dr} = -\frac{1}{Z}\frac{d^2Z}{dz^2}.$$

Since both sides of this equation are functions of only a single variable, both sides must be equal to a constant (λ^2). Thus, two equations result. The first one is,

$$r^2\frac{d^2R}{dr^2} + r\frac{dR}{dr} - \lambda^2 r^2 R = 0.$$

This is a modified Bessel's equation which has a general solution of,

$$R(r) = CI_o(\lambda r) + DK_o(\lambda r), \qquad (3.7)$$

where $I_o(\lambda r)$, $K_o(\lambda r)$ are modified Bessel functions of the first and second kind and zero order, respectively.

Since $T_1(0,z)$ is finite, D must be zero because $K_o(\lambda r)$ goes to infinity at $r = 0$.

The second equation from the separable variable technique is

$$\frac{d^2Z}{dz^2} + \lambda^2 Z = 0.$$

The general solution for $Z(z)$ is

$$Z(z) = A \sin(\lambda z) + B \cos(\lambda z). \qquad (3.8)$$

Since $T_1(r,0) = 0$, $B = 0$ (since cos (0) is unity). Also since $T_1(r,L) = 0$,

$$A \sin(\lambda L) = 0,$$

and if $A \neq 0$, $\sin(\lambda L) = 0$ at $\lambda L = n\pi$, $n = 1,2,\ldots$. Thus,

$$\lambda L = n\pi,$$

or

$$\lambda = \frac{n\pi}{L} .$$

Finally, the overall solution is

$$T_1(r,z) = \sum_{n=1}^{\infty} A_n I_o(\lambda r) \sin\left(\frac{n\pi}{L} z\right) , \qquad (3.9)$$

where $A_n = AC$ for each value of λ.

For the region in the soil, i.e., $r \geq r_1$ the following model formulation was used. The temperature in this region was designated T_3. The Fourier heat conduction equation for this region with no heat source is

$$\frac{\partial^2 T_3}{\partial r^2} + \frac{1}{r}\frac{\partial T_3}{\partial r} + \frac{\partial^2 T_3}{\partial z^2} = 0. \qquad (3.10)$$

The boundary conditions used are:

$$T_3\ (r,0) = 0, \qquad (3.11)$$

$$T_3\ (r,L) = 0, \qquad (3.12)$$

$$T_3\ (\infty,z) = \text{finite or } 0, \qquad (3.13)$$

$$k_s \frac{\partial T_3}{\partial r} (r_1,z) = k_c \frac{\partial T_2}{\partial r} (r_1,z), \qquad (3.14)$$

$$T_3\ (r_1,z) = T_2\ (r_1,z). \qquad (3.15)$$

The last of the two boundary conditions express continuity of flux and temperature between the two adjacent regions, i.e., the heat source and soil (the temperature inside the heat source region is designated as T_2 and k_c and k_s represent the thermal conductivities of the calcine and soil respectively).

Substitution of separated variables in equation (3.10) results in the following ordinary differential equation:

$$\frac{1}{R}\frac{d^2R}{dr^2} + \frac{1}{Rr}\frac{dR}{dr} = -\frac{1}{Z}\frac{d^2Z}{dz^2} .$$

As before, since both sides of the equations are functions of only a single variable, both sides must be equal to a constant (λ^2). Thus, the first of two resultant equations are

$$r^2\frac{d^2R}{dr^2} + r\frac{dR}{dr} - \lambda^2 r^2 R = 0.$$

This is a modified Bessel equation which has a general solution of

$$R(r) = CI_o(\lambda r) + DK_o(\lambda r). \qquad (3.16)$$

Since T_3 (∞,Z) = 0 or finite, C must be zero.

The second equation from separation of variable yields

$$\frac{d^2Z}{dz^2} + \lambda^2 Z = 0.$$

The general solution for Z(z) is

$$Z(z) = A \sin(\lambda z) + B \cos(\lambda z).$$

Since $T_3(r,0) = 0$, $B = 0$ (because cos (0) is unity).

Also since $T_3(r,L) = 0$,

and if $A \neq 0$, $\sin(\lambda L) = 0$ at $\lambda L = n\pi$, $n = 1,2,\ldots$

Therefore, $\lambda L = n\pi$.

or $\lambda = \frac{n\pi}{L}$, and

$$Z = A \sin\left(\frac{n\pi}{L} z\right). \tag{3.17}$$

Finally the overall solution for T_3 is

$$T_3(r,z) = \sum_{n=1}^{\infty} B_n K_o(\lambda r) \sin\left(\frac{n\pi}{L} z\right), \tag{3.18}$$

where $B_n = AC$ for each value of λ.

For the annular region containing the heat source, i.e., $r_o \leq r \leq r_1$, the following model formulation was used. Let the volumetric heat generation rate of the heat source be designated q''' and the temperature in the region T_2. The Fourier heat conduction equation which contains a source term is

$$\frac{\partial^2 T_2}{\partial r^2} + \frac{1}{r}\frac{\partial T_2}{\partial r} + \frac{\partial^2 T_2}{\partial z^2} + \frac{q'''}{k_c} = 0. \tag{3.19}$$

As the above equation is inhomogeneous due to the presence of the heat source term, the analytical solution requires superposition.

$$\text{Let } T_2(r,z) = T_4(r,z) + \phi(z).$$

Using separation of variables and substitution in Eq. (3.19) yields the ordinary differential equation:

$$\frac{d^2 T_4}{dr^2} + \frac{1}{r}\frac{dT_4}{dr} + \frac{d^2 T_4}{dz^2} + \phi''(z) + \frac{q'''}{k_c} = 0. \tag{3.20}$$

The above equation is split into two parts and solved separately using separation of variables. The first part of the Eq. (3.20) is

$$\frac{d^2T_4}{dr^2} + \frac{1}{r}\frac{dT_4}{dr} + \frac{d^2T4}{dz^2} = 0.$$

The boundary conditions are

$$T_4\ (r,0) = 0,$$

$$T_4\ (r,L) = 0.$$

Substitution for $T_4(r,z)$ in Eq. (3.20) followed by division by $R(r)Z(z)$ yields

$$\frac{1}{R}\frac{d^2R}{dr^2} + \frac{1}{Rr}\frac{dR}{dr} = -\frac{1}{Z}\frac{d^2Z}{dz^2},$$

which is as before a modified Bessel equation having a general solution of

$$R(r) = CI_o(\lambda r) + DK_o(\lambda r).$$

The second equation from the separation of variables is

$$\frac{d^2Z}{dz^2} + \lambda^2 Z = 0.$$

The general solution for $Z(z)$ as before is

$$Z(z) = A \sin(\lambda z) + B \cos(\lambda z).$$

Since $T_4(r,0) = 0$ and $T_4(r,L) = 0$, the particular solution is

$$Z(z) = A \sin\left(\frac{n\pi}{L} z\right).$$

Therefore, the overall solution for T_4 is

$$T_4(r,z) = \sum_{n=1}^{\infty} [C_n I_o(\lambda r) + D_n K_o(\lambda r)] \sin\left(\frac{n\pi}{L} z\right). \tag{3.21}$$

The second part of Eq. (3.20) is

$$\phi''(z) + \frac{q'''}{k_c} = 0.$$

The boundary conditions are

$$\phi(0) = 0,$$

$$\phi(L) = 0.$$

Integrating the differential equation twice,

$$\phi(z) = -\frac{q'''}{k_c}\left(\frac{z^2}{2}\right) + C_1 z + C_2.$$

Since $\phi(0) = 0$, the arbitrary constant $C_2 = 0$.

Since $\phi(L) = 0$, $C_1 = \left(\frac{q'''}{k_c}\right)\left(\frac{L}{2}\right)$.

