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PRELIMINARY DESIGN CRITERIA FOR THE INTERIM STORAGE OF HIGH-LEVEL RADIOACTIVE LIQUID WASTES

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PRELIMINARY DESIGN CRITERIA

FOR THE

INTERIM STORAGE OF HIGH-LEVEL RADIOACTIVE LIQUID WASTES

by

H. W. Stivers
Radiological Development
Facilities Engineering
Chemical Processing Department

December 16, 1960

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PRELIMINARY DESIGN CRITERIA FOR THE INTERIM STORAGE
OF HIGH-LEVEL RADIOACTIVE LIQUID WASTES

INTRODUCTION

Approximately five years of process improvements and production line waste handling experience have been accumulated since the newest tank farm, 241-A, was placed into operation. Process improvements and modifications have resulted in waste volume reductions by decreasing the quantity of solids remaining in the liquid wastes from a unit of production. The resulting concentration of fission products has produced mildly boiling solutions in the storage containers permitting further waste volume reductions. Waste handling procedures have been modified to provide safe interim containment of these concentrated wastes, but restrict the usable space in existing facilities. (6) The resulting concentration of fission products in existing facilities has a maximum heat generation rate of 10 to 15 BTU/hour per gallon stored and the contents have a potential boiling life in excess of the anticipated life of the structure. Concentrations of fission products stored in the anticipated smaller tanks have an average heat generation 75 to 100 BTU/hour per gallon stored.

A preliminary design criteria is needed as the bases for an engineering evaluation of tank design concepts to obtain maximum benefits from our operating experiences in the light of the future wastes and the Chemical Processing Department waste management program. (5)

PURPOSE

The purpose of this preliminary design criteria is to provide the bases for an engineering study for the next high-level radioactive waste storage facility.

SUMMARY AND CONCLUSIONS

- (1) Operating experience gained from storing self-boiling wastes in existing tanks, forecasts of future waste characteristics, and the intent of the next tanks reveal some desirable changes in the 241-A tank design for new facilities and future wastes. Briefly, these changes are as follows:
 - (a) Provide a positive method of leak detection with means of routing the leakage to other storage facilities.
 - (b) Provide maintainable instrumentation for control of actively boiling solutions in large-volume, re-usable tanks where there is a potential for non-homogeneous mixtures.
 - (c) Provide means for maximum sludge removal.

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- (d) Provide improved structural integrity for the physical characteristics of these wastes.
2. For the purpose of this study, cell drainages, carbonate wash wastes, and other low-solid content, highly radioactive waste solutions should be combined with the Purex LW high-activity wastes for a final concentrated liquid volume of 50 gallons and a dilute volume of 300 gallons per ton of processed uranium. It should be planned to retain these wastes for an interim cooling period as neutralized, self-boiling liquids to provide as nearly as possible a homogeneous solution. At the 50 gallon/ton volume concentration, it may be desirable to "cut" the sludge with an acid solution so that it could be later transferred to a solidification process.
 3. For the purpose of this study, a total liquid storage capacity of 1,000,000 gallons plus one tank should be provided for the concentrated unit volume in the most economical number of equal units. A total liquid volume of 6,000,000 gallons plus one tank should be provided for the dilute unit volume of 300 gallons per ton. It is assumed that the "one-additional-tank" would be used as the last receiving tank during the period when the contents in the first receiving tank are being transferred to a solidification process. A vapor space should be provided in each tank equivalent to about 12 percent of the liquid volume.
 4. Conceptually the proposed tank farm would consist of several primary tanks of repetitive design constructed of either carbon or stainless steel. The interior of the primary tank would be void of structural supports, cooling coils, and irregularities which would hinder sludge removal. Provision should be made for the following:
 - a. Positive leak detection and recovery of wastes before a "significant amount" (11) enters the adjacent ground.
 - b. Replaceable air-lift circulators to minimize bumping and maintain uniform heat load on the surface condensers.
 - c. Piping, valving, and instrumentation on each tank for the required flexibility and control in handling of wastes, condensates, cooling water, and air to the circulators.
 - d. Visual inspection of the interior of the primary tank with the aid of remotely operated optical equipment.

These tanks would be located so that maximum utilization could be made of existing tank farm facilities. A gross heat evolution curve, a unit heat evolution curve, radiation shielding curves, and a preliminary schematic flow diagram are presented in the Appendix to aid in the study of this facility.

