

MASTER**HEAT TRANSFER IN HIGH-LEVEL WASTE MANAGEMENT**

by

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ABSTRACT

Heat transfer in the storage of high-level liquid wastes, calcining of radioactive wastes, and storage of solidified wastes are discussed. Processing and storage experience at the Idaho Chemical Processing Plant are summarized for defense high-level wastes; heat transfer in power reactor high-level waste processing and storage is also discussed.

INTRODUCTION

This paper summarizes the major heat transfer considerations in the management of both defense and power reactor high-level wastes. Areas addressed include liquid waste storage, waste solidification using fluidized-bed calcination, and heat transfer in the engineered storage of typical waste forms. The defense wastes discussed herein are those generated in the processing of nuclear propulsion and research reactor fuels at the Idaho Chemical Processing Plant (ICPP).

Defense wastes generated at the ICPP are temporarily stored as an acid solution in cooled and uncooled stainless steel tanks within an underground concrete vault. Liquid wastes have been solidified using fluidized-bed calcination since 1963.³ The Waste Calcining Facility (WCF), originally built as a demonstrational facility, has been used to calcine approximately 13,000 m³ of liquid wastes.

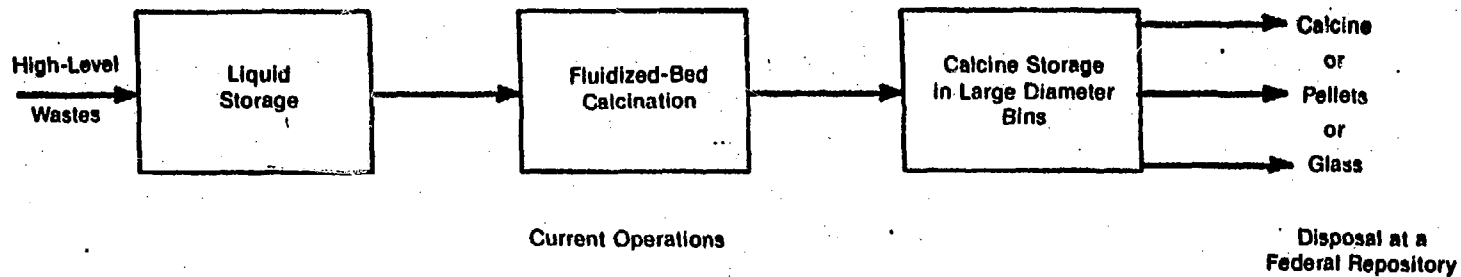
Calcine generated from defense waste is stored in stainless steel bins in underground concrete vaults. The first set of bins was filled during the period 1963-1964 and contains 210 m³ of alumina calcine. A second bin set was filled during the period 1966-1972 and contains 900 m³ of both alumina and zirconia calcine; a third bin set with a maximum capacity of 1100 m³, is currently being filled during operation of the WCF. Heat generation rates for calcine generated from defense wastes are in the range of 0.2 kW/m³ to 0.4 kW/m³.

Long-term management of calcined defense wastes at Idaho is based on options ranging from disposal as calcine to conversion of calcine to other waste forms (pellets and glass) for disposal at a federal repository.⁴ Conversion of calcine to other waste forms and transportation to an off-site federal repository would require that the waste be transported in canisters. The volumetric heat

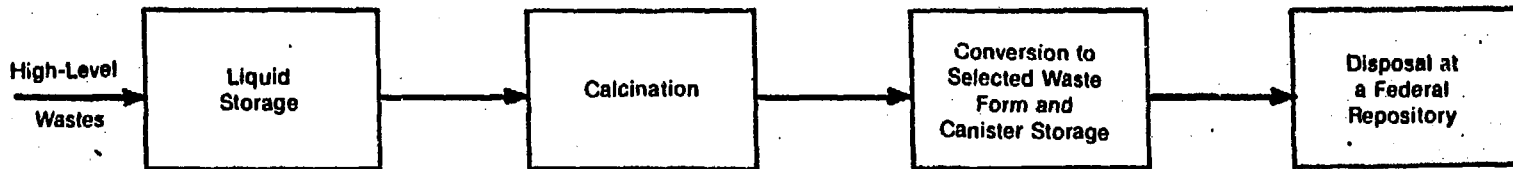
generation rate (200-400 W/m³) of ICPP defense waste calcine is sufficiently low to cause no major heat transfer concerns after canisterization.

Power reactor high-level liquid wastes, other than the 40 m³ of acid thorium wastes and 2300 m³ of neutralized liquid wastes stored at the Nuclear Fuel Services Plant in West Valley, New York, have not been generated in the United States. However, the concept for management of power reactor high-level liquid wastes, internationally, is essentially the same as that practiced routinely at the ICPP. Acid liquid wastes would be stored temporarily in stainless steel tanks inside an underground concrete vault; the liquid wastes would then be calcined (fluidized-bed, spray, or rotary kiln) prior to conversion to one of the candidate waste forms.

Calcine produced from power reactor wastes would have a volumetric heat generation rate in the range of 100-1000 times greater than calcine produced from the acidic defense wastes at the ICPP. Power reactor waste calcine would have a heat generation rate of 240 kW/m³ and 50 kW/m³ at 1 year and 5 years waste decay time, respectively. Liquid power reactor high-level wastes can be stored in stainless steel tanks similar in design to those now used at the ICPP. However, the high heat generation rate of power reactor waste calcine would make calcine storage in large diameter bins impractical. Canister storage or disposal of calcine or other waste forms would be required to keep temperatures below the maxima specified for canister walls or the particular waste form (calcine, glass, sintered glass ceramic, or metal matrix). A schematic diagram of the waste management systems used at the ICPP and a candidate system for power reactor high-level waste management is shown in Figure 1.



(a) Defense High-Level Waste Management at the ICPP



(b) Power Reactor High-Level Waste Management Option

FIGURE 1. SYSTEMS FOR MANAGEMENT OF DEFENSE AND POWER REACTOR HIGH-LEVEL WASTES

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LIQUID WASTE STORAGE

High-level liquid radioactive wastes, associated with nuclear fuel reprocessing, are typically stored on an interim basis (usually less than 5 years) to reduce heat generation rates due to radioactive decay prior to solidification. Present and anticipated ICPP wastes are acidic solutions primarily resulting from reprocessing aluminum, zirconium, and stainless steel clad fuel. High-level waste from mixed-oxide commercial fuel reprocessing would also be acidic, but would contain larger concentrations of fission products resulting in much higher heat generation rates (1000 fold greater) than ICPP wastes.

A typical liquid high-level waste storage system consists of storage tanks contained within concrete vaults, a cooling system, and an off-gas cleanup system. Decay heat is removed by heat exchanger coils submerged in the storage tanks. Cooling water which is pumped through the coils is cooled by a secondary heat exchanger. Vapors from the tank are refluxed by a condenser, diluted, and/or vented. Samplers are provided for the vault sump and tank contents. Liquid level is continuously monitored.

ICPP Defense Waste

High-level wastes at ICPP result from complete dissolution rather than leaching of fuel. The resulting wastes are stored in seven 1135 m³ stainless steel tanks, contained in concrete vaults, until they can be solidified in the WCF. The major chemicals present in the wastes are aluminum, zirconium, fluoride, and nitrate. The wastes are maintained acidic (1 to 3 molar in hydrogen ion) to reduce precipitate formation in the tank and to eliminate high sodium concentrations from neutralizing the waste with NaOH. High sodium content wastes cannot be calcined without further treatment.

The amount of heat generated by the wastes varies with the type of fuel, decay time before processing, and fuel burnup. The activity level in the ICPP high-level wastes varies from 1.0 to 40.0 Ci/L; ^{137}Cs , ^{134}Cs , ^{144}Ce , and ^{90}Sr are the principal nuclides. Cooling coils are provided for decay heat removal from high-level wastes to minimize tank corrosion and to prevent release of volatile fission products. Since fluoride-containing waste is very corrosive, its temperature during interim-storage is maintained below 35°C ; other wastes are maintained below 55°C .

Each storage tank has 30 cooling coils on the shell and/or floor of the tank; each coil has an individual inlet valve and a pressure relief valve. The outlet flow is recorded and the cooling water temperature and coil return header activity are monitored. The water is cooled indirectly in a heat exchanger, collected in a surge tank, and pumped back through the cooling coils, forming a closed system. Individual coils are valved in or out as necessary to maintain the tank contents below 55°C (35°C for fluoride waste). A schematic diagram of a typical ICPP cooled waste tank system is given in Figure 2.

The waste tanks are connected to reflux condensers which condense vapors. The stainless steel shell-and-tube condensers are equipped with sumps and jets for transferring condensate to a waste tank or to the plant waste evaporator. The condenser cooling water is normally shut off and the valve must be manually opened if the condensers are needed.

All high-level waste tanks are contained in individual concrete vaults, approximately 0.61 m (2 ft) thick. Vault roofs are located 3.1 m (10 ft) below grade and are covered with earth for radiation protection. Each vault has a sump to collect liquid entering the vault. The sumps have level indicators to detect tank or groundwater leakage into the vault.