Therefore, the solution for $\phi(z)$ is

$$\phi(z) = -\left(\frac{q'''}{k_c}\right)\left(\frac{z^2}{2}\right) + \left(\frac{q'''}{k_c}\right)\left(\frac{L}{2}\right)(z). \tag{3.22}$$

The final solution for T_2 is

$$T_2(r,z) = \sum_{n=1}^{\infty} [C_n I_o(\lambda r) + D_n K_o(\lambda r)] \sin\left(\frac{n\pi}{L} z\right)$$
$$- \left(\frac{q'''}{k_c}\right)\left(\frac{z^2}{2}\right) + \left(\frac{q'''}{k_c}\right)\left(\frac{L}{2}\right) z. \tag{3.23}$$

The solutions for the temperatures T_1, T_2 and T_3 are

$$T_1(r,z) = \sum_{n=1}^{\infty} A_n I_o(\lambda r) \sin\left(\frac{n\pi}{L} z\right), \tag{3.9}$$

$$T_2(r,z) = \sum_{n=1}^{\infty} [C_n I_o(\lambda r) + D_n K_o(\lambda r)] \sin\left(\frac{n\pi}{L} z\right)$$

$$- \left(\frac{q'''}{k_c}\right)\left(\frac{z^2}{2}\right) + \left(\frac{q'''}{k_c}\right)\left(\frac{L}{2}\right) (z), \qquad (3.23)$$

$$T_3(r,z) = \sum_{n=1}^{\infty} B_n K_o(\lambda r) \sin\left(\frac{n\pi}{L} z\right). \qquad (3.18)$$

The remaining boundary conditions required in the evaluation of the arbitrary constants (A_n, B_n, C_n, and D_n, n = 1, 2, ..., ∞) are the continuity equations for flux and temperature. They are

$$k_s \frac{\partial T_3}{\partial r} (r_1,z) = k_c \frac{\partial T_2}{\partial r} (r_1,z), \qquad (3.24)$$

$$k_a \frac{\partial T_1}{\partial r} (r_o,z) = k_c \frac{\partial T_2}{\partial r} (r_o,z), \qquad (3.25)$$

$$T_3(r_1,z) = T_2(r_1,z), \qquad (3.26)$$

$$T_1(r_o,z) = T_2(r_o,z). \qquad (3.27)$$

Using Eq. (3.27) and substituting the expressions for T_1 and T_2 yields

$$\sum_{n=1}^{\infty} A_n I_o(\lambda r_o) \sin\left(\frac{n\pi}{L} z\right) = \sum_{n=1}^{\infty} [C_n I_o(\lambda r_o) + D_n K_o(\lambda r_o) \sin\left(\frac{n\pi}{L} z\right)$$

$$- \left(\frac{q'''}{k_c}\right)\left(\frac{z^2}{2}\right) + \left(\frac{q'''}{k_c}\right)\left(\frac{L}{2}\right) z.$$

Using the orthogonality condition and expanding the series expressions, together with the definite integrals yields

$$A_n I_o(\lambda r_o)\frac{L}{2} = [C_n I_o(\lambda r_o) + D_n K_o(\lambda r_o)]\frac{L}{2}$$

$$-\left(\frac{q'''}{k_c}\right)\left(\frac{1}{2}\right)\left\{\frac{(-1)^n[2-n^2\pi^2]-2}{\frac{n^3\pi^3}{L^3}}\right\} + \left(\frac{q'''}{k_c}\right)\left(\frac{L}{2}\right)\left(\frac{-L^2(-1)^n}{n\pi}\right),$$

$$A_n I_o(\lambda r_o) = C_n I_o(\lambda r_o) + D_n K_o(\lambda r_o) - \left(\frac{q'''}{k_c}\right)\left(\frac{L^2}{\pi^3}\right)\left\{\frac{(-1)^n[2-n^2\pi^2+n^2\pi^2]-2}{n^3}\right\},$$

$$\text{or } A_n I_o(\lambda r_o) = C_n I_o(\lambda r_o) + D_n K_o(\lambda r_o) - \left(\frac{q'''}{k_c}\right)\left(\frac{L^2}{\pi^3}\right)\left|\frac{2(-1)^n-2}{n^3}\right|.$$

Let the expression $\left(\frac{-q'''}{k_c}\right)\left(\frac{L^2}{\pi^3}\right)\left(\frac{2(-1)^n-2}{n^3}\right)$ = Term.

Therefore the arbitrary constant A_n is

$$A_n = C_n + \frac{D_n K_o(\lambda r_o)}{I_o(\lambda r_o)} + \frac{\text{Term}}{I_o(\lambda r_o)}\ . \qquad (3.28)$$

Using the boundary condition in Eq. (3.26) and substituting the expressions for the temperatures T_2 and T_3 yields

$$B_n K_o(\lambda r_1) = C_n I_o(\lambda r_1) + D_n K_o(\lambda r_1) + \text{Term}.$$

Therefore,

$$B_n = \frac{C_n I_o(\lambda r_1)}{K_o(\lambda r_1)} + D_n + \frac{\text{Term}}{K_o(\lambda r_1)}\ . \qquad (3.29)$$

The boundary condition in Eq. (3.24) is

$$k_c \frac{\partial T_2}{\partial r}(r,z) = k_s \frac{\partial T_3}{\partial r}(r_1,z).$$

The left hand side of the above equation on substitution is

$$k_c \frac{\partial T_2}{\partial r} = k_c \sum_{n=1}^{\infty} [C_n \lambda I_1(\lambda r) + D_n \lambda(-1)K_1(\lambda r)] \sin\left(\frac{n\pi}{L} z\right)\ .$$

Similarly the right hand side of the equation is

$$k_s \frac{\partial T_3}{\partial r} = k_s \sum_{n=1}^{\infty} - B_n \lambda k_1(\lambda r) \sin\left(\frac{n\pi}{L} z\right) .$$

Now using the boundary condition in Eq. (3.25), the left hand side reduces to

$$k_a \frac{\partial T_1}{\partial r} = k_a \sum_{n=1}^{\infty} A_n \lambda I_1(\lambda r) \sin\left(\frac{n\pi}{L} z\right) .$$

But,

$$k_c \frac{\partial T_2}{\partial r}\bigg|_{r=r_o} = k_a \frac{\partial T_1}{\partial r}\bigg|_{r=r_o} ,$$

$$k_c \, [C_n \lambda I_1(\lambda r_o) - D_n K_1(\lambda r_o)\lambda] \, \frac{L}{2} = k_a A_n \lambda I_1(\lambda r_o) \, \frac{L}{2} .$$

Therefore,

$$A_n = \frac{k_c}{k_a} \, [C_n - D_n \frac{k_1(\lambda r_o)}{I_1(\lambda r_o)}] . \tag{3.30}$$

Similarly,

$$k_c \frac{\partial T_2}{\partial r}\bigg|_{r=r_1} = k_s \frac{\partial T_3}{\partial r}\bigg|_{r=r_1} ,$$

$$k_c \, [C_n \lambda I_1(\lambda r_1) - D_n \lambda k_1(\lambda r_o)] \, \frac{L}{2} = k_s(-B_n)\lambda K_1(\lambda r_1) \, \frac{L}{2} .$$

Therefore,

$$B_n = \frac{k_c}{k_s} \left(D_n - \frac{C_n \, I_1 \, (\lambda r_1)}{k_1(\lambda r_1)}\right) . \tag{3.31}$$

From Eqs. (3.28) and (3.30) and equating the expressions for A_n yields

$$\left(\frac{k_c}{k_a}\right) \left(C_n - \frac{D_n \, K_1(\lambda r_o)}{I_1(\lambda r_o)}\right) = C_n + \frac{D_n K_o(\lambda r_o)}{I_o(\lambda r_o)} + \frac{\text{Term}}{I_o(\lambda r_o)} ,$$

$$C_n \left(\frac{k_c}{k_a} - 1\right) = D_n \left(\frac{k_c}{k_a}\frac{K_1(\lambda r_o)}{I_1(\lambda r_o)} + \frac{K_o(\lambda r_o)}{I_o(\lambda r_o)} + \frac{\text{Term}}{I_o(\lambda r_o)}\right) .$$

Let the expression $\left(\frac{k_c}{k_a}\frac{K_1(\lambda r_o)}{I_1(\lambda r_o)} + \frac{K_o(\lambda r_o)}{I_o(\lambda r_o)}\right) = \text{Term 3}$.

Therefore,

$$C_n = \frac{D_n \text{ (Term 3)} + \frac{\text{Term}}{I_o(\lambda r_o)}}{\left(\frac{k_c}{k_a} - 1\right)} .$$

From Eqs. (3.29) and (3.31) equating the expressions for the arbitrary constant B_n,

$$\frac{k_c}{k_s}\left(D_n - \frac{C_n I_1(\lambda r_1)}{K_1(\lambda r_1)}\right) = \frac{C_n I_o(\lambda r_1)}{K_o(\lambda r_1)} + D_n + \frac{\text{Term}}{K_o(\lambda r_1)} ,$$

$$D_n \left(\frac{k_c}{k_s} - 1\right) = C_n\left(\frac{I_o(\lambda r_1)}{K_o(\lambda r_1)} + \frac{k_c}{k_s}\frac{I_1(\lambda r_1)}{K_1(\lambda r_1)} + \frac{\text{Term}}{K_o(\lambda r_1)}\right) ,$$

$$C_n = \frac{D_n \left(\frac{k_c}{k_s} - 1\right) - \frac{\text{Term}}{K_o(\lambda r_1)}}{\frac{I_o(\lambda r_1)}{K_o(\lambda r_1)} + \frac{k_c}{k_s}\frac{I_1(\lambda r_1)}{K_1(\lambda r_1)}} . \tag{3.32}$$

Let the expression $\left(\frac{I_o(\lambda r_1)}{K_o(\lambda r_1)} + \frac{k_c}{k_s}\frac{I_1(\lambda r_1)}{K_1(\lambda r_1)}\right) = \text{Term 2}$.