FUTURE WORK

It is recommended that:

1. This preliminary design criteria be used as the bases for the conceptual design of ge facilities for the positive containment of neutralized, self-boiling, highly radioactive, aqueous wastes.
2. CE&U provide the engineering design as needed to define the comparative project cost estimates.
3. Economic evaluation be derived from project cost estimates of the entire capacity of the facility complete and ready for use.

DISCUSSION

Preliminary Waste Characteristics

For the purpose of this study the following range of chemical compositions were used to describe the chemical characteristics of the stored wastes after formaldehyde kill.

Probable Composition*

Molarity Range for R&D Work (Incl. FP Recovery)

<u>Ion</u>	<u>M</u>	<u>Min.</u>	<u>Max.</u>
H		0.3	1.5
NO ₃ -NO ₂	2.3	0.1	6.0
Fe	0.75	0.1	0.8
Cr	0.04	0.01	0.1
Ni	0.02	0.01	0.2
Al	0.10	0.1	0.2
Na	8.0	0.2	13.0
PO ₄	0.01	0.01	0.1
SO ₄	1.0	0.1	4.0
Si	0.2	0.1	0.4
Mn	0.15	0	0.6
K	0.15	0	0.6
U	0.01	0	0.01
Hg		0	2.0
HCO ₃ or CO ₃	1.2	1.0	2.0

* Before neutralization

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The anticipated sludge volume for the formaldehyde killed LW and carbonate wash wastes would be in the range of 40 to 60 percent at a final volume concentration of 50 gallons per ton of uranium processed.

The preliminary physical properties of the composite waste after formaldehyde kill and neutralization for structural analysis of the tank is expected to be as follows:

	<u>Phases</u>		
	<u>Vapors</u>	<u>Liquid</u>	<u>Sludge</u>
Max. Temperatures (°F)	212 - 230	212 - 230	250 - 300
Specific Gravity		1.35	2.5
pH	9.5	9.5	9.5
Percent Solids		10	90
Viscosity		Not Available	
Thermal Conductivity		Not Available	
% Fission Products	0.1	30 - 40	70-60
Heat Evolution	See Fig. 1(a) and 1(b) in Appendix		
Shielding Requirements	See Fig. 2 and 3 in Appendix		

Predicted Operating Procedure

It may be assumed that a liquid heel at ambient temperatures will be put into the tank prior to initial filling with concentrated wastes. Filling of the tank with wastes would be accomplished in a minimum time lapse to provide a uniformly aged feed for the solidification process. The most critical operating times will be during the filling and emptying of the tank when the heat evolution rate (per unit volume) is high and before and after the air-lift circulators are submerged for operation. After the circulators are submerged, caution should be employed so as not to over-concentrate the wastes thus inducing abnormally high temperatures and irregular heat evolution. It is desirable that tank contents remain mobile for eventual transfer to the solidification process.

The operating vapor pressure of the tank should be maintained at a less pressure than the atmospheric pressure to provide an inward air flow from the outside atmosphere. The tanks would be connected individually to a common vapor header system which could be tied into the existing surface condensers.

[REDACTED]

In the event the tank leaks, the wastes would be collected and routed to another tank. Provision for inspection of the interior surface of the tank by remotely operated optical equipment should be provided to evaluate any possible structural or corrosive damage which could develop from the operation of the tank.

After the wastes have sufficiently cooled, they could be transferred to a solidification process. The tank could then be reclaimed, inspected, and made available to receive fresh boiling wastes from process. Normal operating conditions could permit several re-uses of these tanks.

Requirements of the Storage Tank

The requirements of the storage tank are the most stringent of any large-volume, low-pressure liquid container used by the Chemical Processing Department. The contents are lethal and heat evolved from the decaying of the concentrated wastes is capable of boiling the contents for several decades. In the event of loss of control, the resulting maximum temperatures and pressure surges could be detrimental to a container designed to the recommended maximum conditions in this report. It is not economically feasible to design for the maximum uncontrolled temperatures, vapor pressure surges, and resulting heat evolution rates. Therefore, a reasonable operational control must be employed which will optimize freedom and safety of operation without jeopardizing the integrity of a realistically economic structure. The criteria herein attempts to accomplish this economical balance and the resulting lowest-cost facility meeting these requirements would then be the "most-economical-design."

