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INTRODUCTION

Studies are under way in Chemical Research and Development to provide the technical "know-how" necessary to calcine Purex 1WW and safely store the solid product. The solid product can be stored as the particulate solid from the calciner or, after further treatment, as a solid matrix or melt. In this regard, Chemical Research is studying a $\text{PO}_4\text{-BO}_2$ melt. This document presents expected fission product heat generation rates in the solid product, shows the effect of various variables on heat transfer in the stored solid, and emphasizes certain items for further study. Also, a vault cooling method is suggested for further study.

SUMMARY

Heat generation rates from irradiated uranium as a function of cooling time since reactor discharge are shown below for two irradiation levels.

<u>Cooling Time</u>	<u>Heat Generation Rates, Btu/hr/ton U</u>	
	<u>800 MWD/T, 7 MW/T</u>	<u>1500 MWD/T, 10 MW/T</u>
90 days	8390	13700
1 year	1520	2670
3 years	280	510
5 years	120	226

The lower irradiation level is used throughout this document.

For uranium irradiated to 800 MWD/T at 7 MW/T heat generation rates in a waste storage vault being filled at a separations processing rate of one ton U/day are shown below as a function of cooling time between reactor discharge and waste calcination.

<u>Vault Operating Time, Years</u>	<u>Heat Generation Rates, Btu/hr/ton U/day</u>		
	<u>90 days cooling</u>	<u>3 years cooling</u>	<u>5 years cooling</u>
1	1,100,000	80,000	40,000
3	1,456,000	175,000	107,000
5	1,576,000	242,000	165,000

For higher processing rates multiply the above heat generation rates by the separations processing rate in tons U/day.

Thermal conductivity data as a function of temperature and bulk density (for a particulate solid) are needed. The effect of thermal conductivity upon the temperature drop from the center to the surface of 90-day-cooled waste in standard pipe is vividly shown below for the particulate solid (0.6 cu ft/ton U) and the $\text{PO}_4\text{-BO}_2$ melt (0.26 cu ft/ton U). Mean thermal conductivities for the given geometries are estimated to be 0.2 to 0.3 for the particulate solid and 0.6 to 0.8 for the melt.

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Mean Thermal Conductivity, Btu/(hr)(sq ft)(°F/ft)	Radial Temperature Drop, °F			
	Particulate Solid		PO ₄ -BO ₂ Melt	
	4 in. Pipe	5 in. Pipe	4 in. Pipe	5 in. Pipe
0.2	490	770	1131	1780
0.6	163	257	376	594
1	98	154	226	356
2	49	77	113	178

Studies of various methods of increasing thermal conductivity are needed. Proposed methods include adding ground up metal to particulate waste or forming a solid matrix or melt. Also, for a particulate solid, an increase in bulk density usually increases thermal conductivity.

Fixed-solid systems have other possible advantages. A solid matrix produced from molten metal could possibly be cast into any desired shape and dipped in a lower melting alloy or plated making the "casting" its own container. A solid matrix or melt could eliminate any need for a cooling-air filter at the storage vault to prevent air contamination if the container should fail.

Experimental determination of the resistance to heat transfer at the boundary between the waste and container wall is needed. It cannot be predicted mathematically.

Assuming allowable waste temperatures equal the temperatures attained during processing, allowable container diameters and annual container requirements for a processing rate of 6000 ton U/yr are shown below for two cooling times. Forced convection at an air velocity of 200 ft/min and 10-ft-long containers made from standard pipe were used as bases.

Cooling Time	Container Sizes and Requirements			
	Particulate Solid		PO ₄ -BO ₂ Melt	
	Pipe Dia., in.	Container/yr.	Pipe Dia., in.	Containers/yr
90 days	4	4073	4	1765
3 years	24	120	30	34

Techniques and designs developed or being developed to provide reactor fuel elements with good heat transfer characteristics could be useful for calcined LWW packaging and storage. The recently developed "garbage can" fuel element is an example.