Therefore,

$$\left(D_n \left(\frac{k_c}{k_s} - 1\right) - \frac{\text{Term}}{K_o(\lambda r_1)}\right)\left(\frac{k_c}{k_a} - 1\right) = \text{Term 2}\left(D_n \text{ Term 3} + \frac{\text{Term}}{I_o(\lambda r_o)}\right) ,$$

$$D_n \left(\left\{\frac{k_c}{k_s} - 1\right\}\left\{\frac{k_c}{k_a} - 1\right\} - \text{Term2} \cdot \text{Term3}\right) = \left(\frac{\text{Term}\cdot\text{Term2}}{I_o(\lambda r_o)} + \frac{\text{Term}}{K_o(\lambda r_1)}\left(\frac{k_c}{k_a} - 1\right)\right) ,$$

Therefore,

$$D_n = \frac{\frac{\text{Term} \cdot \text{Term 2}}{I_o(\lambda r_o)} + \frac{\text{Term}}{K_o(\lambda r_1)}\left(\frac{k_c}{k_a} - 1\right)}{\left(\frac{k_c}{k_s} - 1\right)\left(\frac{k_c}{k_a} - 1\right) - \text{Term 2} \cdot \text{Term 3}} . \tag{3.33}$$

Thus, the expression for A_n, Eq. (3.28), can be formulated without the arbitrary constants C_n and D_n by using Eq. (3.33) in Eq. (3.32) and substituting the result along with the expression for D_n, Eq. (3.33), into Eq. (3.28). Similarly, we can eliminate C_n and D_n from the expression for B_n, Eq. (3.29). These results can then be used in Eqs. (3.9), (3.23), and (3.18) to find $T_1(r,z)$, $T_2(r,z)$, and $T_3(r,z)$, respectively.

In the development of this model the z-dependence was included. However, the temperatures of interest were only those which are maximum, i.e., at the midpoint in the z direction where $\sin\left(\frac{n\pi}{L} z\right)$ is unity. This eliminates the need to define precisely the temperature in the region below and above the calcined waste and the inner air (or soil-filled) regions. In fact this model does not define the temperatures well in these regions. To do this properly would require another system of equations for each of these regions and boundary conditions which match temperatures and heat flux at the interfaces. This is a fairly complex development, but is not needed because the maximum temperature occurs at the midpoint in the z-direction and none of the boundary conditions in the z-direction will impact the calculation of the maximum temperature.

Finally an explanation of the boundary condition below Eq. (3.8), i.e., $T_1(r,0) = T_1(r,L) = 0$, is needed. Zero was used as a convenience.

This means all temperatures calculated using this model should be temperatures above ambient conditions (at the top, $z = 0$) and earth equilibrium temperatures (at the bottom, $z = L$).

CHAPTER 4

RESULTS AND DISCUSSION

A source program written in FORTRAN for numerical evaluation of the analytical solutions for the temperatures in the regions is included as Appendix 1. The source program includes two subroutines needed in the calculation of the Bessel routes for the modified Bessel functions and for evaluation of the various constants in the analytical solutions.

Parameters influencing the maximum temperature within the vessel containing the solid waste strongly affect the design of the storage facility. The following parameters are pertinent to the results:

i) Physical properties of the solid waste and soil.

ii) Physical dimension of the heat source, namely the air column radius, the annular source surrounding the air column, the near field dimension in the soil and the burial depth of the containers.

iii) Magnitude of the heat generation rate.

iv) Boundary conditions imposed upon the model.

All the above physical parameters which affect the calculated temperature profiles were studied in the parametric calculations, using the source program. For validating the results for the solid configuration of the bin, the air column radius was taken as a very small value (0.001 inch) and the calculations were performed using the same source program as that of the annular cylinder. The various interpretations of these parametric studies/calculations are as follows:

a) Solid Bin Configuration:

1) The temperature profiles for (the solid geometry) of a 3 ft radius storage bin were calculated with a heat generation rate

of 50 (Btu/hr ft^3). Figure 4.1 and Table 4.1 show the temperature profiles at different vertical cross sections in the storage bin. The temperatures are the highest at the vertical mid-section in the radial direction (1694.97°F) at center vertical mid-section, declining to a value of 408°F at a radius of 16 ft in the near field in the soil). The numerical values are very close to those calculated in the previous study in their model (i.e., 1600°F).[1]

2) The thermal conductivity of the solid waste has a definite influence on the calculated maximum temperatures. This is one of the important design aspects in deciding on a specific waste form, as the calcines resulting from the various feed solutions (as zirconia calcine, alumina calcine, etc.) have varying thermal conductivities. The maximum bin temperatures are influenced by the thermal conductivities of the product. The lower the thermal conductivity of the calcine, the higher is the maximum temperature. This effect is shown in Fig. 4.2 and Table 4.2. The plots are for thermal conductivity as a parameter and the effects of three values (i.e., 0.15, 0.2 and 0.3 Btu/hr ft °F) are shown. However, thermal conductivity has no effect on the soil temperatures and influences the temperature profiles within the bin only.

3) An increase in heat generation rate, results in an increase in the maximum temperature profile. This effect is shown in Fig. 4.3 and Table 4.3. For a heat generation rate of 50 (Btu/hr ft^3), the maximum temperature that is experienced in the bin is as high as 1694.95°F.

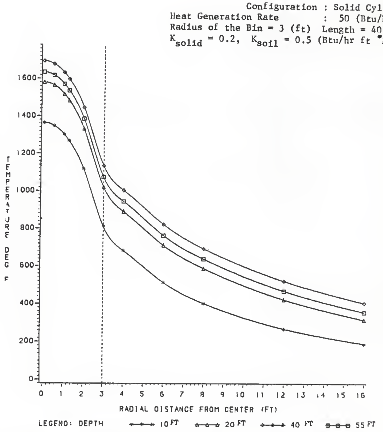


Fig. 4.1 Temperature Profile at Vertical Sections in the Bin- Solid Cylinder Configuration.

Table 4.1. Steady State Temperature Profile at Different Depths in Calcine Bins for the Solid Cylinder Configuration.

Radial Distance (ft)	Temperatures (°F)			
	Depth=10 ft	Depth=20 ft	Depth=40 ft	Depth=55 ft
0.0001	1363.15	1580.00	1694.97	1634.34
0.0002	1363.15	1580.00	1694.97	1634.34
0.0004	1363.15	1580.00	1694.97	1634.34
0.0005	1363.15	1580.00	1694.97	1634.34
0.0010	1363.15	1580.00	1694.97	1634.34
0.5000	1348.50	1564.25	1679.18	1617.62
1.0000	1302.37	1517.93	1632.43	1570.93
1.2500	1268.18	1482.93	1597.31	1536.31
2.0000	1118.62	1331.68	1445.87	1384.68
3.0000	811.43	1020.68	1134.06	1073.12
4.0000	683.77	890.28	1004.03	944.47
6.0000	514.12	712.88	823.57	764.53
8.0000	403.93	589.77	696.37	640.30
12.0000	269.75	425.05	523.86	470.66
16.0000	191.30	318.43	407.99	358.86

Length of the calcine bin = 40 ft.
Bin radius = 3 ft.
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)
Thermal conductivity of calcine = 0.2 (Btu/hr ft °F)
Heat generation rate = 50 (Btu/hr ft^3)

Configuration : Solid Cylinder
Heat Generation Rate 20 (Btu/hr ft^3).
Radius of the Bin = 3 (ft) Length = 40 (ft).
K_{soil} = 0.5 (Btu/hr ft °F) .

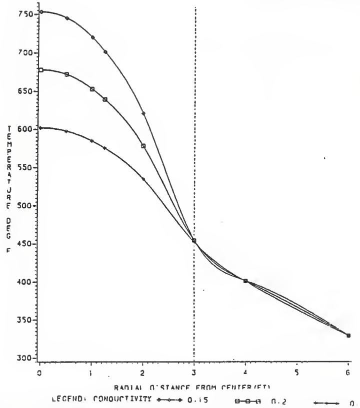


Fig. 4.2 Effect of Waste Thermal Conductivity on Maximum Temperature-Solid Cylinder Configuration.

