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To: R. E. Tomlinson
From: A. M. Platt - J. T. Krieg

STORAGE OF RADIOACTIVE AQUEOUS WASTES FROM PUREX PROCESSES

INTRODUCTION

The radio-active liquid wastes from a separations facility are customarily stored in underground waste storage tanks. The wastes from the proposed Purex Separations Plant will have a high concentration of the fission products and will result in a large evolution of heat. The possibility of these wastes boiling due to radio-disintegration of fission products has made a survey of the problem desirable. This report lists the results of this investigation.

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SUMMARY

An examination of the probable heat evolution of stored radioactive aqueous wastes from the proposed Purex plant indicates that external cooling (such as a shell and tube condenser), is a necessity. It is estimated (See Figure 2) that in concentrating systems peak rates of 10×10^6 Btu per hour and total heat production of 15×10^{10} Btu are involved. Comparative figures for non-concentrating systems are 6×10^6 Btu per hour and 5×10^{10} Btu. The methods proposed for removing this heat fall in two categories: first, methods for simply removing the heat; and second, methods for simultaneously removing the heat and effecting a volume reduction of the aqueous wastes.

The various schemes analyzed and the estimated unit cost per gallon of aqueous waste stored are tabulated below. All concentrating methods (assumed concentration factor = 2.4) are designed to produce condensed waste meeting cribbing tolerances.

<u>Heat Dissipating Unit</u>	<u>Unit Cost per Gallon Waste</u>
<u>Non-Concentrating</u>	
Immersion tube exchanger	59¢
Air cooled fin tube condenser	69¢
Air condenser with water spray	58¢
Shell and tube condenser	56¢
<u>Concentrating Units</u>	
Shell and tube condenser, ion exchange of condensed waste to crib	24¢
Double effect evaporator, shell and tube condenser, condensed waste to crib	25¢
Double effect evaporator, shell and tube condenser, ion exchange of condensed waste to ditch	25¢*

* It should be noted that this installation does not require a pipe line to the cribs while the other two cases require such a line.

In all cases outlined, the basic tank unit is of 750,000 gal. capacity and jacketed on the walls and bottom. This type construction is deemed necessary because of the uncertainty involved in predicting the formation of local "hot-spots" in the sludge, with consequent uncertainty of corrosion rates. Cooling will be needed for a period of six to eight years in the concentrating system in order to prevent excessive evaporation.

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It is recommended (subject to optimum design and construction considerations) that the Purex waste tank farm be constructed of six mild steel tanks, each with a nominal capacity of 750,000 gallons. Jackets should be provided on the walls and bottom of each tank. Each tank should also be equipped with a shell and tube condenser with a condensate draw-off to a common ion exchange system. The ion exchange system may be comprised of two alternately operating parallel resin beds 3-1/2 ft. in diameter and 7 ft. long plus one stand-by unit. The beds would be packed with 30% Dowex-50 and 70% IRA-400. It is estimated that regeneration with caustic would be necessary every 90 days. Such a design would provide sufficient flexibility to handle any process improvements that result in a decrease in waste storage volume.

DISCUSSION

The uranium charged to a Purex plant will be in the form of irradiated slugs containing uranium, plutonium, and fission products. Ultimately, essentially all of these fission products will appear in the concentrated aqueous wastes from the process. These wastes are jettied to underground storage tanks. Here the radio-active decay of the fission products will cause the evolution of large quantities of heat. Boiling will occur unless some provision is made to remove the heat evolved.

Heat Evolution

Methods of calculating the magnitudes of heat evolved during radio-active decay of fission products are described by Gillette.¹⁹ Such calculations are dependent on a knowledge of the irradiation and cooling history of the slugs involved. It is estimated that the slugs used for Purex feed will be irradiated 250 days at a power level of 2.4 megawatts per ton of uranium (600 MWD/T). The cooling period for slugs will be at least 90 days before discharge into the waste storage tanks. Using these data and the methods of Gillette, the expected rate of heat evolution from a given quantity of fission products has been calculated as a function of cooling time (see Figure 1). The data of Borst-Wheeler²⁷ indicate values about 1.8 times higher than those used for this report. On the other hand production tests¹⁵ on B Pile indicate values about 11 times smaller than those used. It is thought that the values shown, although not the highest or lowest on record, do represent a reasonable and safe compromise for design purposes.