The design requirements for two alternate methods of interim storage are summarized in the table below. Alternate I provides a concentrated volume of waste (50 gal/TU) with a high percentage of sludge. The wastes would be neutralized, self-boiling at a slight vacuum, and have a high potential for temperature excursions. It is anticipated that an acid-cut would be required for sludge removal and therefore stainless steel is recommended as the structural material. Alternate II provides a dilute volume of waste (300 gal/TU) with a low percentage of sludge. These wastes would also be neutralized, self-boiling at a slight vacuum, but would have a low potential for temperature excursions. It is anticipated that sludge removal could be accomplished by pumping, sluicing and/or steam jetting and therefore carbon steel is recommended as the structural material.

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METHODS OF INTERIM STORAGE

<u>Design Requirements</u>	<u>Alternate I</u>	<u>Alternate II</u>
1. For Primary Tank ASME, Sec. VIII Construction Material	304-L SS (SA 240)	Carbon Steel (SA 212 B)
2. <u>Functions</u> Positive leak detection and recovery. No interior obstructions. Uniform dissipation of heat. Remote visual inspection. 100% radiograph all welds. Two years minimum retention. 25-year design life. Maintainable instrumentation and equipment.	Same for each alternate	
3. Usable Liquid Volume	1,000,000 gallons plus one tank	6,000,000 gallons plus one tank
4. Vapor Space	12% of the liquid volume	12% of the liquid volume
5. pH	> 9.0	> 9.0
6. Specific Gravity (Average for structural design)	2.0	1.3
7. Percent Solids (Total Settable Solids in Storage)	> 50	< 5
8. Vapor Pressure Maximum (psia) Minimum (psia)	25 13.7	17.2 13.7
9. Maximum Temperature (°F) Incremental Change (°F)	600 150	300 40
10. Rate of Fill Wastes Raw Water	75 gpm for one hour @ 200° F 300 gpm @ 65° F	75 gpm for 3 hrs. @ 200° F 300 gpm @ 65° F
11. Corrosion allowance (inches)	1/16	1/16
12. Shielding	Fig. 2&3 Appendix	Fig. 2&3 Appendix

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Operational flexibility shall be provided for the storage tank as follows:

- a. Receiving of fresh wastes from process;
- b. Receiving of condensate from reflux surface condensers;
- c. Receiving of raw water from 200-E Area distribution system;
- d. Receiving of emergency raw water for liquid level control;
- e. Receiving of wastes from failed primary tanks;
- f. Receiving of residues from the solidification process;
- g. Two spare receiving nozzles accessible near the tanks;

A pump pit for (h, i, j).

- h. Two discharge lines to a waste solidification process;
- i. One discharge line to other storage tanks;
- j. Circulation of wastes during filling;
- k. Maintainable instrumentation control;
- l. Access risers for: Waste samples, sludge level, pH, corrosion coupons, liquid level, radiation and temperature profile, replaceable air-lift circulators, inspection risers for optical equipment (12" min.).

Heat Removal from Concentrated Wastes

Waste volume reductions per unit of production have reduced tremendously the capital investment for liquid, high-level radioactive, waste storage facilities. However, the economical and safe dissipation of heat evolved from the decay of the concentrated wastes has developed complexities. Originally, in 1945, the heat evolution from a relatively "hot" tank was less than one BTU per hour for each gallon stored. New processes and process improvements increased this figure to about two or three BTU per hour for each gallon stored which resulted in mildly boiling solutions at Redox. At Purex, approximately six months of plant operation and modifications were required after startup (in 1956) to concentrate the waste sufficiently to induce self-boiling (an equivalent heat load of approximately one to two BTU per hour for each gallon stored). Currently, the Purex wastes in storage are concentrated sufficiently to obtain a heat generation rate of 10 to 15 BTU per hour per gallon. By the time new facilities are placed into operation, the concentrated wastes could induce average heat evolution rates in the range of 75 to 100 BTU per hour per gallon. Accompanying this high heat evolution rate is the possible intermittent high temperatures and

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pressure surges induced by the characteristics of the stored solution. If uncontrolled and over-concentrated, the temperatures and vapor pressures can become greater than those recommended for the design criteria. Therefore, this study should incorporate maximum protection without prohibitive costs for the safe and uniform dissipation of the evolved heat. This could be done by utilizing replaceable air-lift circulators and the existing surface condensers at other tank farms.

Fig. 1(a) in the Appendix illustrates the anticipated gross heat evolution, while Fig. 1(b) illustrates the anticipated unit heat evolution as a function of time for the characteristics of the future wastes. Decay curves are also illustrated in Fig. 1(a) for liquid storage volumes of 150,000 and 250,000 gallons. Decay curves for other volumes can be extrapolated from these curves.