Table 4.2. Effect of Waste Thermal Conductivity on Calculated Steady State Temperature Distribution in and Around Calcine Bins for Solid Cylinder Configuration.

Radial Distance (ft)	Temperature (°F)		
	$K_{\text{soild waste}}$ (Btu/hr ft °F)		
	0.15	0.2	0.3
0.0001	753.31	678.13	601.99
0.0002	753.31	678.13	601.99
0.0004	753.31	678.13	601.99
0.0005	753.31	678.13	601.99
0.0010	753.31	678.13	601.99
0.5000	745.44	672.25	597.73
1.0000	720.69	653.44	585.29
1.2500	702.00	639.56	575.98
2.0000	621.38	579.06	535.48
3.0000	455.06	454.63	452.43
4.0000	401.98	401.61	400.87
6.0000	329.75	329.42	328.77

Bin radius = 3 ft, length = 40 ft
Length of bin = 40 ft
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)
Heat generation rate = 20 (Btu/hr ft^3)

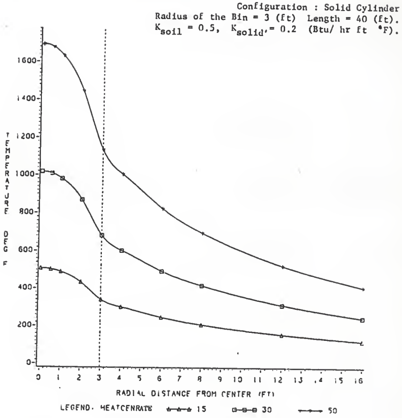


Fig. 4.3 Effect of Heat Generation Rate on Maximum Temperature- Solid Cylinder Configuration.

Table 4.3. Effect of Heat Generation Rate on Calculated Steady State Temperature Distribution in and around Calcine Bins for Solid Bin Configuration.

Radial Distance (ft)	Temperature (°F)		
	Heat Generation Rate (Btu/hr ft^3)		
	50.0	30.0	15.0
0.0001	1694.97	1017.09	508.53
0.0002	1694.97	1017.09	508.53
0.0004	1694.97	1017.09	508.53
0.0005	1694.97	1017.09	508.53
0.001	1694.97	1017.09	508.53
0.500	1679.19	1008.13	503.56
1.000	1632.44	980.19	489.55
2.00	1445.88	868.31	433.45
4.00	1004.03	602.42	301.20
6.00	823.58	494.14	247.07
8.00	696.37	417.82	208.91
12.00	523.87	314.32	157.16
16.00	408.00	244.80	122.40

Bin radius = 3 (ft)
Length of bin = 40 (ft)
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)
Thermal conductivity of solid = 0.2 (Btu/hr ft °F)

4) Increase in bin diameter increases the peak temperature appreciably (as shown in Fig. 4.4 and Table 4.4). This is in direct proportion to the total inventory of the solids in the bin and therefore of the total heat input. This relationship is an important consideration in design of the maximum diameter of the storage bin (directly buried bins) while not exceeding the allowable temperature limits. For a heat generation rate of 20 (Btu/hr ft^3), and with a thermal conductivity of 0.3 (Btu/hr ft °F) a bin radius of only 4 ft results in peak temperature as high as 1993°F (for a solid configuration of the bin).

5) The effect of burial depth of the bins below the ground level is shown in Fig. 4.5 and Table 4.5. The burial depth below grade has marginal effects on the peak temperatures. In all the above cases the peak temperatures are evaluated at respective vertical mid-section of the bins depending upon the burial depths. This follows from the boundary conditions that the bin system is assumed to be a continuous heat source extending the system length 'L'.

The following are a similar set of calculations for the annular configuration of bin geometry, developed in this study. The parametric values of variables used in the computation of temperatures (such as the heat generation rates, thermal conductivities, the bin configuration, etc.) are similar for easy comparison with solid bin configuration on common basis. In the parametric studies, the temperature profiles at the vertical section as a function of the radial distances were computed for different variables.

Configuration: Solid Cylinder
Heat Generation Rate = 20 (Btu/ hr ft^3)
K_{soil} = 0.3 , K_{solid} = 0.1 (Btu/ hr ft °F).

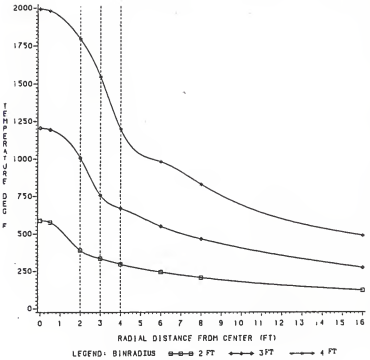


Fig. 4.4 Effect of Bin Radius on Maximum Temperature- Solid Cylinder Configuration.

Table 4.4. Effect of Bin Radius on Calculated Steady State Temperature Distribution in and around Calcine Bin.

Radial Distance (ft)	Temperature (°F)		
	Bin Radius (ft)		
	2.0	3.0	4.0
0.0001	588.54	1204.88	1993.52
0.0002	588.54	1204.88	1993.52
0.0004	588.54	1204.88	1993.52
0.0005	588.54	1204.88	1993.52
0.0010	588.54	1204.88	1993.52
0.5	577.25	1193.5	1981.81
2.0	390.37	1006.62	1795.13
3.0	334.80	757.37	1546.56
4.0	296.65	669.76	1197.56
6.0	243.28	549.41	981.29
8.0	205.67	464.57	829.93
16.0	120.40	272.23	486.88

Heat generation rate = 20 (Btu/hr ft^3)
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)
Thermal conductivity of solid waste = 0.1 (Btu/hr ft °F)

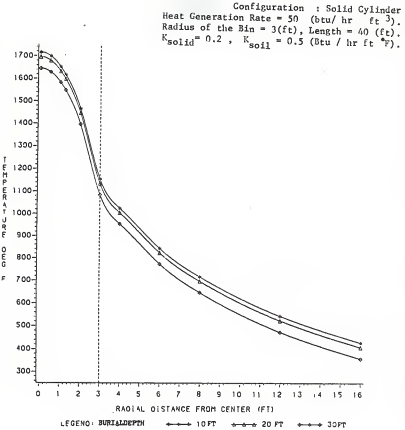


Fig. 4.5 Effect of Burial Depth on Maximum Temperature- Solid Cylinder Configuration.

Fig. 4.5. Effect of the Burial Depth of the Bins Below Grade on the Maximum Temperature.

Radial Distance (ft)	Temperature (°F) Burial Depth (ft) 10	20	30
0.0001	1718.21	1694.97	1642.54
0.0002	1718.21	1694.97	1642.54
0.0004	1718.21	1694.97	1642.54
0.0005	1718.21	1694.97	1642.54
0.0010	1718.21	1694.97	1642.54
0.500	1699.18	1679.18	1629.27
1.00	1652.43	1632.27	1582.43
1.25	1617.31	1597.24	1547.81
2.00	1465.87	1445.82	1395.91
3.00	1154.06	1131.91	1084.36
4.00	1021.03	1004.03	952.27
6.00	842.57	823.57	772.37
8.00	716.37	696.37	641.35
12.00	541.82	523.86	473.21
16.00	425.10	407.99	354.91

Length of the calcine bin = 40 (ft)
Bin radius = 3 (ft)
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)
Thermal conductivity of calcine = 0.2 (Btu/hr ft °F)
Heat generation rate = 50 (Btu/hr ft^3)

b) Annular Configuration:

6) Introduction of an air column at the center reduces the peak temperatures, for the same set of conditions of that of the solid bin configuration. Figure 4.6 and Table 4.6 show the temperature profile at the vertical mid-section for the annular configuration. As can be seen the annular configuration is advantageous because of the reduction in the peak temperature by 300°F (approximately, 1694°F vs 1416°F for the annular configuration). Also seen from the relationships, the air column temperature remains constant and is also the maximum temperature in the bin system. However, the soil temperatures in the near field are not affected and are the same for both of the configurations. The temperature profile at different cross sections within the annular bin configuration are correspondingly lower when compared with that of the solid configuration.

7) Figure 4.7 and Table 4.7 show the effect of the variation of the thermal conductivities of the waste products for annular bin configurations. As can be seen the values are only slightly lower when compared with those of the values for the solid bin configurations.

8) The rate of heat generation for the annular configuration similarly affects the maximum temperatures as in the previous case, but the maximum temperatures are lower when compared with solid configuration (Fig. 4.8 and Table 4.8).