In order to apply these data in estimating the heat evolution from an underground storage tank, it must be remembered that during filling operations both the average age and the quantity of fission products in the tank are continuously changing. Under these circumstances, the average rate of heat evolution may be found by suitable integration of the curve in Figure 1. Figure 2 presents such an estimate of the rate

of heat evolution versus time for a single waste storage tank. The calculations for this figure assumed a waste storage volume of about 460 gallons per ton of uranium processed²³ (200 tons U per month) and a single 750,000 gallon storage tank. Using this basis, a single tank would be filled in about 250 days. The peak rate of heat evolution occurs just as filling is completed and will be about 6×10^6 Btu/hr.

If the tank is allowed to self concentrate (by boiling off water) during filling, considerably higher rates of heat evolution are encountered. For a volume reduction of 2.4, peak heat evolution rates of about 9×10^6 Btu/hr. may be expected.

Figure 2 also indicates the rate of heat evolution during tank filling that may be encountered for plant operating rates from 150 to 250 tons of uranium per month.

In all cases the quantity of energy released is more than sufficient to boil the tank completely dry unless the heat is removed by external means.

Heat Dissipation

It has been estimated²¹ from the behavior of BiPO_4 waste storage tanks that ground losses are about 200,000 to 300,000 Btu/hr. when the tank temperature is in the range of 220-240°F. This value agrees reasonably well with unsteady state^{1,4,6} heat transfer calculations using a thermal conductivity of soil of about 1 Btu/(hr.)(ft.)(°F.). In practical terms, such a low ground loss will mean that a self concentrating tank will have to be externally cooled for about 6.3 years in order to prevent boiling. A non-concentrating tank will have to be cooled for about 4.6 years.

Several methods appear to be feasible for dissipating the excess energy involved. They may be classified into two general groups: 1) those methods which simply dissipate the heat without attempting to utilize the energy involved; and 2) those methods which utilize the evolved energy to reduce the volume of the stored wastes.

Non-Concentrating Systems

The energy developed during radio-active decay could be dissipated without utilization by either direct cooling using a heat exchanger, or by indirect cooling such as allowing the wastes to boil and refluxing the overhead vapors. DuPont has adopted an integral tube and tank arrangement for the Savannah River Plant. The exchanger consists of 2 inch vertical pipes with triangular spacing of 4 feet in a 74 foot diameter tank. At Arco, deionized water is sprayed on the top and flows down the sides of the tank, collects in a steel-lined concrete sump, and is pumped via a heat exchanger back to the spray heads; a pancake coil is provided to cool the bottom of the tank.

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Another possible installation would encase the walls and bottom of the tanks with jackets. The primary advantage of this installation accrues because of the excellent cooling of the tank shell. If fission products concentrated in the sludge in the tank, there is a possibility of local "hot spots" forming. The application of cooling water directly to the tank walls would remove this heat rapidly enough so that the tank wall would remain at a desirably low temperature even though there were local hot spots in the sludge.

The following types of heat exchangers should be considered for the indirect systems of refluxing overhead vapors: 1) air-cooled finned tube exchangers (such as currently installed on BiPO_4 waste tanks); 2) water sprayed finned tube exchangers, and 3) water-cooled shell and tube exchanger.

Concentrating Systems

Alternately, a number of methods of utilizing the evolved heat to achieve a reduction in volume of the liquid wastes present themselves. Experiments performed by Chemical Research indicate that when Purex wastes are concentrated by a factor of 2.5, the crystallization point is about 100°C . and at room temperature the slurry contains about 65% solids. Any process improvements which tend to reduce the solids content of the waste would permit a greater volume reduction.

A volume reduction factor of 2.4 was assumed in analyzing the following systems: 1) total condensation of overhead vapors combined with an ion-exchange bed to decontaminate the condensate to cribbing tolerances, 2) a double effect evaporator plus a total condenser with a condensate adequately decontaminated crib tolerance, and 3) a double effect evaporator, total condenser, and ion-exchange bed with sufficient decontamination for effluent discharge to an open ditch.