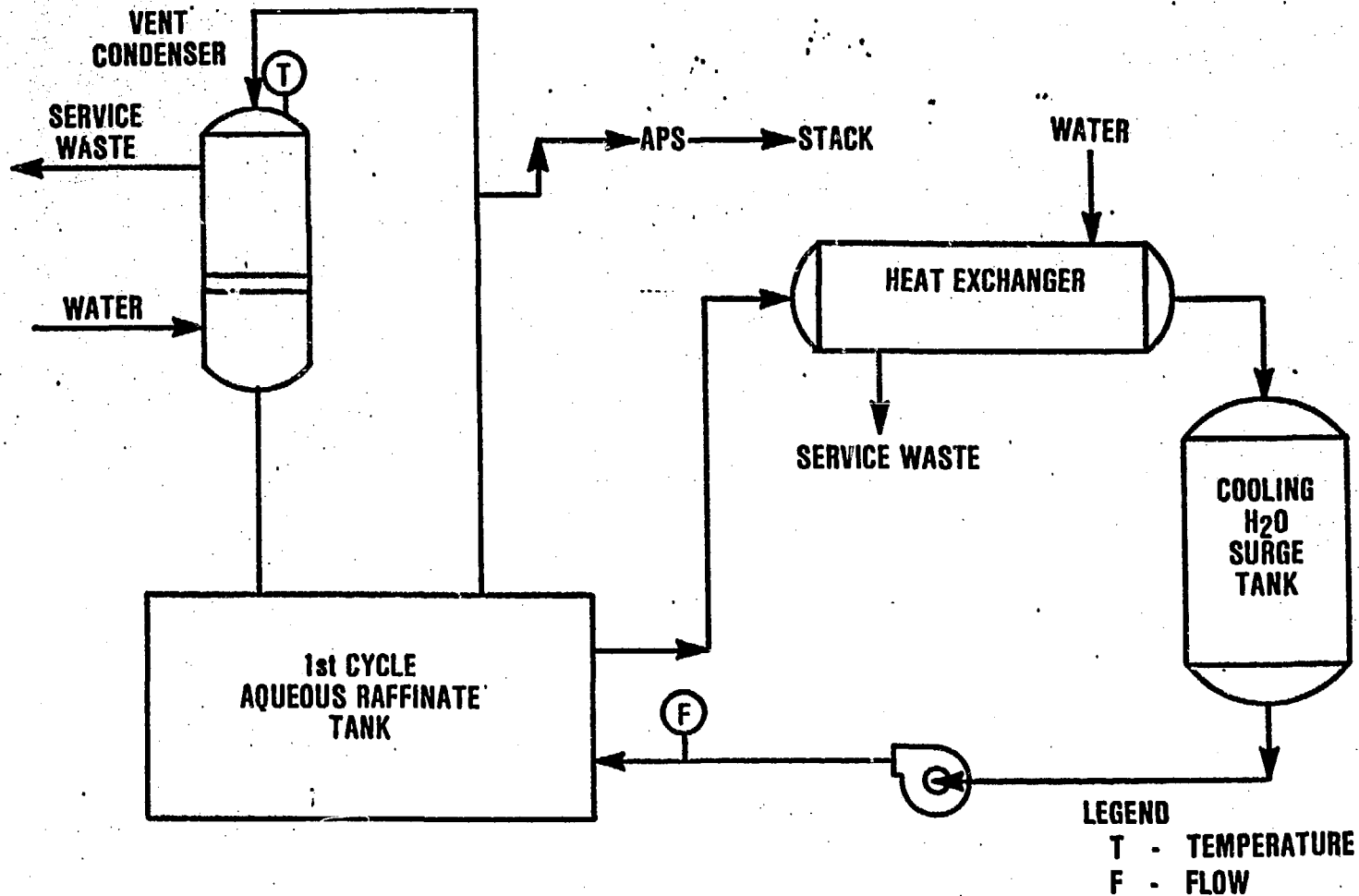


FIGURE 2. PROCESS SCHEMATIC OF AN ICPP WASTE TANK COOLING WATER SYSTEM

As a condition for fuel processing, at least one empty 1135 m³ cooled tank is reserved at ICPP to hold the contents of any storage tank should a tank failure occur. An operating spare is also required.

The volumetric heat generation rates of the wastes at ICPP are calculated from specific average energy releases, generally in Mev/dis, and measured activity in dis/s-mL. Design heat transfer considerations are summarized in Table I.

General operating experience has been:

1. No tank has ever leaked.
2. Measured corrosion rates on tanks ≤ 1.0 mil per year.
3. No cooling coil leaks have ever been detected.
4. Heat removal capacity of tanks has been conservative by at least a factor of 10.
5. Projected service life of tanks is 50 to 100 years.
6. No tank vapors are known to have been released to the stack through relief valves.

Power Reactor High-Level Wastes

Pertinent domestic high-level waste storage facilities currently are limited to those constructed but not in operation at the BNFP Separations Facility as described in their Safety Analysis Report. High-level waste at BNFP would be stored in 1135 m³ stainless steel tanks. Heat generation rates correspond to concentrations of 300 gallons of waste per metric ton of reprocessed fuel having a design burnup of 35,000 MWd/MTU fuel at a specific power of 40 MW/MTU and cooling time of 160 days. Longer cooled waste can be more concentrated. Design basis heat removal is 11.0×10^6 W per tank. Maximum interim storage time period before solidification is 5 years. The high-level tank systems include: 1) a decay heat removal system, 2) dilution air for tank atmosphere, 3) a ballast

TABLE I

WASTE TANK DESIGN HEAT TRANSFER CONSIDERATIONS

Waste Activity:	1.0 to 40 Ci/L
Principal Nuclides:	^{137}Cs , ^{134}Cs , ^{144}Ce , and ^{90}Sr
Maximum Heat Generation Rate:	1.22 kW/m ³
Recirculating Water Velocity	= 1.6 m/sec
Inside Film Coefficient	= 6,101 W/m ² -°C (1075 Btu/hr ft ² °F)
Outside Film Coefficient	= 624 W/m ² -°C (110 Btu/hr ft ² °F)
Resistance	
Inside Fouling	0.0004 m ² -°C/W
Outside Fouling	0.0012 m ² -°C/W
Metal	0.0002 m ² -°C/W
LMTD	32.0 °C
U _o	273 W/m ² -°C (48.1 Btu/hr ft ² °F)
Required Heat Transfer Area	— 157 m ²
No. Coils Required	— 17 coils
Average Coil Length	— 60 m

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tank system to minimize settling of solids, and 4) instruments for liquid level, specific gravity, tank pressure, and thermocouples. Corrosion allowance for the tanks is 1/8 inch. Expected corrosion rate is less than 0.1 mil/year.

The decay heat removal system consists of 48 two-inch schedule 80 stainless steel coils. The coils average 740 feet in length. The coils are arranged in six banks wound helically through the tank. Any one header can be valved out. The tank is contained in a stainless steel lined vault.

WASTE CALCINATION

Process Heat

Since 1963 the Waste Calciner Facility (WCF) located at the ICPP has converted over 13,000 m³ of high-level radioactive liquid waste to 1900 m³ of calcined solids at a net feed rate of over 360 L/hr. Initially the heat required for calcination of these wastes was provided by a circulating liquid-NaK system which transferred heat from an oil-fired furnace to a fluidized-bed of solids contained in the calciner vessel.

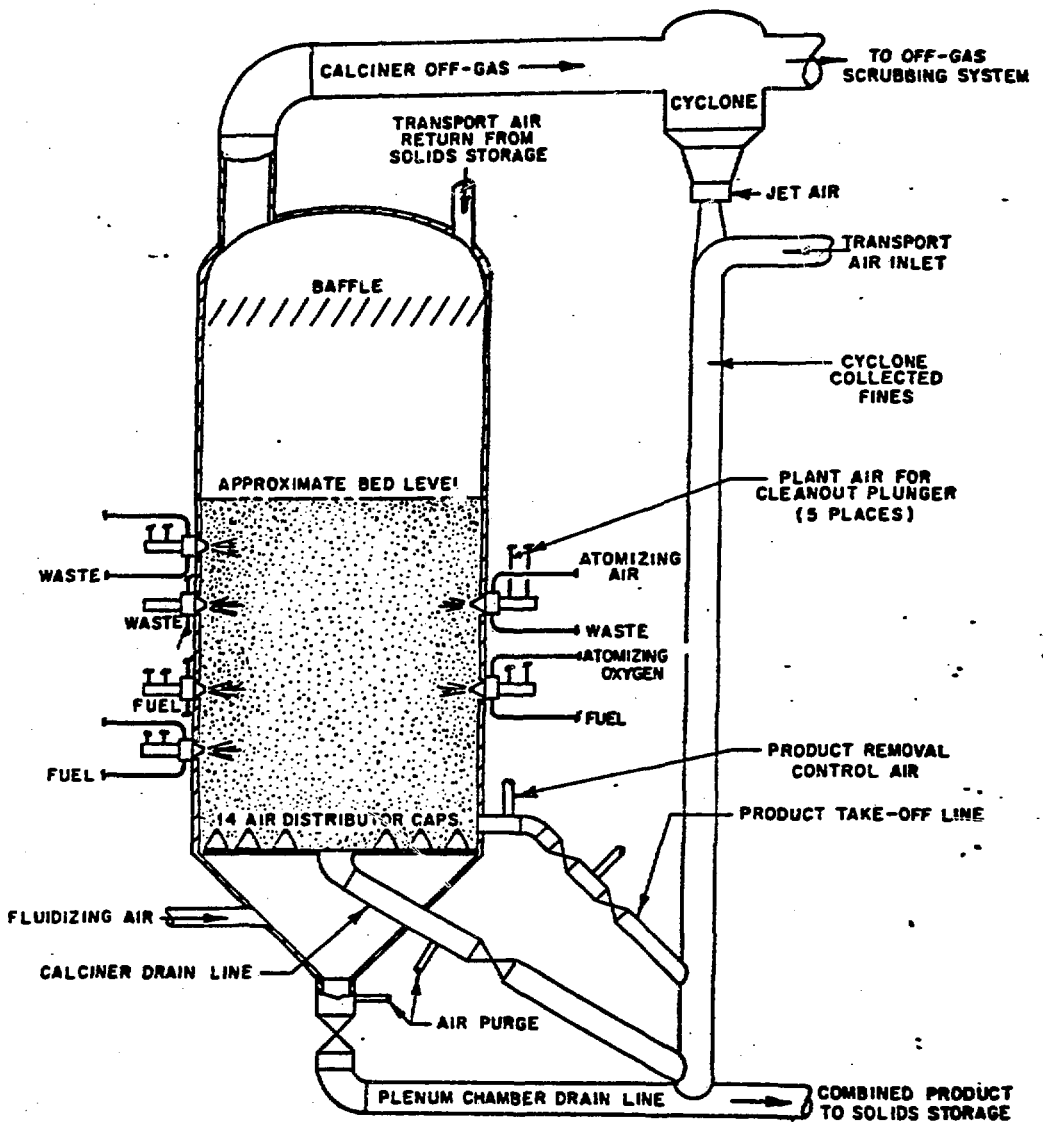
To improve the WCF reliability and provide for increased waste feed capacity, an in-bed combustion system was installed in 1970 as a replacement for the NaK heating system. (In-bed combustion is the injection of a combustible liquid directly into a fluidized-bed to furnish the heat requirements of the fluidized-bed process.) Adaptation of the WCF to in-bed combustion heating required minimum modifications and capital expenditure relative to alternative systems, and the system has many inherent advantages.⁵

The in-bed combustion system for the WCF provides sufficient heat to calcine up to 400 L/hr of high-level radioactive liquid waste

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 given in Figure 3. During operation, kerosene is sprayed through three external atomizing nozzles into the hot fluidized-bed where the fuel burns, releasing heat and producing carbon dioxide and water vapor plus a small amount of carbon monoxide. Oxygen required for the fuel combustion is provided by: 1) preheated fluidizing air, 2) pure oxygen used as the fuel-nozzle atomizing gas, and 3) nitrates in the liquid waste. Heat requirements of this calcining process are set by the rate and type of liquid waste being fed to the calciner.