Stack-induced convection instead of forced convection can be used to cool the vault if air requirements can be decreased sufficiently. This might be accomplished by interim liquid storage and then storage as a matrix or melt of high thermal conductivity in large-surface-area containers.

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DISCUSSION

The temperature attained by calcined LWV stored in a container is primarily a function of the heat generation rate, solids volume, storage vault and container geometry, thermal conductivity, and cooling method. These variables and calculations to "ball park" the effect of some of them are discussed in the following sections. Since many cooling methods and vault and container geometries can be devised, cooling by forced convection and containers made from standard pipe have been assumed for heat transfer calculations to simplify the discussion.

Heat Generation Rates

Fission product heat generation rates from uranium irradiated to 800 MWD/T and 1500 MWD/T at a specific power of 7 MW/T and 10 MW/T, respectively, are shown in Figure 1. The irradiation history of 800 MWD/T at 7 MW/T is expected to approximate that in use when calcined LWV disposal begins and is used throughout this document. The higher irradiation level might eventually be attained and is shown for information purposes.

Figure 2 shows heat generation rates in a storage vault as a function of vault operating time for an irradiation history of 800 MWD/T at 7 MW/T and cooling times of 90 days, 3 years, and 5 years between reactor discharge and waste calcination. Given heat generation rates are for a fission product addition rate to the vault equivalent to a separations processing rate of one ton U per day. For other heat generation rates multiply the given rates by the separations processing rate in tons of uranium per day.

Interim Storage

Interim storage as a liquid prior to calcination decreases tremendously the heat generation rates in the stored solid which, in turn, increases allowable container sizes and decreases final storage vault size and cooling requirements. For example, Figure 1 shows heat generation rates of 8390, 280, and 120 Btu/hr/ton U for a cooling time of 90 days, 3 years, and 5 years after reactor discharge, respectively. Using Figure 2 calculations show that the heat which must be removed from a storage vault after 5 years operation at a separations processing rate of 6000 ton U/year is 2.59×10^7 Btu/hr for waste stored directly out of Purex (90 day cooled) and 4.0×10^6 and 2.70×10^6 Btu/hr for a cooling time of 3 years and 5 years, respectively. The effect of interim storage upon container wall temperatures, waste center temperatures, and container sizes is brought out in the following sections.

Solids Volume

The volume of calcined LWV per ton of uranium affects container and vault sizes and the amount of heat in a container of a given size. The eventual composition and volume of calcined LWV are unknown. Calcined LWV may be stored as a particulate solid or possibly as a $\text{PO}_4\text{-BO}_2$ melt. Using the present Purex flowsheet as a basis, the particulate solid volume is estimated to be 0.4 to 0.9 cu.ft./ton U with a bulk density of 0.7 to 1.3. The melt volume is estimated to be 0.26 cu.ft./ton U with a bulk density of 2.3. For purposes of this document, a particulate solid volume of 0.6 cu.ft./ton U and the above melt volume are used.

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Container Cooling by Forced Convection

For this document it is assumed that containers are cooled by forced convection. Steady state wall temperatures in a stream of 70 F air have been calculated for single vertical pipes filled with solid LW. Details and limitations of the calculations are given in the Appendix. The effects of pipe diameter and two air velocities upon wall temperature are shown below for calcined LW whose fission product content has cooled 90 days since reactor discharge.

Std. (Sched. 40) Pipe Diameter, Inches	Wall Temperature, °F			
	Particulate Solid		PO ₄ -BO ₂ Melt	
	200 ft/min	400 ft/min	200 ft/min	400 ft/min
3	386	289	737	534
3.5	456	330	866	628
4	521	381	1000	726
5	674	489	1270	930

The effects of interim liquid storage and pipe diameter upon wall temperature are shown below for calcined LW whose fission product content has cooled 3 years since reactor discharge. The cooling air velocity is 200 ft/min.