The decontamination factors required for the various methods of concentrating the wastes appear to be attainable. Experiments at other sites (B.N.L., O.R.N.L. and A.N.L.) indicate that a de-entrainment factor of at least 3×10^4 is obtainable^{10,12} for overhead vapors from neutralized or basic wastes. The de-entrainment factor for the second effect evaporator is expected to be somewhat less and of the order of 10^3 . It is expected that decontamination factors of 10^2 to 10^3 can be obtained through a mixed ion exchange bed (anion and cation resins) or ion exchange coupled with a solid reaction bed such as FeS or $\text{Ca}_3(\text{PO}_4)_2$. Data obtained by Chemical Research indicates that two alternately operating parallel resin beds three and one-half feet in diameter and seven feet long plus one standby would be sufficient to handle a tank farm. The beds would be packed with 30% Dowex-50 and 70% IRA-400. Regeneration using a 4 M caustic solution would take place every 90 days during peak load periods. The average rate of flow thru the bed during concentration would be about 3100 gallons per day. A summary of decontamination factors for the specified methods of concentrating are presented in the following table.

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Predicted Decontamination Factors

<u>Method</u>	<u>First Effect</u>	<u>Second Effect</u>	<u>Ion Exchange</u>	<u>Total</u>	<u>DF Required*</u>
Condenser + Ion Exchange	3×10^4	--	10^3	3×10^7	3×10^7
Double-Effect Evaporator + Condenser	3×10^4	10^3	--	3×10^7	3×10^7
Double-Effect Evaporator + Condenser + Ion Exchange	3×10^4	10^3	10^2	3×10^9	5×10^8

*Note: These values based on first increment added to storage. It is expected that Purex wastes as discharged will have an activity of about 1.5×10^2 β curies/liter before concentration and an activity of 3.6×10^2 β curies/liter after concentration. However, the ageing of wastes during concentration will reduce these activities by factors in excess of three.

Economic Appraisal

An economic appraisal of the various waste storage installations outlined above is presented in the attached Tables I, II, and III.

It will be noted that the waste storage costs for self-concentrating systems are considerably reduced below those for non-concentrating systems due to the concentration of the wastes and resultant additional space for waste storage. The three concentrating systems are competitive. The data for the total condenser-ion exchange-crib system indicate a slight edge over the other methods -- apparently due to the equipment investment. In addition, this unit is the most attractive operationally. It contains no moving parts such as steam traps and the complex instrumentation that would have to be installed on a double effect evaporator system. As such, it is well suited for operation with a minimum of attention.

It will be noted that in all cases jackets are included on the sides and bottom of the tanks. This recommendation is based on the following premises. First, potential process and/or equipment improvements may reduce the solids content of the aqueous wastes.

Such improvements would result in waste volumes less than 460 gal./ton U with a resultant increase in the rate of heat evolution. Second, it is possible to self concentrate wastes beyond a factor of 2.4. Using the present HW #1 Purex Flowsheet,²³ a concentration factor of 3.6 could be employed to produce slurry containing 95% solids. With flowsheet modifications, concentration factors in excess of 3.6 are not unreasonable. If the wastes were concentrated to such a point, the possibility of local "hot spot" formation would be augmented. In this case, the use of cooling water in the tank jacket should prevent damage to the tank due to local overheating.

Corrosion

While the cost factors developed indicate that the concentration systems are an economical approach to the storage of wastes, it should be borne in mind that certain problems are introduced which must be answered in a final evaluation of this problem. Primary among these problems is the possibility of excessive corrosion in tanks at elevated temperatures. Although Purex type wastes have not been used in corrosion tests, experiments with BiPO_4 ²² and Redox²⁴ wastes indicate that vapor corrosion is greater than liquid or gas-liquid interface corrosion. These tests indicate that vapor corrosion rates of from 10 to 30 x 10⁻⁵ inches per month may be expected, with pit penetrations up to 10 times this amount. The similarity of Redox and Purex wastes (as shown below) indicate that corrosion rates should be of the same magnitude.

TYPICAL REDOX AND PUREX WASTES

	Redox <u>M</u>	Purex <u>M</u>
NaNO_3	3.81	2.95
NaNO_2	0.09	0.46
NaAlO_2	1.29	0.63
NaOH	1.32	0.62
Na_2SO_4	0.05	0.12
Na_2SiO_3	0.001	0.012
$\text{Na}_2\text{U}_2\text{O}_7$	0.001	0.006
Na_2CrO_4	0.04	
K_2CrO_4	0.001	
NH_3	0.05	
$\text{Cr}_2\text{O}_3 \cdot 2\text{H}_2\text{O}$	0.002	
$\text{Fe}(\text{OH})_3$	0.02	0.04
$\text{Mn}(\text{OH})_2$	0.006	

However, this tentative conclusion will have to be verified or disproven by experimental evidence. Experiments are in progress by Chemical Development to check corrosion with Purex type wastes using different concentration levels.