Primary advantages of in-bed combustion relative to indirect heating are: 1) maximum heat transfer efficiency and utilization, 2) very suitable for high-temperature operation, and 3) a relatively simple installation. Improved heat transfer and utilization allows greater waste throughput with the same sized calciner at minimum metal wall temperatures. Operation at 500°C bed temperatures rather than 400°C as with indirect heating resulted in reduction of ruthenium volatility by more than 1000 fold. A significant secondary advantage of in-bed combustion is more nitrates are decomposed to H_2 and O_2 rather than NO_2 and NO .

Disadvantages of in-bed combustion relative to indirect heating are: 1) the high velocity flame from the fuel nozzle tends to cause a high particle attrition, resulting in difficulty in maintaining bed level, 2) combustion efficiencies greater than 92% cannot be regularly achieved by in-bed combustion, hence light hydrocarbons are produced which can cause plugging in off-gas filters, 3) larger off-gas systems are required with in-bed combustion to handle the additional combustion gas, and 4) high flame temperature causes melting of bed particles in some flowsheets (particularly high sodium content) tested in the pilot plant.



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Figure 3. WCF Calciner Vessel with In-Bed Combustion

Hydrocarbon fuel burns readily in a fluidized-bed at temperatures greater than 335°C in the presence of a nitrate-oxygen atmosphere. Therefore, normal operations at 500°C present no particular hazard. Automatic fuel and oxygen shutoff valves, temperature alarms, and other instruments are provided as part of the safety interlock system. Eight years of successful operation with in-bed combustion have demonstrated the safety and efficiency of the process. The use of in-bed combustion is directly applicable to calcining wastes from reprocessed power reactor fuel.

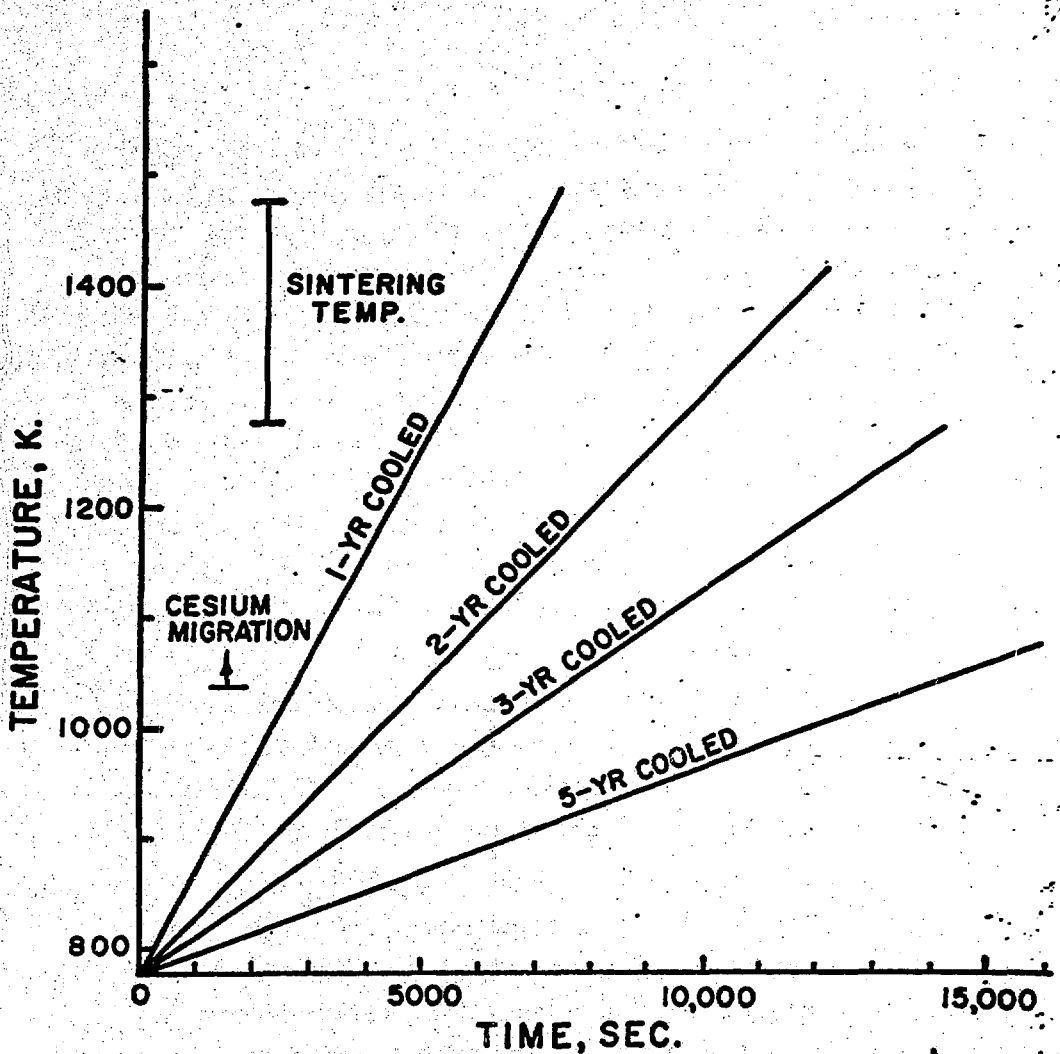
Collapsed Bed

A consideration that must be addressed in the fluidized-bed calcination of the high-heat-generation reprocessing wastes is the overheating of a collapsed bed with a portion of the bed sintering or melting following an accidental loss of fluidizing air.⁶ The most likely cause of a loss of fluidization in the calciner is loss of fluidizing air due to a blower failure or loss of electric power. A fluidized bed remains fluidized so long as adequate fluidizing air is supplied; a bed collapse occurs if fluidizing air is lost. Protection against loss of fluidization in the WCF is provided by spare blowers and a back-up power supply. The loss of fluidizing air is a relatively rare event. The existing WCF has averaged about one bed collapse every two years as a result of a commercial power outage. Fluidization was normally restored in less than a minute by activation of the emergency power system. One (the longest) bed collapse of an hour's duration occurred when the emergency-power system failed to respond following a power outage.

An uncorrected loss-of-fluidization of the calciner bed when processing power reactor wastes would result in a serious operating problem rather than a serious hazard to the public. The solids in the calciner would heat slowly, if no corrective action were taken, with sintering and possibly melting occurring in the center of the bed after several hours. The calciner wall, if uninsulated, would not melt through. Upon reaching steady-state -- i.e., maximum temperatures -- the collapsed bed would contain possibly a molten core surrounded by sintered calcine surrounded in turn by unsintered calcine, all contained within an intact vessel. Fission products, volatilized from the calcine, would be released only through the calciner off-gas system which would hold the quantity released to allowable values.

A quantitative appraisal of potential consequences of an uncorrected loss of fluidization depends in part on the waste age and burnup and on the reprocessing flowsheet. The subsequent analyses are based on full burnup fuel (33,000 MWD/MTU).

The calcine temperature increase following a loss of fluidization would be sufficiently slow to allow ample time for corrective action. Figure 4 shows maximum temperatures for calcines of different ages in an uncooled bed (the slopes are a ratio of decay-heat generation to heat capacity). Depending on waste age, there would be 0.7 to 4 hours available before any of the solids reached the lowest temperature -- 1025°K -- at which fission products begin to migrate (initially from hot to cooler zones) and at least 1.4 to 8 hours before any sintering would occur (the sintering temperature is somewhere between 1275°K and 1475°K). The automatic protection systems and the operators could certainly respond in these time intervals.



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Figure 4. Temperature Increase in a Collapsed Calciner Bed.

The basic assumptions in Figure 4 are:

- (1) The decay-heat generation per kg uranium is taken from ORNL-4422, Fig. 4.5 for 33,000 MWD/MTU burn-up.
- (2) The high-level waste ratio is 85 kg solids/MTU.
- (3) The calciner operates at 773°K.
- (4) The heat capacity of the solids is 1000 Joule/kg-K.