9) For the same heat generation rate and the total heat input, increasing the air column radii reduces the peak temperature (i.e., 1630 to 1416°F for air column radii 0.5, 1.0 and 1.5 ft

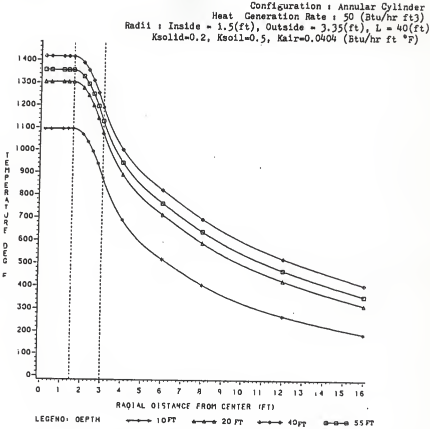


Fig. 4.6 Temperature Profiles at Vertical Sections in the Bin- Annular Cylinder Configuration.

Table 4.6. Steady State Temperature Profile at Nodal Points in Calcine Bins for Annular Configuration.

Radial Distance (ft)	Temperature (°F)			
	Depth (ft)			
	10.0	10.0	40.0	55.0
0.10	1094.71	1303.72	1416.21	1356.34
0.40	1094.86	1303.76	1416.23	1356.36
1.00	1095.69	1304.04	1416.35	1356.51
1.25	1096.24	1304.24	1416.41	1356.62
1.50	1096.90	1304.52	1416.47	1356.78
2.00	1070.69	1277.56	1389.25	1328.50
2.25	1038.19	1244.44	1356.19	1295.69
2.50	994.13	1199.94	1311.63	1250.94
2.75	939.81	1145.00	1256.69	1196.13
3.00	875.44	1080.00	1191.31	1130.81
4.00	689.61	891.21	1003.22	944.71
6.00	518.44	713.97	823.11	764.83
8.00	407.10	590.79	696.07	640.73
12.00	271.43	425.86	523.85	741.10
16.00	192.20	319.03	408.13	359.25

Inside radius = 1 ft (air)
Outside radius = 3.35 ft (calcine)
Length of bin = 40 ft
Thermal conductivity of solid waste = 0.2 (Btu/hr ft °F)
Heat generation rate = 50 (Btu/hr ft^3)

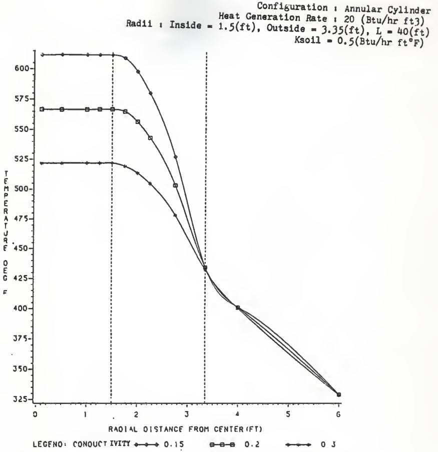


Fig. 4.7 Effect of Waste Thermal Conductivity on Maximum Temperature-Annular Bin Configuration.

Table 4.7. Effect of Waste Thermal Conductivity on the Maximum Temperature for Annular Bin Configuration.

Radial Distance (ft)	Maximum Temperature (°F)		
	Thermal Conductivity of Calcine (Btu/hr ft °F)		
	0.15	0.2	0.3
0.1	611.76	566.71	521.95
0.5	611.78	566.72	521.97
1.0	611.82	566.76	522.01
1.25	611.85	566.78	522.03
1.5	611.86	566.80	522.06
1.75	609.13	565.06	519.37
2.0	597.88	556.63	513.81
2.25	580.13	543.31	504.95
2.75	527.25	503.63	478.45
3.35	434.88	434.50	432.43
4.00	401.64	401.29	400.58
6.00	329.55	329.24	328.62

Inside radius = 1.5 ft (air)
Outside radius = 3.35 ft (calcine)
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)
Heat generation rate = 20 (Btu/hr ft^3)

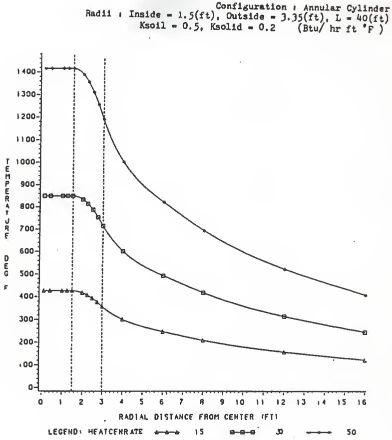


Fig. 4.8 Effect of Heat Generation Rate on Maximum Temperature- Annular Bin Configuration.

Table 4.8. Effect of Heat Generation Rate on Calculated Steady State Temperature Distribution in Calcine Bins for Annular Bin Configuration.

Radial Distance (ft)	Temperatures (°F) Heat Generation Rate (Btu/hr ft^3)		
	50.0	30.0	15.0
0.1	1416.21	850.04	425.05
0.4	1416.23	850.05	425.06
1.00	1416.35	850.12	425.09
1.25	1416.41	850.16	425.11
1.50	1416.47	850.20	425.13
2.00	1389.25	834.69	416.54
2.25	1356.19	814.69	406.57
2.50	1311.63	787.69	393.19
2.75	1256.69	754.88	376.73
3.00	1191.31	715.69	357.13
4.00	1003.22	601.93	300.96
6.00	823.11	493.86	246.93
8.00	696.07	417.64	208.82
12.00	523.85	314.31	157.16
16.00	408.13	244.88	122.44

Inside radius = 1.5 ft (air)
Outside radius = 3.35 ft (calcine)
Thermal conductivity of solid waste = 0.2 (Btu/hr ft °F)
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)

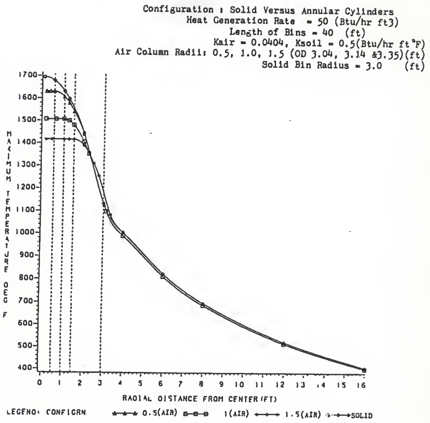


Fig. 4.9 Comparison of Maximum Temperatures for Solid Bin Versus Annular Bin for the Same Heat Input.

Table 4.9. Comparison of Maximum Temperatures for Solid Bin Versus Annular Bins for Same Heat Input.

Radial Distance (ft)	Temperature (°F)			
	Solid Cylinder	Air 0.5 ft	Air 1.0 ft	Air 1.5 ft
0.1	1694.06	1630.77	1526.28	1416.21
0.2	1692.19	1630.77	1526.28	1416.22
0.3	1689.00	1630.76	1526.29	1416.22
0.4	1684.81	1630.76	1526.30	1416.23
0.5	1679.18	1630.75	1526.30	1416.25
1.0	1632.44	1606.31	1526.94	1416.35
1.25	1597.31	1578.38	1519.81	1416.41
1.50	1554.81	1541.06	1499.56	1416.47
2.00	1445.88	1440.94	1426.38	1389.25
4.00	1004.03	1003.36	1003.46	1003.22
6.00	823.58	823.05	823.20	823.11
8.00	696.37	695.93	696.09	696.07
12.00	523.87	523.57	523.76	523.85
16.00	408.0	407.78	407.98	408.13

Heat generation rate = 50 (Btu/hr ft^3)
Length of the bins = 40 (ft)
Thermal conductivity of air (at 1400°K) = 0.0404 (Btu/hr ft °F)
Thermal conductivity of solid waste = 0.2 (Btu/hr ft °F)
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)
Bin radii = 3 ft (nominal)
Air column radii = 0.5, 1.0 and 1.5 (ft)

respectively). By collapsing the air column to approximately zero leads to the solid cylinder configuration with a peak temperature of 1694°F. For the purpose of comparison and validating the results, total heat input was assumed to be the same in all three annular and solid configurations compared by proportionately increasing the outer diameter.

10) Table 4.10 shows the temperatures in the annular bin with the inner annulas filled with soil instead of air as in the previous case. As can be seen, the resultant temperature profiles at respective nodes are only marginally lower than the annular bin configuration filled with air at the center (Fig. 4.6 and Table 4.6 i.e., 1416.21 versus 1412.59°F). So it is apparent that the reduction of peak temperature by 300°F over the solid configuration stems from the increased surface area of the annular bin for heat dissipation. Thus the central air column can be back filled with earth without affecting the temperature profiles.