Std. (Sched. 10) Pipe Diameter, inches	Wall Temperature, °F	
	Particulate Solid	PO ₄ -BO ₂ Melt
20	226	414
24	270	505
30	340	644

Higher heat generation rates from the melt resulting from a lower waste volume per ton of uranium explains its higher wall temperatures. Lower wall temperatures at higher air flows show the advantage of increasing air velocity either by increasing the total air volume fed and/or decreasing the array's free area transverse to the air flow. Long and narrow vaults and/or small container wall-to-wall distances could be used to increase air velocity.

Air velocities used represent 100,000 and 200,000 cu ft/min through a vault with a transverse free area of 500 sq ft when the container array is in place. These flow rates represent a large filter and large cost should a filter be required on the air stream. For example, the ventilation air filter at the Purex Plant was designed for a flow of 126,000 scfm. The filter is 83 ft by 53 ft by 15 ft thick and it cost about \$550,000 to build. Should a filter be required, the benefits of higher air flow will have to be balanced against filter costs as well as mechanical equipment costs.

Thermal Conductivity

To calculate temperature drop through calcined LW or maximum temperatures attained, the thermal conductivity must be known. Thermal conductivity for a homogeneous solid is a function of temperature. For a non-homogeneous solid

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it is a function of both temperature and bulk density. Therefore, thermal conductivity data as a function of temperature and bulk density (for a particulate solid) are needed to compare various geometries, container sizes, and cooling methods.

To show the effect of thermal conductivity, the temperature drop from the center to the surface of a cylinder of solid waste has been calculated as a function of standard pipe size and mean thermal conductivity. Figures 3 and 4 show the results for a particulate solid and $\text{PO}_4\text{-BO}_2$ melt, respectively. The fission product content has cooled 90 days between reactor discharge and waste calcination. The mean thermal conductivity represents a mean value of the different thermal conductivities due to temperature changes along a radial path in the waste. Further details are given in the Appendix.

As shown in Figures 3 and 4, slight increases in mean thermal conductivity of materials with mean thermal conductivities of less than about one result in spectacular decreases in the temperature difference. It is estimated from available data that the mean thermal conductivity for the conditions and geometries used is about 0.2 to 0.3 Btu/(hr)(sq ft)(°F/ft) for particulate LW and 0.6 to 0.8 Btu/(hr)(sq ft)(°F/ft) for $\text{PO}_4\text{-BO}_2$ melt. This illustrates the incentive for increasing the thermal conductivity of the stored solid.

Center Temperatures

Approximate center temperatures for standard pipe containers can be obtained by inserting the previously calculated wall temperatures and an equation for the variation of thermal conductivity with temperature into the basic conduction equation used to calculate the temperature drop through the waste. This accounts for the major effects upon center temperature, that is, heat transfer through the solid waste and wall-air interface. Lesser effects of heat transfer by radiation (from a hot container surrounded by hot containers) and heat transfer through the waste-solid container wall boundary and container wall are neglected. Further details of the calculations and thermal conductivity assumptions are given in the Appendix. Using the wall temperatures calculated previously and a fission product cooling time of 90 days, approximate center temperatures, T_c , for standard (sched. 40) pipe filled with calcined waste are shown below for several pipe sizes and two air velocities.

Pipe Diam., in.	Approximate Center Temperature, °F			
	Particulate Solid		$\text{PO}_4\text{-BO}_2$ Melt	
	200 ft/min	400 ft/min	200 ft/min	400 ft/min
3	660	590	910	710
3.5	800	710	1090	860
4	930	830	1290	1030
5	1240	1110	1700	1380

Approximate center temperatures for standard (sched. 10) pipe filled with calcined waste with a fission product cooling time of 3 years are shown below for an air velocity of 200 ft/min.