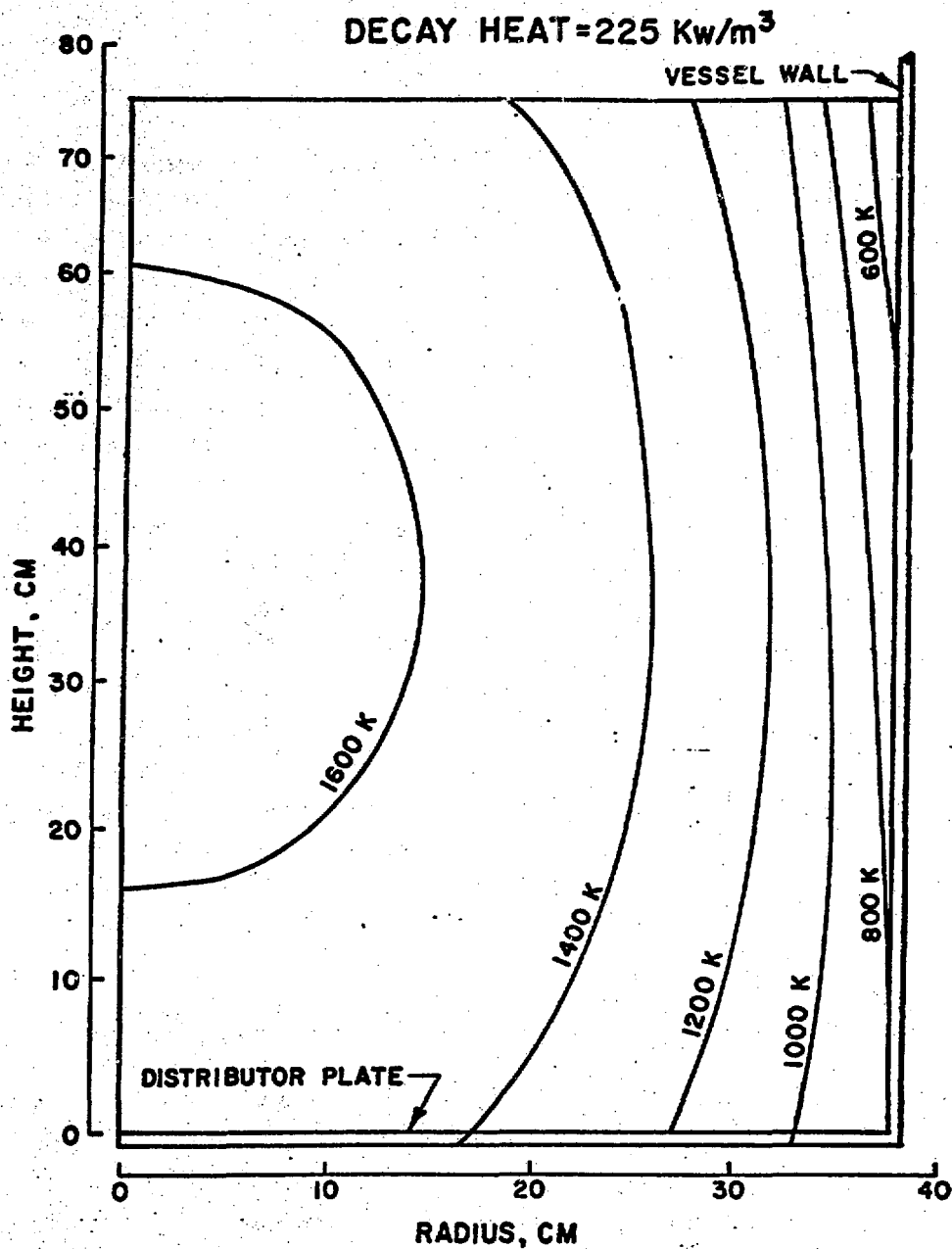
In the event of an uncorrected loss of fluidization, the overheated (and possibly with a molten core) solids would be retained within the calciner vessel provided the vessel is installed with an uninsulated outer wall to allow dissipation of the decay heat. A bare vessel wall can dissipate, by convection and thermal radiation, the decay-heat from a collapsed bed without reaching temperatures that would damage the wall. Figure 5 shows calculated temperature isotherms for postulated collapsed beds of calcine with postulated "maximum" decay-heat. The calciner dissipates the decay-heat by radiation and convection through the walls and by radiation from the grid plate and upper surface to the lower and upper plenums, respectively. Table II summarizes the maximum calculated wall and grid-plate temperatures. The maximum wall temperature -- 850°K -- is within the operating limits of stainless steels. The calculated grid-plate temperatures vary from the wall temperature -- 850°K -- at the edge to 1525°K at the center. With older wastes, the temperatures would be lower. A grid-plate fabricated of, or reinforced with, a high-temperature metal -- e.g., tantalum -- would remain intact in event of the postulated loss-of-fluidization.

Table II

Calciner Vessel Conditions for a Collapsed Bed

Calcine age (out of reactor), yr	1.0	1.8
Solids yield, m ³ /MTU	0.038	0.062
Decay heat, kW/m ³	225	103
Maximum wall temperature, K	850	710
Maximum grid plate temperature, K	1525	1250

The use of an uninsulated calciner vessel is recommended (unless only old waste is solidified) to assure that the vessel cannot melt through in case of an uncorrected loss of calciner fluidization.



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FIGURE 5. CALCULATED TEMPERATURE ISOTHERMS IN A COLLAPSED CALCINER BED WITH A DECAY-HEAT GENERATION OF 225 KW/m³

With the overheated calciner bed confined within the calciner vessel, the primary impact of an uncorrected loss-of-fluidization would be operational. The sintered or molten core of the calciner would be difficult to remove. Restoration of the calciner to operation following the postulated incident would require either 1) removal and disposal of the entire calciner vessel with its sintered bed, or 2) dissolution of the hot bed. Both would be difficult.

The protective systems proposed to assure that a loss-of-fluidization does not result in a sintered bed are 1) a set of automatic, redundant, fluidizing-air systems, and 2) a set of operator-controlled, redundant, bed-removal systems. The fluidizing air systems would be the primary protective systems; the fluidizing air will remove the decay heat and keep the calciner bed in a fluidized state which would allow the operator at his discretion to either resume operation or remove the bed. As shown in Figure 4, a rapid response to a loss of fluidization is not necessary; however a reliable response is needed to avoid the possibility of an inoperable calciner. The bed removal systems would be shut-down systems.

The severity of a collapsed fluidized bed processing ICPP defense wastes would generally be much less than for a bed of commercial fuel because heat generation rates are much less. A molten inner core would not occur when processing ICPP wastes.

HEAT TRANSFER FROM SOLIDIFIED WASTE FORM

ICPP Calcine Storage

Early defense wastes calcined in the WCF consisted essentially of a mixture of aluminum nitrate and fission products. The first set of bins was designed to maintain the temperature of the calcined solids below the 400°C calcination temperature (using an in-bed heat exchanger) to prevent the possible evolution of volatile fission

products.¹ A plan view of the first bin set is shown in Figure 6. An isometric drawing of the first bin set is shown in Figure 7.

Calcined solids were transferred pneumatically from the calciner vessel to a cyclone located over the top of the bins; calcine fell 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, is approximately 200 m³.

The second and third calcined solids storage facilities consist of seven 3.7 m diameter cylindrical bins, also enclosed in a reinforced concrete vault. The second and third set of calcined solids storage facilities differ primarily in the height of the bins. Capacity of the second solids storage facility is approximately 900 m³ and was filled in 1972; the third set of bins now being filled has a capacity of 1140 m³ and currently contains 640 m³. An isometric drawing of the second and third bin sets is also shown in Figure 7. The fourth bin set is similar to the second and third bin sets, although the capacity is only 500 m³.

Beginning with the fifth bin set, future bins will be built on the basis of an annular configuration because of the projected increase in volumetric heat generation rate and decreased calcine sintering temperature.

Calcine Cooling

The first bin set was designed for cooling by forced air convection; however, decay heat was removed by 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 walls and the concrete vault. The first bin set was designed on the basis of a calcine volumetric heat generation rate of 1200 W/m³. Calcine placed in Bin Set 1 had an average heat generation rate of