Conclusions and Recommendations:

Based on the temperature distribution calculated in the numerical examples and the parametric studies, the direct burial of solidified nuclear wastes in the soil without external cooling seems feasible. However, the results and temperatures calculated based upon these models should be considered only as a feasibility study and the conclusion that such simplified underground storage can dissipate appreciable amount of heat by way of conduction to soil is only qualitative. The mathematical models are valid only for the set of physical conditions assumed in the

Table 4.10. Effect of Back Fill Soil in the Central Air Column for the Annular Bin Configuration.

Radial Distance (ft)	Temperature in Vertical Mid-Section of the Bin (°F) Central Column	
	Air	Soil Back-fill
0.1	1416.21	1412.59
0.5	1416.25	1412.63
1.00	1416.35	1412.74
1.25	1416.41	1412.82
1.50	1416.47	1412.91
1.75	1410.31	1406.56
2.00	1389.25	1385.94
2.25	1356.19	1253.00
2.75	1256.69	1253.75
3.35	1083.75	1080.94
4.00	1003.22	1000.75
6.00	823.11	820.89
8.00	696.07	694.09
12.00	523.85	522.14
16.00	408.13	406.64

Inside radius = 1.5 (ft) Outside radius = 3 (ft)
Length of the bin = 40 (ft)
Heat generation rate = 50 (Btu/hr ft^3)
Thermal conductivity of solid waste = 0.2 (Btu/hr ft °F)
Thermal conductivity of soil = 0.5 (Btu/hr ft °F)
Thermal conductivity of air = 0.0404 (Btu/hr ft °F)

model, together with the set of boundary conditions to simplify mathematics. The radiological safety implications and the technical aspects of construction of such simplified storage conforming to safety regulations applicable to such burial facilities must be carefully analyzed. In other words, the modelling studies pursued in this research work pertain only to the heat dissipation aspect, which is but one important consideration in the overall scope of design of such simplified storage. Towards that objective the modelling studies are useful and should be able to predict reasonably close temperature estimates for practical systems.

In fact, such simplified (directly buried) storage facilities were considered during the design of the second set of bins at INEL.[12] The main reason why they were discarded later on stem from limitations imposed by maximum size of these bins (solid bins) without exceeding the sllowable maximum temperatures in the bin. However, as is validated in the current model study, annular bin configuration could be a viable alternative with a reduction of maximum bin temperatures as much as 300°F (with an air column radius of 1.5 ft) and perhaps more if the bin could be increased in size. This, of course, should be weighed against construction and cost aspects.

Calculated temperatures based on the steady state models for the solid cylinder configuration were validated by the previous investigators[1] for small diameter heat sources (such as simulated fuel elements of typical diameter 0.14 ft (3.5 inches O.D), by burying heater elements at the National Reactor Testing Station at power levels of 350 and 950 watts (equivalent to 329 and 880 (Btu/hr ft^3) for the fuel element). The temperature profiles were measured at specific grid points (radial distances). The general agreement between the calculated and experimental temperature distribution measured in the field point towards the validity of the model predictions. The models for the large diameter heat sources, resembling waste storage bins could not be field verified due to the problems of simulating them with practical sized heaters of such proportions.

Future modelling studies that could be undertaken in this area are the following:

1) More exact mathematical formulations could include the contact resistances of the various heat conduction regions and the influence of the convective air layer. Due to difficulty in the mathematical formulations and arriving at analytical solutions, they were not included in this study.

2) Another area for future work could be consideration of overall temperature profile estimates for a set of bins buried in a typical layout geometry such as the ones which would be involved in practice. This would require a three dimensional mathematical model (namely radial, depth and azimuthal) for individual bins and the composite temperature profile for a cluster of such bins (assuming a point source or a line source in such calculations representing individual bins). Such a system can at best represent practical burial facilities.

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APPENDIX 1

COMPUTER PROGRAM

The numerical technique for evaluation of the analytical solutions (equations developed in Chapter 3) was programmed on an IBM-370 computer. The main steps involved in the machine solutions consisted of:

i) An arbitrary number (100) summation of the Bessel routines were first evaluated for the arguments R_o and R_1. The arbitrary constants, i.e., ANN, BNN, CNN and DNN in the analytical solutions are evaluated using the summation process. In practice, a summation of the first 100 terms of the series is sufficient and converges to a steady value for the numerical constants.

ii) Summation of the trigonometric terms (required in the computation of the temperatures at specific nodes) are evaluated for the argument $R_2(k)$, representing the radial distances from the center. The number of summation terms are identical to that in the first step.

iii) The temperatures at specific nodes are evaluated as a two dimensional array, U(k,j), representing both the radial and depth directions (r,z), by using the analytical solutions developed for the temperatures T_1, T_2 and T_3. The analytical solutions for T_1, T_2 and T_3 are computed separately and are valid only in respective regions for which they were developed. However, at the boundaries between two adjacent regions, the solutions are valid because of the boundary conditions assumed for the sake of continuity.

iv) The nodes are divided into 5 discrete locations in the radial direction in each of the three individual regions, i.e., air, heat-source and soil. This amounts to a total of 15 radial nodes where the temperatures are evaluated and in turn for different depths. As designated in the program, (1-5) represent the region bounded by the air in the innermost radius, (6-10) the region bounded by the solid waste (heat source) and (11-15) the region in the soil.

v) The outputs are printed out in a tabular format representing the depths (i.e., 10 ft, 20 ft, 40 ft and 55 ft), where 40 ft represents the vertical mid-section of the container, at which the temperatures are the maximum. Depths 20 ft and 60 ft represent the top and bottom of the container from the ground level respectively. The total depth of the system is assumed to be 80 ft including portions of soil above and below the physical dimension of the container. This procedure is analogous to the one used by the previous investigators and results in a steady state solution for the temperature evaluated.

vi) A similar procedure is repeated in the case of the solid cylinder configuration, by assuming the inner radius to be a very small value (typically 0.001 inch) and using the same program. This reduces the inner air region to zero (approximately) and thus simulates the solid geometry.

vii) The computed values for the temperatures for different depths are plotted using standard plotter program.

Nomenclature for the Source Program Two-Cyl Watfiv

ANN,BNN,CNN,DNN	= The constants of the analytical solutions for evaluation of temperatures T_1, T_2, T_3.
Con	= $-(Q/k_c)(L^2/\pi^2)$.
Con1	= $-(Q/k_c)(L^2/\pi^2)(z^2/2)$.
Con2	= $(z/2)(Q/\pi)(L^2/k_c)$.
Denom	= Denominator in the expression for DNN
IORO,IOR1,I1RO, and I1R1	= Modified Bessel Functions of the first kind of the zero and first order respectively.
I,J,K,S	= Counting integers
k_a	= Thermal conductivity of air at respective temperature (Btu/hr ft °F).
k_c	= Thermal conductivity of the solid waste (Btu/hr ft°F)
k_s	= Thermal conductivity of the soil (Btu/hr ft°F).
Kizero,Kione	= Library subroutines required in the evaluation of the various constants in the analytical solution.
KORO,KOR1,K1RO, and K1R1	= Modified Bessel functions of the second king of zero and first order respectively.
L	= Finite depth of the system (ft)
Lambda	= $[n\pi z(j)/L]$, the argument for the trignometric function needed in the evaluation of temperatures at different depth.
N	= Number of terms used in the summation for the temperatures.
Term, Term2 & Term3	= The abbreviations for the various terms used in the development of the analytic solutions.
Numr	= Numerator in the expression for DNN
Q	= Volumetric heat generation rate of the heat source (btu/hr ft^3).
Qterm	= Con1 + Con2.
R_o	= Inside radius of the air column (ft).
R_1	= Outside radius to heat source (ft).

$R_2(K)$ = Radial distances where the temperatures are calculated (ft).

T_1 = Calculated temperature at the air in the inner radius (°F).

T_2 = Calculated temperatures in the solid waste in the storage container (°F).

T_3 = Calculated temperatures in the soil surrounding storage container (°F).

U(k,j) = Calculated temperatures at specific points in and around the waste container (°F).

XI = The value of the Bessel functions KO(X) or Kl(X).

XK = The value of the Bessel functions IO(x) or Il(x).

Xl,Xl,X3 = Arguments for the evaluation of the Bessel routes required in the calculations (ft).

Z = The depth below the ground surface at which points the temperatures are evaluated (ft).

II. Data Input Formats for the Computer Program:

The arrangement for the input data cards and the cards order are given below:

First Card - Title card - Contains the case number and the parameter being studied.

Second Card - Contains thermal conductivity values for the air, solid waste and soil respectively.