Preliminary Instrumentation Requirements

Instrumentation control for the proposed facility will be more complete and maintainable. Of primary importance is the provision of replaceable thermocouples in the stored liquid, and in the structure to determine maximum temperatures and corresponding thermal gradients across the structural member. Instrumentation must be provided to automatically detect a significant leak from the tank.

Liquid level control should be provided to prevent accidentally over-filling the tank

Instrumentation control should be provided to detect an unusual rate of temperature rise in the stored liquid so that counter-control could be employed without delay.

Other primary instrumentation which is common to the tank farm operation includes the following:

- a. Liquid level measurement;
- b. Vapor pressure;
- c. pH;
- d. Specific gravity;
- e. Process flow control;
- f. Air supply to air-lift circulators
- g. Off-gas vapor pressures.

Auxiliary Facilities

The principal auxiliaries included in this study for the safe operation of the tank farm include the following:

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a. Control Building

A building similar in size to the 241-A Control House. Included in this building would be process flow control instrumentation and electrical switchgear, a change room with lockers and a minimum of four individual shower stalls, lavatory, a 90 square foot operators office. Temperature recorders will be provided in this building for all of the thermocouples installed in the tanks.

b. Diversion Box

There would be a diversion box similar to the 241-A-152 diversion box, which would provide the transfer flexibility of the incoming wastes from the process. The other function of this box would provide the transfer flexibility of "aged" wastes to a waste calcination facility. This box should be designed to handle acidic solution if the Alternate I method of storage is employed.

c. Instrument Control Building

Each tank should be provided with a small instrument control building to house the air supply instrumentation for the air-lift circulators. Heat to this building would be supplied by electrical heaters or steam.

d. Off-Gas System

The off-gas system should consist of a vapor seal to prevent overloading the storage tank; one cyclone separator for the tank farm and a return to a storage tank to remove the majority of the fission products, and a tie-in to the existing surface condensers. A new valve pit would be provided at the existing condensate receiver for individual, carbon steel return lines to each tank.

e. Sanitary water should be provided to the control building for domestic purposes.

f. Raw water should be provided to the storage tank for liquid level control, and to the tank farm for general wash-down of diversion boxes, pump pits, and tank risers, etc.

g. Electricity should be furnished at the tank farm for area lighting, operation of transfer pumps, operation of instruments, and heating of buildings.

h. Compressed air should be provided at the tank farm for the operation of air lift circulators, instrumentation, and general purpose cleaning.

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1. Low pressure steam should be provided at the tank farm to aid in the maintenance of instrumentation equipment and cleaning purposes.

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Fig. 1a - Fission Product Heat Generation in Underground Storage Tanks

Ref: (1) HW-42488, "Fission Product Heat Generation Tables," WH Swift
and GL O'Neill, April 10, 1956.

(2) Reference (3) in Bibliography

The accompanying heat generation and decay curves were calculated with the aid of the referenced heat generation tables and assuming the following irradiation and production histories:

<u>Definition</u>	<u>Units</u>	<u>Value</u>
Irradiation Level in Megawatt Days/Ton	(MWD/T)	700
Pile Power Level in Megawatts/Ton	(MW/T)	7.5
Days of Cooling Since Pile Discharge	(T/C)	90
Unit Volume of Wastes in Gallon/Ton	(Gal/T)	50
Average Production Rate in Tons/Day	(T/D)	21
Heat Loss from Primary Tank	(BTU/hour)	0

The axis of abscissas is the time since pile discharge expressed in days while the axis of the ordinates is the gross heat production of the decaying fission products expressed in BTU/hour.

Under these conditions the maximum heat generation rate is 14,500,000 BTU/hour for a liquid volume of 150,000 gallons and 19,000,000 BTU/hour for a liquid volume of 250,000 gallons. A maximum heat generation rate of 30,000,000 BTU/hour is anticipated for the entire tank farm storing 1,000,000 gallons of waste.

Fig. 1 also illustrates the fuel element decay time (90 days), separations processing time, the fill times for individual tanks containing the respective volumes, cooling time in storage, the allotted time for reprocessing and inspection of storage tank which completes one cycle of interim cooling. The 150,000 gallon size provides a mean decay time of 1,115 days while the 250,000 gallon size provides a mean decay time of 1,040 days from the time of discharge from the reactor.

After transferring the cooled fission products to the solidification process, the primary tank could then be inspected and made available to receive fresh wastes from the separations process.