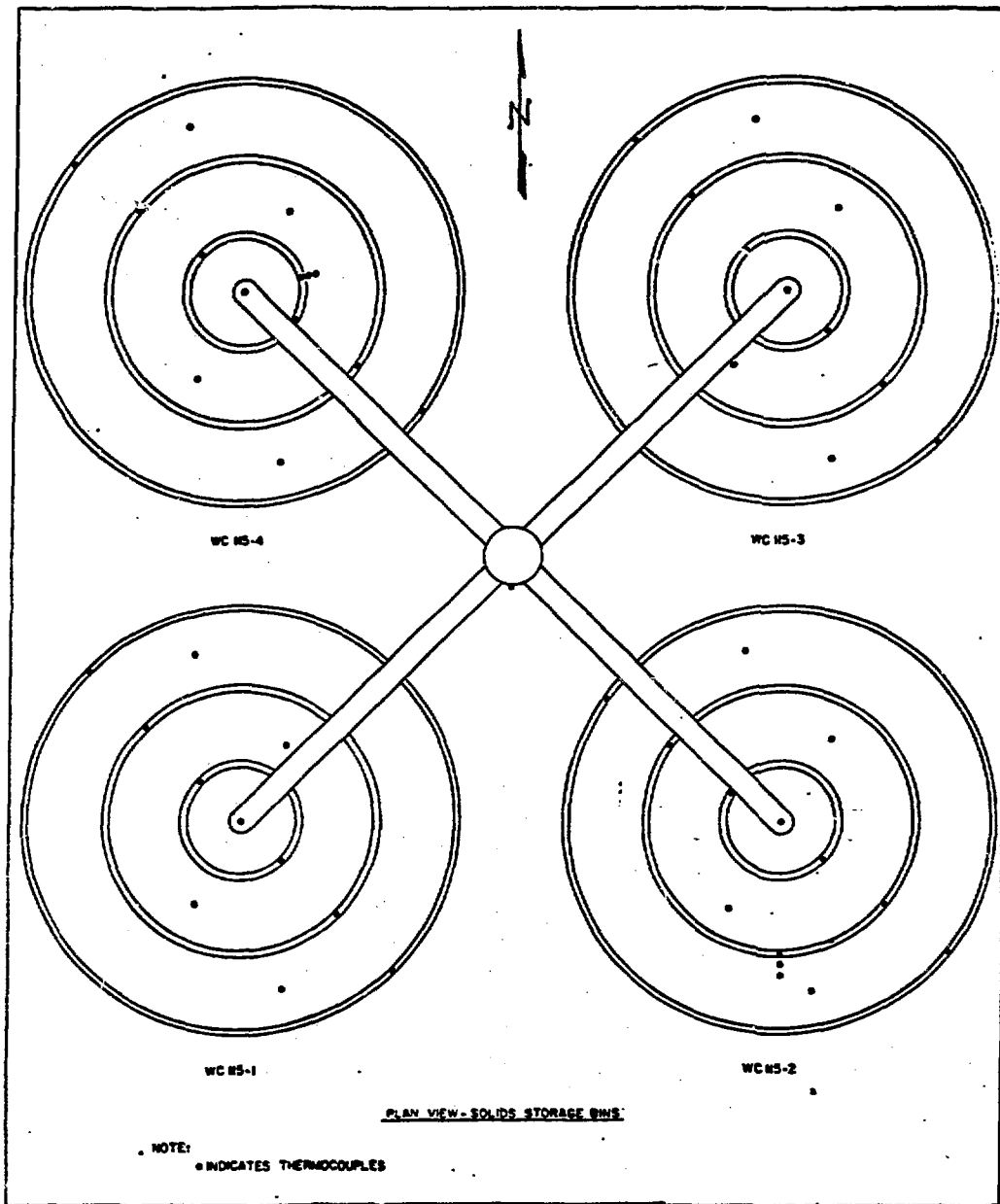


FIGURE 6. PLAN VIEW OF FIRST BIN SET

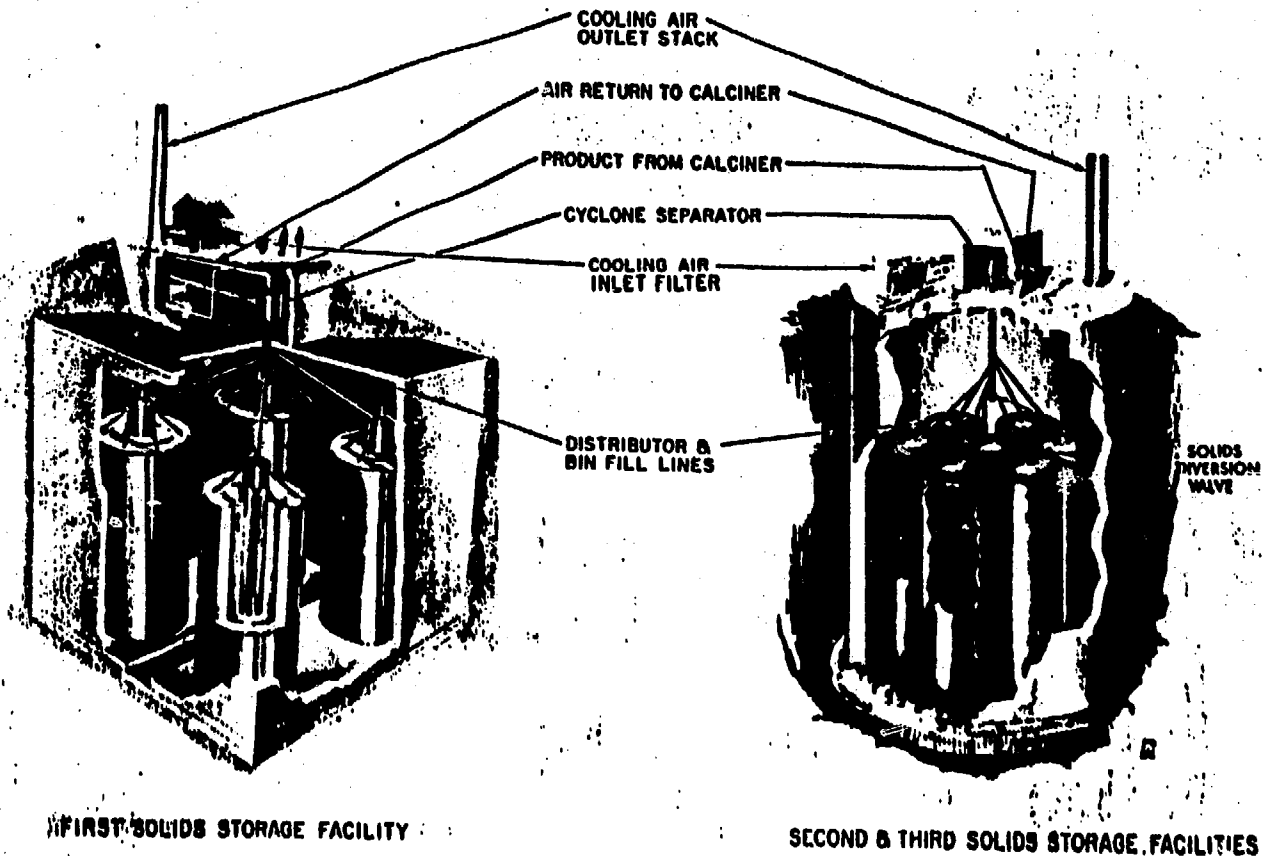


FIGURE 7. CALCINE STORAGE FACILITIES AT THE ICPP

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300 W/m³. The highest measured calcine centerline 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/m³. Cooling air was shut off for a 12-month period beginning in 1969. Heat removal was then primarily by convection to the vault air and a combination of natural convective and radiative transfer, respectively, from the vault air and bin walls to the vault wall. Heat was eventually dissipated to the surrounding soil. The maximum calcine temperature measured during the test in 1969 was 695°C.

The Third Bin Set contains calcine with an average heat generation rate of 50 W/m³; maximum calcine temperature measured to date has been 300°C. The Fourth Bin Set is not scheduled for accepting calcine until 1982; the expected heat generation rate is 100 W/m³. The fifth and future bin sets will be required to accept calcine with a heat generation rate in the range 400-500 W/m³. Maximum calcine temperature in the fourth and subsequent bin sets will be limited to 600-650°C to prevent sintering of calcine containing a mixture of sodium and zirconium. Basic heat transfer design equations and properties significant to heat transfer in defense waste calcine storage are shown in Table III. Design basis calcine heat generation rates and operating experience to date are summarized in Figure 8 and Table IV, respectively.

Storage of Solidified Power Reactor Waste Forms

High-level liquid wastes from the processing of Light Water Reactor (LWR) fuels would require solidification based on safety, regulatory, and economic considerations. While the liquid to calcine volume reduction factor for ICPP defense waste ranges from 7-8 the liquid to calcine volume reduction factor for LWR high-level wastes would range from 10-20. Conversion to other waste forms would result

TABLE III

HEAT TRANSFER CONSIDERATIONS IN CALCINED SOLIDS STORAGE

Basic Heat Transfer Design Equation For Cylindrical Bins:

$$\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial^2 T}{\partial r} + \frac{\partial^2 T}{\partial z^2} + \frac{Q(t)}{K_c} = \frac{1}{\alpha} \frac{\partial T}{\partial t}$$

For Storage Bins @ ICPP:

$$\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial^2 T}{\partial r} + \frac{\partial^2 T}{\partial z^2} + \frac{Q_0}{K_c} = 0$$

Calcine Properties Significant to Bin Storage:

Thermal conductivity, W/m-°C	0.19-0.35
Storage bulk density, g/cc	Zr 1.55-1.7
<u>Sintering temperatures</u>	<u>°C</u>
Zr-Fluoride calcine	700
Alumina calcine	1200
Zr-Sodium calcine	650

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FIGURE 8

CALCINED SOLIDS STORAGE AT THE ICPP

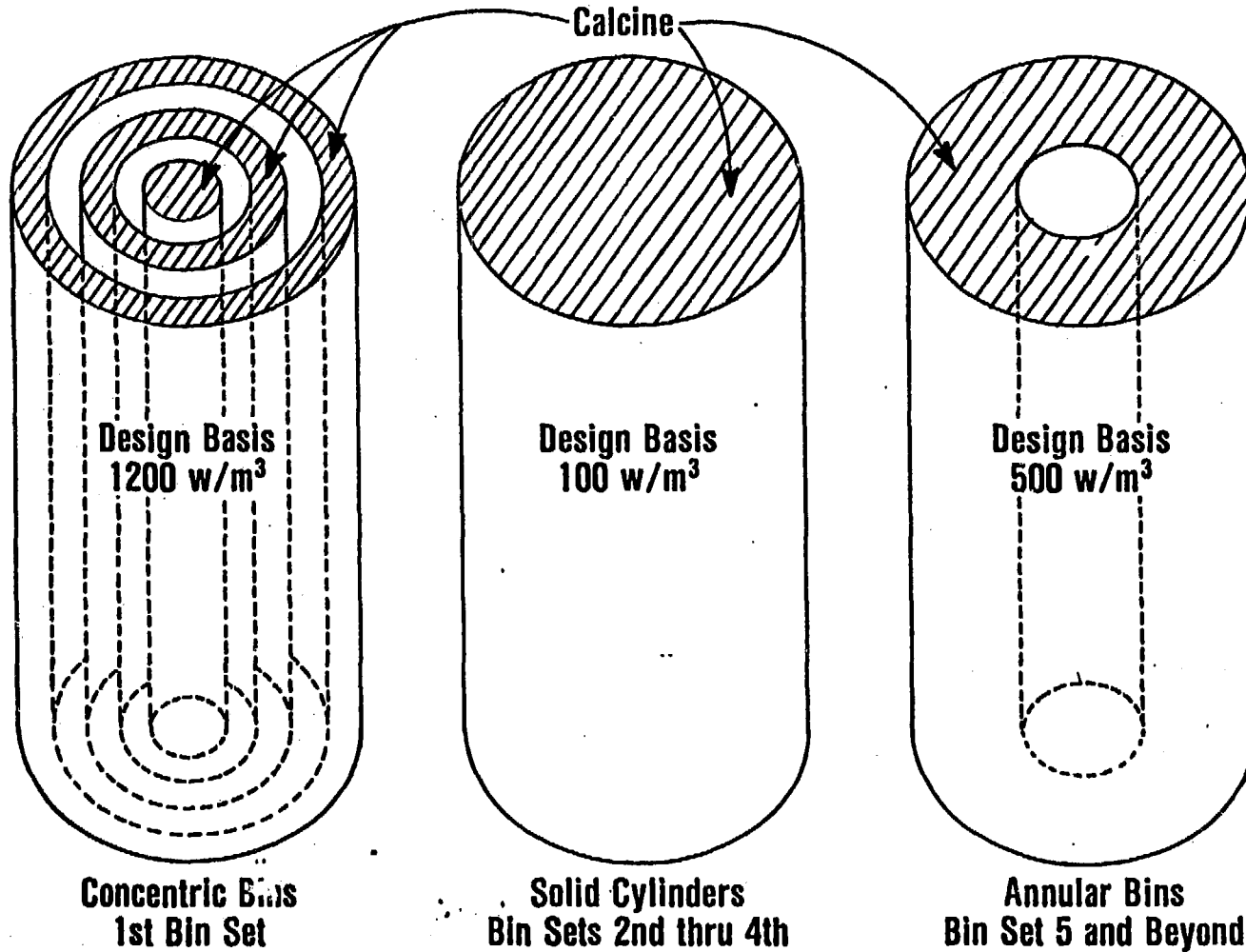


TABLE IV

CALCINED SOLIDS STORAGE EXPERIENCE AT THE ICPP

	Calcined Solids Bin Set		
	<u>1</u>	<u>2</u>	<u>3</u>
Calcine heat generation rate, w/m ³	300	250	50
Measured maximum calcine temp., °C	185	695	300
Maximum measured bin wall temp., °C	— —	85	55