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<u>Pipe Diam., in.</u>	<u>Approximate Center Temp., °F</u>	
	<u>Particulate Solid</u>	<u>PO₄-BO₂ Melt</u>
20	630	670
24	810	870
30	1090	1190

Container Sizes

The container size is limited by the maximum temperature that can safely be attained by either the container wall or waste solid. From available corrosion data⁽⁶⁾ the wall temperatures listed earlier are not expected to be limiting for containers made of mild steel or low-alloy steel though the effect of such temperatures on vault walls needs further study. By assuming the maximum allowable waste center temperature, t_c , is the maximum temperature attained by the waste during processing (assumed to be 930 F for the particulate solid and 1300 F for the melt), container sizes can be determined from the approximate center temperatures given earlier. For 90-day cooled waste and an air velocity of 200 ft/min, the allowable pipe size is 4 inches for both the particulate solid and melt. Assuming containers are 10 feet long, a processing rate of 6000 ton U/year requires 4073 and 1765 containers per year for the particulate solid and melt, respectively.

For waste cooled 3 years and an air velocity of 200 ft/min, allowable pipe sizes are 24 inches and 30 inches for the particulate solid and melt, respectively. This represents 120 and 34 containers (10 feet long) per year for the particulate solid and melt, respectively, at a processing rate of 6000 ton U/year.

Methods of Increasing Thermal Conductivity

Various off-site investigators^(1, 2) have suggested and studied various methods of increasing thermal conductivity. These include adding ground up metal to a particulate solid or forming a solid matrix by incorporating the particulate solids into silicates, molten metals, such as lead or aluminum, or molten salts. These have not been evaluated but molten lead immediately appears questionable from a cost standpoint because of its high specific gravity and resultant high cost per unit volume. Aluminum or its alloys, with their low specific gravity, low melting point, and high thermal conductivity, are possibilities but their use is questionable with LWW solids due to the potential for a "thermite" reaction. A solid matrix would decrease the possibility of fission product contamination should the primary container fail and possibly eliminate any need for a cooling-air filter. Also, the molten system could be cast into any desired shape and dipped in a lower melting alloy or plated to provide a protective coating and allow the "casting" to be its own container.

The PO₄-BO₂ melt proposed by G. B. Barton, Chemical Research, will have a higher thermal conductivity than particulate waste.

For a particulate solid, increasing its bulk density is another possible method. For such systems the apparent thermal conductivity at a given temperature is a function of bulk density and usually increases as bulk density increases.

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Along this line, Chemical Research has pressed particulate waste. Bulk density increased from 0.6 to about 1.8.

Fuel Element Studies

The problem of keeping containers of radioactive waste cool is similar to that of keeping fuel elements cool in a reactor. Fabrication techniques and/or designs that have been developed or are under development to provide fuel elements with good heat transfer characteristics could prove useful for packaging waste. For example, the "garbage can" fuel element⁽⁷⁾ recently developed provides a large surface area with short paths for heat transfer.

Forced versus Stack-Induced Convection

Container cooling by forced convection (i.e., by mechanically pumping the cooling air by means of a fan or blower) was one of the bases used in previous sections and will undoubtedly be seriously considered for any ultimate waste storage scheme. However, from the ultimate disposal point of view it is not too desirable since the mechanical system could fail and it requires periodic surveillance, maintenance, and replacement. Stack-induced convection could eliminate these drawbacks but only under certain conditions. The inherently low available draft from a large stack restricts flow rates and probably precludes the use of filters on the air stream.

The information in previous sections indicates storage conditions might be devised which would allow cooling by stack-induced convection. Interim storage for three to five years decreases air requirements and/or increases allowable container sizes. Storage of a solid with a high thermal conductivity decreases air requirements and/or increases container size. Presuming a solid with a high thermal conductivity and no large resistance to heat flow at the waste-solid container-wall boundary, the major resistance to heat transfer is the air film at the container surface. Containers with large surface areas (finned tubes, annular containers, finned annular containers, etc.) could be used to further decrease air requirements. A decrease in air flow reduces the pressure drop through the vault and raises the air temperature. Inspection of the stack-draft equation in the Appendix shows that such a temperature rise increases the available draft. Finally, storing the waste as a fixed solid, such as a melt or solid matrix, could eliminate the need (if any) for a filter for the cooling air should the waste container fail.