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TABLE I

ESTIMATED EXCHANGER REQUIREMENTS

Peak Load - 10,000,000 Btu/hr.

<u>Exchanger</u>	<u>Btu</u> <u>U, hr-ft²-F</u>	<u>log mean</u> <u>Δt, F</u>	<u>A, ft.²</u>	<u>Cost,</u> <u>\$/ft.²</u>	<u>Direct</u> <u>Cost \$</u>
Immersion Tubes	20	100	4,200	6	25,000
Jackets, Walls & Bottom	10	100	10,000	10	100,000
Air Cooled Fin Tubes	10	80	12,500	8	100,000
Air Cond. With Water Spray	40	100	2,500	8	20,000
Shell & Tube Cond.	150	100	670	6	4,000
Double Effect Evap.	150	30	2,300	10	23,000
+ Shell & Tube Cond.	150	100	670	6	4,000

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TABLE II

COST OF NON-CONCENTRATING WASTE STORAGE UNITS

<u>Heat Dissipating Unit</u>	<u>Cost</u>
<u>Immersion Type Tubes</u>	
Tank, 750,000 gal.	\$315,000
Jackets, Sides and Bottom	100,000
Tubes	25,000
Cooling water*	2,000
Cost per gal. waste	58.9¢
<u>Air Cooled Fin Tube Condenser</u>	
Tank, 750,000 gal.	315,000
Jackets, Sides and Bottom	100,000
Air Condenser	100,000
Cost per gal. waste	68.7¢
<u>Air Condenser With Water Spray</u>	
Tank, 750,000 gal.	315,000
Jackets, Sides and Bottom	100,000
Air Condenser	20,000
Cooling water*	2,000
Cost per gal. waste	58.3¢
<u>Shell and Tube Condenser</u>	
Tank, 750,000 gal.	315,000
Jackets, Sides and Bottom	100,000
Water cooled condenser	4,000
Cooling water*	2,000
Cost per gal. waste	56.1¢

* Cooling water costs are based on \$12.50 per 10⁶ gallons.

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TABLE III

COST OF CONCENTRATING WASTE STORAGE UNITS

<u>Heat Dissipating Unit</u>	<u>Cost</u>
<u>Shell & Tube Condenser + Ion Exchange</u> <u>(Waste to Crib)</u>	
Tank, 750,000 gal.*	\$315,000
Jackets, Sides and Bottom	100,000
Condenser	4,000
Ion Exchange	5,000
Cooling water**	5,500
Line to Crib***	10,000
Cost per gal. waste	24.4¢
<u>Double Effect Evaporator</u> <u>(Waste to Crib)</u>	
Tank, 750,000 gal.*	\$315,000
Jackets, Sides and Bottom	100,000
Evaporator	23,000
Condenser	4,000
Cooling water**	5,500
Line to Crib***	10,000
Cost per gal. waste	25.4¢
<u>Double Effect Evaporator + Ion Exchange</u> <u>(Waste to Ditch****)</u>	
Tank, 750,000 gal.*	\$315,000
Jackets, Sides and Bottom	100,000
Evaporator	23,000
Condenser	4,000
Ion Exchange	1,000
Cooling water**	5,500
Cost per gal..waste	24.9¢

* These tanks will store an equivalent of 1,800,000 gal. using a volume reduction factor of 2.4 based on Chem. Res. Data.

** Cooling water costs based on \$12.50 per 10⁶ gallons.

*** The cost of the line to crib was very roughly estimated. This value may be in error by 100%.

**** It should be noted that this installation does not require a pipe line to the cribs while the other two cases require such a line.

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FIGURE

Value of Final Product Radiation Per Watt
of Pile Power Level Vs Days Cooling

SLUG RADIATION TIME - 250 DAYS

840 900 960 1020 1080 1140 1200 1260 1320 1380 1440 1500 1560 1620 1680 1740 1800
Cooling After Pile Discharge

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