Special Tests

- (1) Core samples from 2 bins of Bin Set No. 2 in 1978 revealed no calcine sintering.
- (2) Cooling air in Bin Set No. 2 was shut off for 12 months beginning in 1969; maximum calcined solids and bin wall temperatures reached 695 and 85 °C, respectively.

in volume reduction factors ranging from 6-10 and 10-15 for glass and sintered glass ceramic, respectively. Since a metal matrix waste form is essentially based on filling the voids within calcine with a metal, the volume reduction factor for a metal matrix is essentially the same as that for calcine.

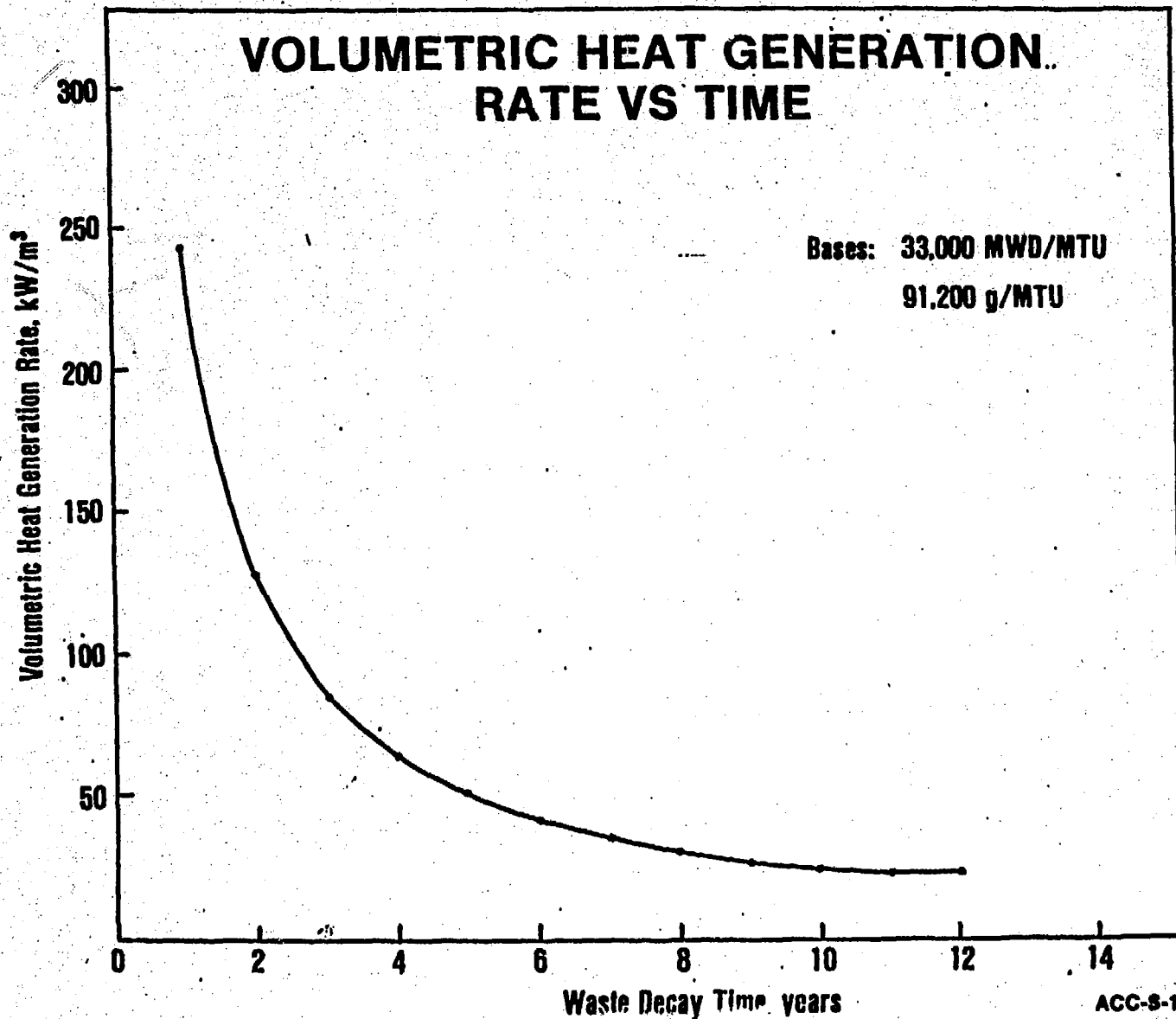
Engineered storage or transportation to a federal repository of any waste form will require that heat dissipation be consistent with the specified maximum product temperature for a given waste form or the maximum specified canister wall temperature. The only waste forms considered herein are calcine, glass, sintered glass ceramic (SGC), and metal matrix; these waste forms are representative of properties significant to heat transfer during canister storage. The reference high-level waste is that produced from a 1500 MTU/y LWR fuel reprocessing plant.

The volumetric heat generation rate of power reactor high-level wastes is significantly higher than defense high-level wastes, since the wastes are largely a mixture of fission product oxides. Approximately 90% of the decrease in the rate of decay heat occurs during the first 5 years out of the reactor. Volumetric heat generation rates for a typical calcine generated from LWR high-level wastes are shown in Figure 9.

Representative waste forms for high-level wastes are calcine, glasses or sintered glass ceramics, and metal matrices. Calcines are produced at processing temperatures from 350-600°C; glasses and SGC's are made in the temperature range of 950-1150°C; metal matrices considered herein would be based on filling the interparticle void fractions with a molten metal.

Calcine should be stored at a maximum temperature of 700°C to prevent significant volatility of ruthenium and cesium. Borosilicate glass will flow under its own weight at 550°C, and this is assumed

FIGURE 9. HEAT GENERATION RATE FOR POWER REACTOR HIGH-LEVEL WASTE CALCINE



to be the maximum storage temperature for glass. Sintered glass ceramics remain stable in a solid form up to 800°C. Metal matrices are stable at temperatures below 700°C (calcine maximum temperature) but are limited to temperatures below the melting point of the metal used.

Calculational Bases for Heat Transfer

Canisters for engineered storage or transportation to a federal repository must be of a practical size for handling. Concepts for canister configuration have been based on cylindrical canisters with a maximum outer diameter of 61 cm and a maximum length of 4.6 meters. Two basic configurations are considered in this paper, unfinned solid cylinders and annular cylinders. Nominal canister dimensions and waste volumes are shown in Table V;² properties of typical waste forms having significant impact on heat transfer are shown in Table VI. Calcine was assumed diluted with inert material to reduce the volumetric heat generation rate in those cases where the calcine temperature or canister wall temperature would exceed the specified maximum at the time the canister is filled. For glass and sintered glass ceramics, the waste content was adjusted to ensure satisfying the product and canister wall temperature constraints at the time of canister filling. A maximum canister wall temperature of 300°C was assumed based on stress corrosion of stainless steel at elevated temperatures for long periods of time.

Resistance to heat transfer through the cylinder walls was assumed negligible, and the thermal conductivity was assumed independent of temperature. Since the change in decay heat generation rate at a specified time is much smaller than the change in temperature as a function of the decreasing heat generation rate,

TABLE V

CANISTER CONFIGURATIONS AND WASTE VOLUMES

<u>Canister Configuration</u>	<u>Length, cm</u>	<u>I.D., cm</u>	<u>O.D., cm</u>	<u>Vol., m³</u>	
Solid Cylinder	305	—	20	0.1	
			25	0.16	
			30	0.22	
			35.5	0.30	
Annular Cylinder	305	25	x	46	0.34
		30			0.28
		30	x	61	0.59
		40.5			0.49
		46			0.39
		51			0.27

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TABLE VI

TYPICAL PROPERTIES OF POWER REACTOR HIGH-LEVEL WASTE FORMS SIGNIFICANT TO HEAT TRANSFER

Property	Calcine	Glass	SGC ⁽¹⁾	Metal Matrix
Waste in waste form, wt%	100	25-35	50-67	55-75
Density, g/cc	2.0-2.3	3.0-3.4	2.4-3.0	2.5-5.0
Thermal conductivity, w/m-°C	0.17-0.29	0.86-1.2	0.6-0.8	5.0-35.0
Waste form volume, m ³ /MTU	0.038-0.046	0.086-0.12	0.05-0.076	0.04-0.05
Maximum permissible waste form temp., °C	700	550	800	Pb-300(2) Al-600

(1) Sintered Glass - Ceramic

(2) Maximum waste form temperature limited by melting point of metal

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calculations were based on a "pseudo steady state". For calculational purposes, infinitely long cylinders were also assumed.

Calculated Temperatures and Permissible Heat Generation Rates

If wastes are placed in canisters at short decay times, product and wall temperature decrease significantly in the first two years. In general, product and canister wall temperatures for canisters loaded at long decay times remain near the original loading temperature for many years. The maximum permissible product (T_m) or canister wall temperature (T_w) limits the volumetric heat generation rate of the waste at time of loading.