Third Card - Contains the inner radius R_0, (Air column) and outer radius R_1, (of the source).

Fourth Card - Contains the heat generation rates (Q) and depth of the system (L).

Fifth Card - Contains the radial distances $R_2(k)$ from the center nodes at which the temperatures are evaluated - (1-5) for the air column, (6-10) for the source and (11-15) for the soil.

Sixth Card - Contains the depths at which temperatures are evaluated.

The source program listing is as follows:

```
$JOB           RAM,TIME=(,20),PAGES=30
C    PROGRAM FOR CALCULATING THE STEADY STATE TEMPERATURE IN THE CALCINE
C    BINS  FOR THE CASE OF ANNULAR GEOMETRY.
C    PROGRAM COMPILED BY S.RAMACHANDRAN ON SEP 2,1984.
C    THE FOLLOWING PROGRAM IS FOR CALCULATING THE STEADY-STATE
C    TEMPERATURES IN THE SOLID WASTE STORED IN  UNDERGROUND  BINS
C    TYPICALLY LIKE THE CALCINE STORAGE BINS AT INEL,IDAHO.
C    THE ANALYTICAL SOLUTIONS TO THE FOURIER HEAT CONDUCTION EQUATIONS
C    ARE PROGRAMMED THROUGH THE MACHINE SOLUTIONS TAKING INTO ACCOUNT THE
C    RESPECTIVE REGIONS.  TYPICALLY THE INNER REGION(RADIUS RO) IS BOUND
C    BY AIR IN THE INNERMOST RADIUS, SURROUNDED BY THE HEAT SOURCE
C    VIZ. THE WASTE CALCINES IN THE NEXT REGION (R1).  THE
C    OUTER REGION IS BOUNDED BY INFINITE SOIL MEDIUM (R2).
C    ******************************************************************
C
C
C    **********DECLARATIONS AND DIMENSION STATEMENTS*******************
       DIMENSION    IOR0(200),KOR0(200),IOR1(200),KOR1(200),I1R0(200),
      1K1R0(200),I1R1(200),K1R1(200),IOR2(200,200),R2(15),
      2KOR2(200,200),Z(10),U(50,50),DNN(200),CNN(200),
      3ANN(200),DNN(200),TITLE(20)
C
C
       REAL RO,R1,Q,L,KA,KC,KS,LANDA,ZETA,NTERM,NUMR,
      1 IORO,KORO,IOR1,KOR1,I1RO,
      2 K1RO,I1R1,K1R1,R2,K1,K2,KOR2,IOR2,BNN,ANN,
      3 Z,U,DNN,CNN
C
C
        INTEGER N,I,J,K
C
C    ******************************************************************
C     READING  THE INPUT DATA CARDS                                    *
C    ******************************************************************
206    READ(5,705,END=9999)TITLE
       READ,KA,KC,KS
       READ,RO,R1
       READ,Q,L
       READ,(R2(K),K=1,15)
       READ,(Z(J),J=1,4)
C    ******************************************************************
C
C    ******************************************************************
C    FORMAT STATEMENTS.                                                *
C    ******************************************************************
705    FORMAT(20A4)
       WRITE(6,705)TITLE
       WRITE(6,1200)KA
1200   FORMAT(2X,' THERMAL CONDUCTIVITY - AIR(BTU/HR-FT-F)=',F5.2)
       WRITE(6,1201)KC
1201   FORMAT(2X,'THERMAL CONDUCTIVITY - SOLID-WASTE(BTU/HR-FT-F)=',F5.2)
       WRITE(6,1202)KS
1202   FORMAT(2X,'THERMAL CONDUCTIVITY - SOIL(BTU/HR-FT-F)=',F5.2)
       WRITE(6,1101)RO,R1
1101   FORMAT(2X,'INSIDE RAD,RO(FT)=',F5.2,2X,'OUTSIDE RAD,R1(FT)=',F5.2)
       WRITE(6,1103)Q,L
1103   FORMAT(2X,'HEATGEN.RATE(BTU/HR-FT3=)',F5.2,2X,'LENGTH=',F5.2)
```

```
      PI=3.14159
      N=100
      PI2=PI*PI
      PI3=PI*PI*PI
      CON=-Q*L**2/KC/PI3
      K1=KC/KS
      K2=KC/KA
      DO 101  I=1,N
      X1=I*R0*PI/L
      X2=I*R1*PI/L
C     ***************************************************************
C     COMPUTATION OF THE BESSEL ROUTINES FOR CALCULATING THE          *
C     ARBITRARY CONSTANTS IN THE EXPRESSIONS FOR TEMPERATURES.        *
C     THE BESSEL ROUTINES FOR THE ARGUMENTS ARE EVALUATED THROUGH     *
C     TWO SUBROUTINES INCLUDED IN THE PROGRAM                         *
C                                                                     *
C     ***************************************************************
      CALL KIZERO(X1,XK,XI)
      KOR0(I)=XK
      IOR0(I)=XI
      CALL KIZERO(X2,XK,XI)
      KOR1(I)=XK
      IOR1(I)=XI
      CALL KIONE (X1,XK,XI)
      K1R0(I)=XK
      I1R0(I)=XI
      CALL KIONE(X2,XK,XI)
      K1R1(I)=XK
      I1R1(I)=XI
C     ***************************************************************
C     EVALUATION OF THE ARBITRARY CONSTANTS REQUIRED IN  THE          *
C     EXPRESSIONS FOR THE ANALYTICAL SOLUTIONS FOR THE TEMPERATURES.  *
C     ANN,BNN,CNN AND DNN ARE THE ARBITRARY CONSTANTS REQUIRED IN     *
C     THE ANALYTICAL SOLUTIONS FOR THE TEMPERATURES T1,T2 & T3.       *
C     THE SUMMATION OF FIRST 100 TERMS IS FOUND SUFFICIENT TO GIVE A  *
C     CONVERGED VALUE  FOR THE ARBITRARY CONSTANTS.                   *
C     ***************************************************************
C
      TERM=((L**2/PI3)*(-Q/KC)*(2*(-1)**I-2))/I**3
      TERM2=(K1*I1R1(I)/K1R1(I))+(IOR1(I)/KOR1(I))
      TERM3=(KOR0(I)/IOR0(I))+(K2*K1R0(I)/I1R0(I))
      DENOM=(K1-1)*(K2-1)-(TERM2*TERM3)
      NUMR=(TERM*TERM2/IOR0(I))+(TERM*(K2-1)/KOR1(I))
      DNN(I)=NUMR/DENOM
      CNN(I)=(DNN(I)*TERM3+TERM/IOR0(I))/(K2-1)
      BNN(I)=K1*(DNN(I)-(CNN(I)*I1R1(I)/K1R1(I)))
      ANN(I)=K2*(CNN(I)-(DNN(I)*K1R0(I)/I1R0(I)))
  101 CONTINUE
      DO 299 I=1,N
      DO 299 K=1,15
      KOR2(K,I)=0.
      IOR2(K,I)=0.
  299 CONTINUE
      DO 245 I=1,N
      S=I
      DO 245 K=1,5
      X3=S*PI*R2(K)/L
```

```
      CALL KIZERO(X3,XK,XI)
      IOR2(K,I)=XI
 245  CONTINUE
C     ****************************************************************
C     REGION K(1,5) REPRESENT THE INNERMOST RADIUS BOUNDED           *
C     BY AIR COLUMN AND THE NODAL TEMPERATURES AT DIFFERENT          *
C     DEPTHS ARE EVALUATED.                                          *
C     ****************************************************************
      DO 2003 J=1,4
      DO 2003 K=1,5
      U(K,J)=0
      DO 204 I=1,N
      S=I
      ZETA=PI*Z(J)/L
      U(K,J)=U(K,J)+ANN(I)*IOR2(K,I)*SIN(S*ZETA)
 204  CONTINUE
 2003 CONTINUE
      DO 250 I=1,N
      S=I
      DO 250 K=6,10
      X3=S*PI*R2(K)/L
      CALL KIZERO (X3,XK,XI)
      KOR2(K,I)=XK
      IOR2(K,I)=XI
 250  CONTINUE
C     ****************************************************************
C     THE REGION K(6,10) REPRESENTS THE REGION BOUNDED BY THE        *
C     HEAT SOURCE - THE TEMPERATURES AT DIFFERENT NODES AT VARIOUS   *
C     DEPTHS ARE EVALUATED.                                          *
C     ****************************************************************
      DO 2000 J=1,4
      DO 2000 K=6,10
      U(K,J)=0.
      DO 201 I=1,N
      S=I
      ZETA=PI*Z(J)/L
      CON1=-(Q/KC)*(L**2/PI2)*(.5*ZETA**2)
      CON2=(ZETA/2)*Q*L**2/KC/PI
      QTERM=CON1+CON2
      U(K,J)=U(K,J)+(CNN(I)*IOR2(K,I)+DNN(I)*KOR2(K,I))*SIN(S*ZETA)
 201  CONTINUE
      U(K,J)=U(K,J)+QTERM
 2000  CONTINUE
      DO 251 I=1,N
      S=I
      DO 251 K=11,15
      X3=S*PI*R2(K)/L
      CALL KIZERO(X3,XK,XI)
      KOR2(K,I)=XK
 251  CONTINUE
C     ****************************************************************
C     THE REGION BOUNDED BY THE INFINITE SOIL MEDIUM IS REPRESENTED *
C     BY K(11,15) AND THE NODAL TEMPERATURES ARE EVALUATED FOR       *
C     DIFFERENT DEPTHS.                                              *
C     ****************************************************************
      DO  2002 J=1,4
      DO 2002 K=11,15
```

```
      U(K,J)=0
      DO 203 I=1,100
      S=I
      ZETA=PI*Z(J)/L
      U(K,J)=U(K,J)+BNN(I)*KOR2(K,I)*SIN(S*ZETA)
203   CONTINUE
2002  CONTINUE
      WRITE(6,1105)
1105  FORMAT(' ***************************************************')
      WRITE(6,701)
701   FORMAT( 2X,'THE STEADY TEMPERATURE IN THE CALCINE BIN ')
      WRITE(6,999)
 999  FORMAT(' RADII(FT)         DEPTH=10     DEPTH=20     DEPTH=40     DEPTH
     1=55')
      WRITE(6,706) (R2(K),(U(K,J),J=1,4),K=1,15)
706   FORMAT(5(F12.2))
      WRITE(6,1106)
1106  FORMAT(' ***************************************************')
      GO TO 206
9999  STOP
      END
C     *************************************************************
C     ENO OF THE MAIN PROGRAM.                                    *
C     *************************************************************
C
C    **************************************************************
C     BESSEL FUNCTION SUBROUTINE FOR CALCULATING THE ROUTES OF THE  *
C     MODIFIED BESSEL FUNTIONS REQUIRED IN THE EVALUATION OF THE    *
C     ARBITRARY CONSTANTS.                                          *
C     *************************************************************
C     THIS SUBROUTINE IS REQUIRED TO COMPUTE I(0) AND K(0)          *
C     *************************************************************
      SUBROUTINE KIZERO (X,XK,XI)
    1 IF (X-3.75) 2,2,10
    2 A= (X/3.75)**2
    30XI = (((((0.0045813*A+0.0360768)*A+0.2659732)*A+1.2067492)*A+3.089
     19424)*A+3.5156229)*A+1.0
    4 IF(X-2.0)5,5,40
    5 A= (X/2.0)**2
    60XK= (((((7.40E-6*A+1.0750E-4)*A+0.00262698)*A+0.03488590)*A+0.22006
     19756)*A+0.42278420)*A-0.57721566-XI*ALOG(0.5*X)
    7 RETURN
   10 A=3.75/X
   110XI = ((((((((.3923767E-2*A-.1647632E-01)*A+.2635537E-01)*A-.2057
     170E-01)*A+.9162808E-02)*A-.1576490E-02)*A+.2253187E-02)*A+.13285
     29E-01)*A+ .3989422E+00
   12 B=-0.5
   13 IF(X-87.3)14,43,43
   14 XI=XI*X**B*EXP(X)
   40 A= 2.0/X
   410XK= ((((((5.3208E-4*A-2.51540E-3)*A+0.00587872)*A-0.01062446)*A+
     10.02189568)*A-0.078323580)*A+1.25331414)*EXP(-X)/X**0.5
   42 RETURN
   43 XI =XI*X**B/EXP(-X)
   44 GO TO 40
      END
C     *************************************************************
```

```
C   THIS SUBROUTINE  IS REQUIRED TO COMPUTE I(1)  AND K(1)*
C   ****************************************************************************
      SUBROUTINE KIONE(X,XK,XI)
    1 IF(X-3.75)2,2,8
    2 A=(X/3.75)**2
    30XI =    ((((((3.2411E-4*A+3.01532E-3)*A+2.658733E-2)*A+0.15084934)*
     1A+0.51498869)*A+0.87890594)*A+0.5)*X
    4 IF (X-2.0)5,5,10
    5 A=(X/2.0)**2
    60XK=    ((((((-4.686E-5*A-1.1040E-3)*A-1.919402E-2)*A-0.18156897)*A
     1-0.67278579)*A+0.15443144)*A+1.0)/X+ALOG(0.5*X)*XI
    7 RETURN
    8 A=3.75/X
    90XI=  ((((((((-4.200587E-3*A+1.787653E-2)*A-2.895312E-2)*A+2.282
     196E-2)*A-1.031555E-2)*A+1.638014E-3)*A-3.620183E-3)*A-3.988024
     2E-2)*A+.3989422E+00)/(X**0.5*EXP(-X))
   10 A=2.0/X
   110XK= ((((((-6.8245E-4*A+3.25614E-3)*A-7.80353E-3)*A+0.0150426)*A-.
     1365562E-01)*A+0.2349861)*A+1.2533141)*EXP(-X)/X**0.5
      RETURN
      ENO
$ENTRY
1 CASE-1 ANNULAR CONFIGURATION(TEMPERATURE AT DIFFERENT VERTICAL SECTION)
0.5,0.2,0.5
0.5,3.04
50.0,80.0
.1,.2,.3,.4,.5,1.,1.5,2.,2.5,3.04,4.,6.,8.,12.,16.
10.0,20.0 ,40.0,55.0
1 CASE-2 ANNULAR CONFIGURATION(TEMPERATURE AT DIFFERENT VERTICAL SECTION)
0.5,0.2,0.5
1.5,3.35
50.0,80.0
.1,.2,.3,.4,.5,1.,1.5,2.,2.5,3.04,4.,6.,8.,12.,16.
10.0,20.0 ,40.0,55.0
1 CASE-3 ANNULAR CONFIGURATION(TEMPERATURE AT DIFFERENT VERTICAL SECTION)
0.5,0.2,0.5
1.0,3.14
50.0,80.0
.1,.2,.3,.4,.5,1.,1.5,2.,2.5,3.04,4.,6.,8.,12.,16.
10.0,20.0 ,40.0,55.0
```