The effect of a filter in the stack-induced-convection scheme is shown by comparing the pressure drop of the filter with available stack draft. The ventilation air filter at the Purex Plant was designed for an initial pressure drop of 4 inches of water. Available draft from a 200-ft-high, 7-ft-I.D. stack is shown below for several air temperatures and air flows assuming an atmospheric air temperature of 70 F.

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<u>Air Flow, scfm</u>	<u>Available Draft, inches of H₂O</u>	
	<u>350 F Air</u>	<u>600 F Air</u>
10,000	0.98	1.43
20,000	0.97	1.42
50,000	0.66	1.0
75,000	0.25	0.44

Solid-Solid Boundary

The resistance to heat transfer at the solid-solid boundary between the container wall and waste solid cannot be predicted mathematically. It must be experimentally determined. It could cause unpredicted high temperatures--especially if the differences in thermal expansion of the container and solid causes the wall to expand away from the solid. Study of this problem is needed.

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APPENDIX

Forced Convection Cooling

Wall temperatures for standard pipe filled with calcined LW were calculated using the following equations: (3)

$$q = hA \Delta T \quad 1$$

$$h = \frac{(0.26)(k_f)}{D_o} \left(\frac{C_p u_f}{k_f} \right)^{0.3} \left(\frac{D_o G}{u_f} \right)^{0.6} \quad 2$$

where q = heat transferred, Btu/hr;

h = surface coefficient of heat transfer, Btu/(hr)(sq ft)(°F);

A = area of heat transfer surface, sq ft;

ΔT = temperature difference between surface and cooling air, °F;

D_o = outside cylinder diameter, ft;

k_f = thermal conductivity for the cooling air at the film temperature,
 $T_f = \frac{T_w + T_a}{2}$, Btu/(hr)(sq ft)(°F/ft);

u_f = absolute viscosity of cooling air at the film temperature,
 lb/(hr)(ft);

C_p = specific heat of air at constant pressure, Btu/(lb)(°F);

G = mass velocity of cooling air, lb/(hr)(sq ft);

T = temperature, °F, T_a for air; T_w for wall temperature;
 T_f for film temperature.

To calculate wall temperatures for a given pipe size, a curve of the variation of ΔT with changes in h is drawn for equation 1 using the known values of q and A . By assuming values of T_w , values of ΔT , T_f , and variables of equation 2 are determined and a second curve of ΔT versus h drawn for equation 2. The wall temperature for the pipe at the existing conditions is determined from the ΔT at the intersection of the curves.

The wall temperature determined by the above method is a steady-state temperature for the assumed conditions. Steady state will not exist in the actual case since the heat to be transferred from the container is constantly decreasing. However, for the cases of interest where heat generation rates are high and containers small for short-cooled waste or where heat generation is nearly constant for long-cooled waste, the maximum transient temperatures will approximate the steady-state temperatures calculated by the above method.

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Equation 2 expresses an estimate of the heat transfer coefficient for forced convection at right angles to a single cylinder. In the actual case arrays of containers must be cooled. Little data are available on forced convection heat transfer across arrays of cylinders. In general, the literature agrees that the mean heat transfer coefficients for arrays of cylinders is 10 to 30 per cent higher than the coefficient for a single cylinder. This increase is achieved by increases in individual-row coefficients beyond the first row due to increased air turbulence. The coefficient for the first row that the cooling air passes and the next two or three is not expected to differ appreciably from that predicted by equation 2 for single cylinders at similar air velocities. Under most vault operating conditions these rows have the highest heat generation rates and limiting conditions occur there.