Air Cooling

The heat generation rate for low thermal conductivity calcines is dictated by the maximum product temperature. Wastes stored in an annular canister are limited by the canister wall temperature. Since fluid-bed calcine has a low volume per MTU processed, dilution with an inert material does not necessarily result in waste volumes greater than other waste forms. An annular canister is a practical means of storing calcines using air cooling, because the canister can contain practical volumes of waste without exceeding the maximum calcine temperature. Glass, sintered, or metal matrix waste forms placed in solid cylinders are limited in heat generation rate because of the maximum canister wall temperature. However, lead metal matrices may be limited by the melting point of lead. Product temperature, canister wall temperatures, and heat generation rates for various waste forms placed in air-cooled cylinders are summarized in Tables VII and VIII.

TABLE VII

CALCULATED TEMPERATURES AND HEAT GENERATION RATES
USING AIR-COOLED SOLID CYLINDERS

Product	k W/m-°C	Diam. cm	T _w °C	T _m °C	Q ₃ kW/m
CALCINES	0.173	20	245	695	31
		25	210	695	21
		30	195	695	15
		35.5	180	695	11
	0.242	20	265	700	41
		25	240	700	27
		30	230	700	20
		35.5	210	700	15
SGC	0.692	20	300	475	48
		25	300	525	37
		30	300	565	31
		35.5	300	595	26
GLASS	1.038	20	300	420	50
		25	300	450	35
		30	300	475	30
		35.5	300	495	26
METAL MATRIX Pb	8.65	25	300	320	37
		30	300	320	33
		35.5	297	320	25
		40.5	293	320	21
		61	281	320	13
Al	35.0	25	300	305	37
		30	300	305	31
		35.5	300	305	26
		40.5	300	310	23
		61	300	310	15

TABLE VIII

CALCULATED TEMPERATURES AND HEAT GENERATION RATES
USING AIR-COOLED ANNULUS CYLINDERS

Product	k W/m-°C	I.D. cm	O.D. cm	T _{iw} °C	T _{ow} °C	T _m °C	Q kW/m ³
CALCINES	0.173	51	61	300	294	452	83
		46	61	300	291	527	55
		30	46	300	286	521	54
		25	46	300	277	578	39
	0.242	51	61	300	294	408	83
		46	61	300	291	461	55
		41	61	300	286	510	41
		35.5	61	300	277	548	31
30		46	300	286	455	54	
25		46	300	278	496	39	
SGC	0.692	51	61	300	296	336	83
		46	61	300	293	354	55
		30	46	300	289	350	54
		25	46	300	280	360	39
GLASS	1.038	51	61	300	296	323	83
		46	61	300	294	335	55
		30	46	300	290	331	54
		25	46	300	291	347	42
METAL MATRIX Pb	8.64	51	61	300	299	304	85
		46	61	300	299	307	57
		30	46	300	299	306	57
		25	46	300	298	306	43
Al	35.0	51	61	300	300	301	85
		46	61	300	300	301	57
		30	46	300	300	301	56
		25	46	300	300	301	43

TABLE VIII

CALCULATED TEMPERATURES AND HEAT GENERATION RATES
USING AIR-COOLED ANNULUS CYLINDERS

Product	k w/m-°C	I.D. cm	O.D. cm	T _{iw} °C	T _{ow} °C	T _m °C	Q kW/m ³
CALCINES	0.173	51	61	300	294	452	83
		46	61	300	291	527	55
		30	46	300	286	521	54
		25	46	300	277	578	39
	0.242	51	61	300	294	408	83
		46	61	300	291	461	55
		41	61	300	286	510	41
		35.5	61	300	277	548	31
30		46	300	286	455	54	
25		46	300	278	496	39	
SGC	0.692	51	61	300	296	336	83
		46	61	300	293	354	55
		30	46	300	289	350	54
		25	46	300	280	360	39
GLASS	1.038	51	61	300	296	323	83
		46	61	300	294	335	55
		30	46	300	290	331	54
		25	46	300	291	347	42
METAL MATRIX Pb	8.64	51	61	300	299	304	85
		46	61	300	299	307	57
		30	46	300	299	306	57
		25	46	300	298	306	43
Al	35.0	51	61	300	300	301	85
		46	61	300	300	301	57
		30	46	300	300	301	56
		25	46	300	300	301	43

TABLE VIII

CALCULATED TEMPERATURES AND HEAT GENERATION RATES
USING AIR-COOLED ANNULUS CYLINDERS

Product	k w/m-°C	I.D. cm	O.D. cm	T _{iw} °C	T _{ow} °C	T _m °C	Q kW/m ³
CALCINES	0.173	51	61	300	294	452	83
		46	61	300	291	527	55
		30	46	300	286	521	54
		25	46	300	277	578	39
	0.242	51	61	300	294	408	83
		46	61	300	291	461	55
		41	61	300	286	510	41
		35.5	61	300	277	548	31
		30	46	300	286	455	54
		25	46	300	278	496	39
SGC	0.692	51	61	300	296	336	83
		46	61	300	293	354	55
		30	46	300	289	350	54
		25	46	300	280	360	39
GLASS	1.038	51	61	300	296	323	83
		46	61	300	294	335	55
		30	46	300	290	331	54
		25	46	300	291	347	42
METAL MATRIX Pb	8.64	51	61	300	299	304	85
		46	61	300	299	307	57
		30	46	300	299	306	57
		25	46	300	298	306	43
Al	35.0	51	61	300	300	301	85
		46	61	300	300	301	57
		30	46	300	300	301	56
		25	46	300	300	301	43

Water Cooling

Wastes at shorter decay times can be stored using water cooling. Undiluted calcines can be stored in 25-cm diameter cylinders after six years decay, or earlier if diluted. Glass at 35 wt% waste concentration can be stored at approximately one year; high waste concentration metal matrices can be stored at two years decay time or less. Waste products can be stored in water-cooled annular cylinders at a decay time as low as one year. The heat generation rate permitted in water-cooled cylinders is usually limited by the maximum product temperature. However, the high thermal conductivity of the metal matrix results in little temperature drop between the canister centerline and the wall, and canister wall temperature (60°C) becomes limiting. Product and wall temperatures using water cooling and corresponding heat generation rates are summarized in Tables IX and X.

Annual Canister Requirements

Since high-level waste may be costed on an individual canister basis, the number of canisters filled annually may significantly influence storage costs. Annual waste volume and the number of canisters should be minimized to reduce costs and maximize safety with respect to the number of canisters requiring shipment to a federal repository. The annual number of solid cylinders for various waste forms stored in air or water as a function of waste decay time is shown in Figure 10; the number of annular canisters for candidate waste forms is shown in Figure 11.

Air Cooling

Calcines and metal matrices have the least product volume per MTU and require the fewest canisters. Glasses and SGC's have

TABLE IX

CALCULATED TEMPERATURES AND HEAT GENERATION RATES
FOR WASTES IN WATER-COOLED SOLID CYLINDERS

Product	k W/m-°C	Diam. cm	T _w °C	T _m °C	Q kw/m ³
CALCINES	0.173	20	33	700	45
		25	30.5	700	28
		30	29.5	700	20
		35.5	29	700	15
SGC	0.242	20	36	700	62
		25	34	700	39
		30	32	700	28
		35.5	31	700	21
GLASS	1.038	25	53	800	126
		30	49	800	90
		35.5	47	800	66
		40.5	45	800	50
METAL MATRIX Pb	8.65	30	60	149	133
		40.5	60	176	98
		51	60	203	77
		61	60	229	63
AT	35.0	30	60	82	133
		40.5	60	89	98
		51	60	96	77
		61	60	102	63