Heat Transfer and Modelling Studies for the Analysis of Waste Storage Facilities

by

Subbaratnam Ramachandran

B. Tech, Indian Institute of Technology, Madras, 1968

AN ABSTRACT OF A MASTER'S THESIS

submitted in partial fulfillment of the requirements for the degree of

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Kansas State University
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1985

Abstract

Dissipation of heat from first cycle wastes resulting from reprocessing of spent nuclear fuel is a problem when considering the extended storage times involved. The storage tanks for the high-level liquid wastes (aqueous raffinates from the first solvent extraction cycle) at the Hanford Reservations and the Savannah River Plant have experienced leaks, presumably due to the large amount of heat evolved in the wastes. Pursued in this research work is a brief account of the thermal histories of the tanks that had leaked and the remedials for safe storage of these wastes in future installations.

These aqueous wastes are converted into granular calcines at the Idaho National Engineering Laboratory, Idaho Falls, and are stored in the form of a 'dry calcine' in underground stainless steel bins to provide an additional barrier against leakage. The current storage facilities for these converted calcines, which are thermally hot, consist of bins cooled externally using forced circulation. Pursued in this study is the design of simplified storage facilities that are directly buried in soil, which takes advantage of the inherent mode of heat rejection through conduction. By doing so the need for external cooling is totally avoided, making such storage cost effective and safe from the leakage of radioactivity. An earlier model, undertaken for the heat transfer analysis for such simplified storages, pertained to a solid cylinder configuration and estimation of the resultant temperature profile inside the bin and near field effects in the surrounding soil. The model pursued in this research work assumes an annular geometry for the bin configuration. Mathematical formulations for the heat

conduction are arrived at using the Fourier heat conduction equation and the analytical solutions in the various regions evolved using separation of variables. The numerical examples simulating field conditions for the annular configuration show an overall reduction in the maximum storage temperature in the bin by 300°F for a practical size container, when compared with the solid configuration. Also pursued in the modelling studies are various parametric studies such ss the effect of heat generation rate of the stored wastes, the bin's geometry and the thermal conductivities of the solid wastes and the surrounding soil upon the resultant temperature profiles.