Radiation losses are not included in the wall temperature calculation since, beyond the first row, a hot container is essentially surrounded by other hot containers.

Waste Temperature Drop and Center Temperatures

The general form of the steady state conduction equation for the conduction of heat through a cylinder of material uniformly generating heat is: ⁽⁴⁾₍₈₎

$$\frac{1}{r} \frac{d}{dr} \left(r k \frac{dT}{dr} \right) + W = 0 \quad 3$$

where W = volumetric heat generation rate, Btu/(hr)(cu ft);

r = radius of waste cylinder or, in this document, inside radius of pipe, ft;

k = thermal conductivity of solid waste, (apparent thermal conductivity of particulate solid) Btu/(hr)(sq.ft.) (°F per ft).

T = waste temperature, °F.

Considering W and k constant along the radial path, r, integration of equation 3 yields: ⁽⁴⁾₍₈₎

$$\Delta T = T_c - T_s = \frac{W r^2}{4 k} \quad 4$$

where ΔT = steady state temperature drop through the waste, °F;

T_c = waste center temperature, °F;

T_s = waste surface temperature, °F.

Equation 4 was used to show the effect of thermal conductivity upon temperature drop by using the mean thermal conductivity of the material along a radial path for k.

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To calculate the waste center temperatures given in this document, the variation of thermal conductivity with temperature was assumed to be expressed by the linear relationship:

$$k = k_o (1 + aT) \quad 5$$

where k = thermal conductivity at temperature T (°F);

k_o = a constant, taken as 0.123 for the particulate solid and 0.604 for the melt;

a = a constant, taken as 0.00127 for the particulate solid and 0.000256 for the melt.

Inserting equation 5 for k in equation 3 and integrating yields,

$$T_c + \frac{a}{2} T_c^2 - T_s - \frac{a}{2} T_s^2 = \frac{W}{4k_o} r^2 \quad 6$$

To simplify, container wall temperatures were used as waste surface temperatures, T_s .

Equation 3 is for infinite cylinders and when applied to finite cylinders end effects are neglected.

Again, as stated in the preceding section, steady state will not exist in the actual case, but the maximum transient temperatures will approximate the steady state temperatures.

Heat generation rates will not be uniform due to the escape of gamma radiation from the cylinder. However, much of the gamma radiation from a cylinder will be absorbed by its neighbors.

Available Stack Draft

Available stack draft was calculated using the following equation:⁽⁵⁾

$$d_1 = 0.52 PH \left[\frac{1}{T} - \frac{1}{T_1} \right] - fW^2CH/A^3$$

where d_1 = available draft, in. of H_2O ;

H = height of stack above air inlet, ft;

P = atmospheric pressure, psia;

T = atmospheric temperature, °Rankine;

T_1 = temperature of stack gas, °Rankine;

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f = a constant, for this document values are for steel stacks or
0.0011 with gases at 350 F and 0.0015 at 600 F;

W = weight of gases passing per sec, lb;

C = stack circumference, ft;

A = area of passage, sq ft.

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HEAT GENERATION IN IRRADIATED UREANIUM

FIGURE 1

HEAT GENERATION IN A WASTE STORAGE VAULT

Irradiation level - 800 MW/D
Power level - 7 MW/t
Separations Processing Rate - Variable (ton/day)
Curves Give Heat Generation During Vault Filling

Cooling Time Before Vault Storage - 90 Days

5 Yrs.

3 Yrs.

Btu/Hr
Ton U/Day

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FIGURE 3

TEMPERATURE DROP IN A CYLINDER OF

CALCINED INKW.

Waste - Particulate Solid

Heat Generation Rate - 13980 BTU/Hr./cu. ft. of waste

Cooling time - 90 days since reactor discharge

RADIAL TEMPERATURE DROP (CENTER TEMP. - SURFACE TEMP.), °F

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