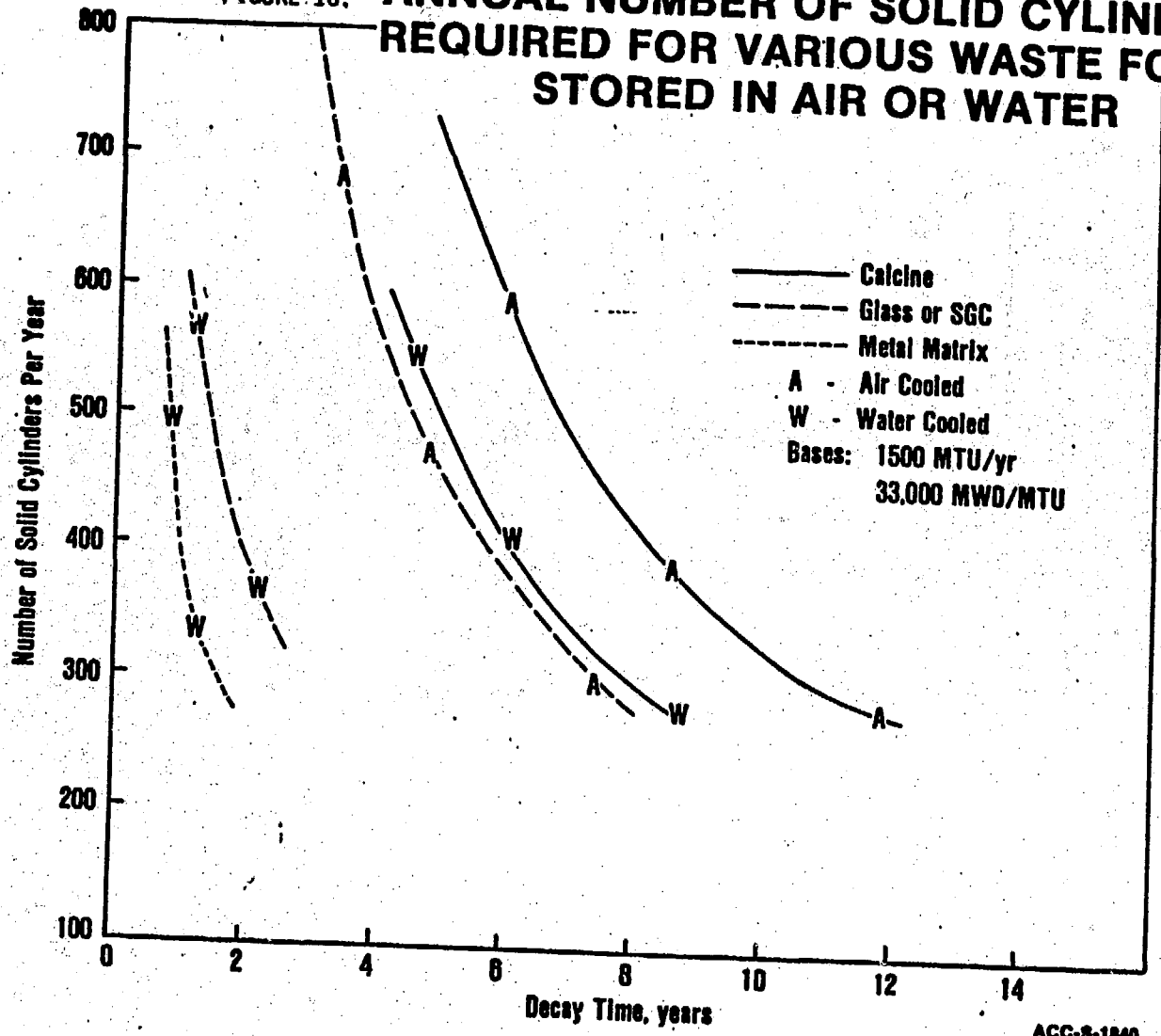
TABLE X

CALCULATED TEMPERATURES AND HEAT GENERATION RATES
USING WATER-COOLED ANNULAR CYLINDERS

Product	k W/m-°C	I.D. cm	O.D. cm	T _{lw} or T _{ow} °C	T _m °C	Q kW/m ³
CALCINES	0.173	40.5	61	44	700	87
		30	61	39	700	39
		25	46	44	700	87
	0.242	40.5	61	51	700	122
		30	61	44	700	54
		25	46	52	700	122
SGC	0.692	40.5	61	60	382	173
		30	61	60	509	107
		25	46	60	363	163
GLASS	1.038	40.5	61	60	274	173
		30	61	60	362	108
		25	46	60	262	164
METAL MATRIX Pb	8.65	40.5	61	60	85	177
		30	61	60	94	93
		25	46	60	83	110
Al	35.0	40.5	61	60	59	182
		30	61	60	68	114
		25	46	60	66	177

FIGURE 10.

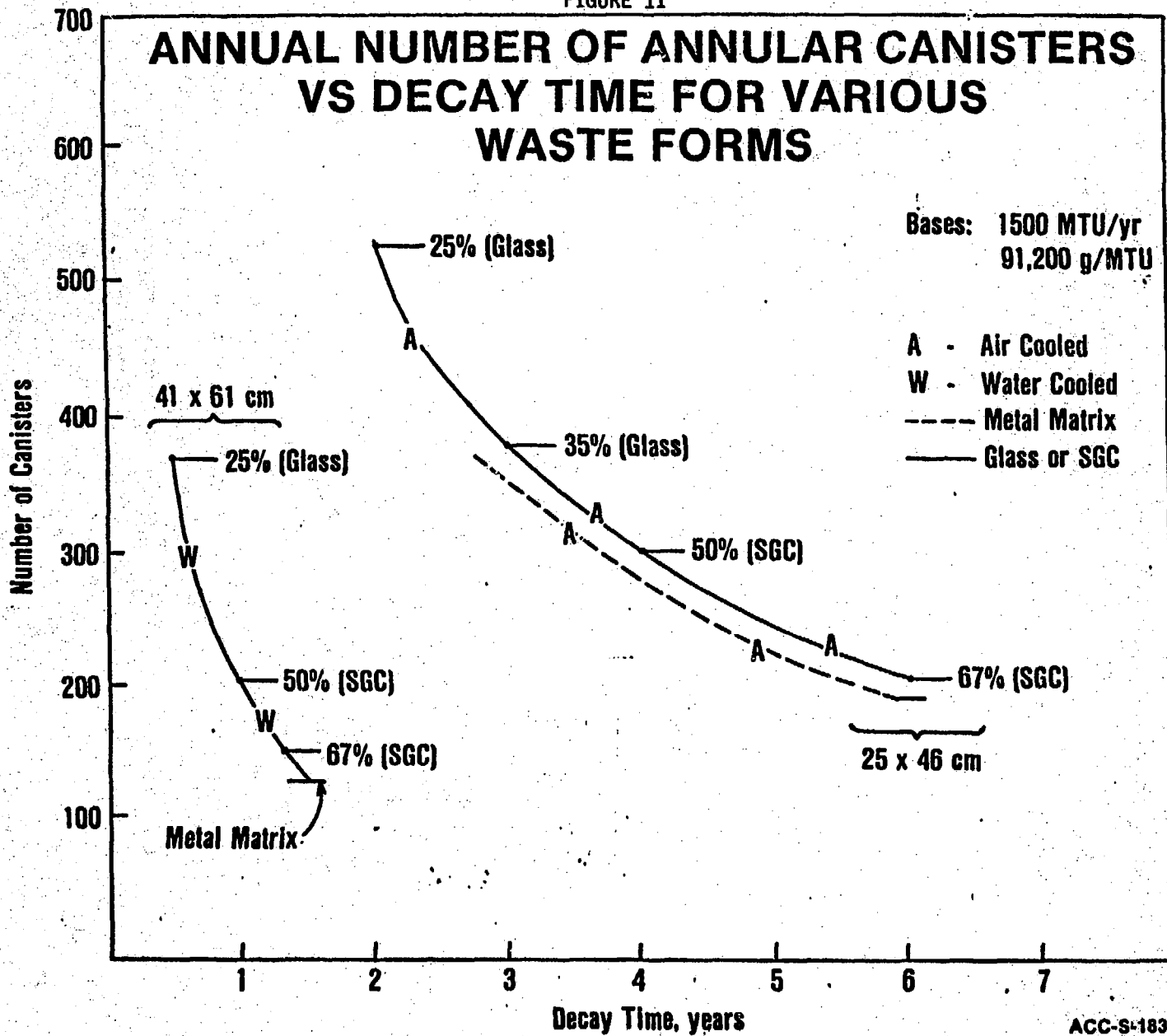
ANNUAL NUMBER OF SOLID CYLINDERS REQUIRED FOR VARIOUS WASTE FORMS STORED IN AIR OR WATER



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FIGURE 11

ANNUAL NUMBER OF ANNULAR CANISTERS VS DECAY TIME FOR VARIOUS WASTE FORMS



lower concentrations and require more canisters. An annular canister may be required if calcine is to be loaded at short decay times. High thermal conductivity metal matrices can be used to reduce the number of cylindrical canisters required annually.

Water Cooling

Water cooling is much more practical and fewer canisters are required for storage of short decay time waste. High waste concentration waste forms such as calcines, SGC's, or metal matrices require the fewest storage canisters. For three-year or older wastes, calcines and metal matrices can be placed in fewer annular canisters than glass or sintered glass ceramics. Metal matrices, because of high thermal conductivity and waste concentration, result in the fewest canisters at short decay times.

CONCLUSIONS

High-level defense wastes at the Idaho Chemical Processing Plant have been managed successfully for nearly a quarter of a century. Liquid wastes that generate sufficient decay heat to require cooling are stored in acid solution in stainless steel tanks inside an underground concrete vault. Wastes have been calcined and stored in stainless steel bins in underground concrete vaults since 1963. Cooling of the calcined solids by a combination of natural convection and thermal radiation to the vault walls has been demonstrated as a practical method for heat removal. Process heat input in fluidized-bed calcination using in-bed combustion of kerosene has been practical since 1970. Heat transfer in other candidate defense waste forms (within canisters) such as glass, sintered glass ceramics, and pellets do not present unique heat transfer concerns, since the volumetric heat generation rates are $<200 \text{ W/m}^3$; a canister would

thus contain a quantity of waste equivalent in power generation to a 20-100 watt light bulb.

Power reactor high-level wastes could be managed in a manner similar to the defense wastes at the ICPP; liquid wastes would be stored in stainless steel tanks similar in design to those at the Barnwell Plant. Fluidized-bed, spray, or rotary kiln calcination are candidate processes for solidification. In-bed combustion of kerosene using a fluidized bed could be used for providing process heat.

Solidified power reactor wastes have a significantly higher volumetric heat generation rate than defense waste calcine. Though the heat generation rate decreases rapidly during the first five years, the rate is still 100-1000 times greater than defense wastes after five years decay time. Solid cylinders and annular cylinders are practical for storage of the various waste forms in air or water. The decay time at which a given waste form can be placed in the canister is governed by the heat generation rate and the resultant maximum product or canister wall temperature. The annular canister permits all waste forms to be loaded at shorter decay times with fewer canister per year; because of the increased heat transfer surface area. All waste forms can be stored at shorter decay times in water-cooled canisters. Product centerline temperatures decrease rapidly for wastes loaded at less than five years decay time. A practical strategy would be to place high concentration waste in water-cooled canisters at 2-3 years decay time; canisters could then be transferred to air-cooled storage after two years to minimize the total number of canisters.

REFERENCES

1. B. P. Brown, E. S. Grimmett, and J. A. Buckham, Development of a Fluidized-Bed Calcination Process for Aluminum Nitrate Wastes in a Two-Foot-Square Pilot Plant Calciner, Part I, Equipment Development and Initial Process Studies, IDO-14586 (June 1962).
2. B. R. Dickey, "Long-Term Management of High-Level Defense Wastes at the Idaho Chemical Processing Plant", 87th National AIChE Meeting, New York, NY (November 1977).
3. T. K. Thompson, Development of In-Bed Combustion Heating for Calcination of Radioactive Wastes, IN-1278 (July 1968).
4. A. P. Hoskins, J. R. Berreth, Heat Transfer Considerations in Canister Storage of High-Level Solidified Wastes, ICP-1090 (April 1976).
5. J. T. Nichols, ICPP Development Programs for Fluidized-Bed Calciner Heat Input, ICP-1169 (September 1978).
6. R. E. Schindler, et.al., Development of a Fluidized-Bed Calciner and Post Treatment Processes for Solidification of Commercial Fuel-Reprocessing Liquid Wastes, ICP-1136 (December 1977).