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LIQUID COOLING / RE-PROCESS INSPECTION

185-D

250,000 GAL. 1115 DAYS MEAN DECAY TIME

240-D 250,000 GAL. 1040 DAYS MEAN DECAY TIME

1,000,000 GAL. LIQUID VOLUME

30,000,000 BTU/HR

FIG. 1a FISSION PRODUCT HEAT GENERATION
IN UNDERGROUND STORAGE TANKS

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4/25/60

840 960 1080 1200 1320 1440 1560 1680

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Fig. 1b - Fission Product Unit Heat Generation in Underground Storage Tanks

Ref: Un documented criteria and data from GC Oberg dated July 29, 1960.

In addition to the basic assumptions for Fig. 1a, the following assumptions were used for the unit heat generation curves in Fig. 1b.

- a. Storage tank capacity 250,000 gallons, 50 foot diameter, discharge of air lift circulator 6.5 feet above tank bottom, water heel of 95,000 gallons.
- b. A relationship between gallons/ton and amount of sludge was assumed.
- c. Supernate was assumed to contain all Cs plus 25 percent Sr plus the difference in actual sludge and that estimated at a volume concentration of 50 gallons per ton. Ten percent of the sludge volume was also estimated to be suspended by the air-lift circulators.

The purpose of this graph is to aid the heat transfer engineer in defining the design of the heat removal system for the interim storage period. The elapsed time measures the filling, decaying and transferring times during this period, and it is initiated when the high activity waste stream is first routed to the tank. The left ordinate axis is a scale of the unit waste heat generation expressed in BTU/hour (gal.) for the supernate and sludge. The right ordinate axis is a measure of the total waste volume expressed in gallons for the sludge and supernate in a 250,000-gallon storage tank. At a final volume concentration of 50 gallons a ton, the sludge and supernate are each estimated at 50 percent of the total volume; therefore, at the end of the fill period, the supernate and sludge volumes emerge as a single line at a volume of 125,000 gallons each.

The unit heat generation and/or total heat generation can be determined for the sludge, the supernate, or total volume for any desired elapsed time during the interim storage period.

Example: Assume an elapsed time of 75 days.

From Fig. 1b

(1) Volume of sludge in tank	38,000 gal.
(2) Volume of supernate in tank	92,000 gal.
(3) Waste heat generation in supernate	17 BTU/hour (gal.)
(4) Waste heat generation in sludge	225 BTU/hour (gal.)

- a) Total waste volume in tank = (1) + (2) = 131,500 Gal.
- b) Total heat generation in sludge = (1) x (4) = 8,500,000 BTU/hour
- c) Total heat generation in supernate = (2) x (3) = 1,560,000 BTU/hour
- d) Total heat generation in tank = b + c = ~ 10,000,000 BTU/hour

From Fig. 1a, the total heat generation after 75 days of filling (175 days since Pile discharge) would be ~ 9,600,000 BTU/hour which indicates the two Figures to be in close agreement.

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FIG. 1.6. FISSION PRODUCT UNIT HEAT GENERATION IN UNDERGROUND STORAGE TANKS

WASTE VOLUME, GAL $\times 10^3$

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Fig. 2 - Gamma Intensity Curves

The accompanying gamma intensity curves provide the gross gamma radiation levels (mr/hour) for a horizontal cylindrical tank and a vertical cylindrical tank with reinforced concrete shells of 21 and 15 inches, respectively, when covered with dry sandy soil. It is recommended to use 0.1 mr/hour for the design gamma radiation level at the surface of the earth cover above the tanks. This radiation level would require 60 and 69 inches (5 feet and 6 feet) of earth cover for the respective containers.

Radiation profiles of Purex LWW wastes in existing storage tanks were used to establish the dose rates at the roof of the vertical tank. The dose rates at the roof of the horizontal tank were increased proportionately for the liquid level being nearer the crown of the shell. A dose rate of 10,000 and 17,000 R/hour at the ceiling was used for the vertical and horizontal tanks, respectively.

Spectrographic data taken in Purex in July 1959 and December 1959 were used to determine the percentage of dose rate for each contributing isotope. Adjustments were made for spectrum "hardening" to approximate the increase on percent of the higher energy components in spectrum above the liquid surface resulting from self-absorption of gamma rays within the liquid of the tank contents.

The following table summarizes these percentages of energies:

Approx. Energy (MEV)	Percentages of Dose Rate	Dose Rate from Energy	
		Vert. Tk.	Horiz. Tk.
0.14	5	600	850
0.50	10	1200	1700
0.75	75	9000	12,750
1.4	5	600	850
2.5	5	600	850

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Fig. 3 - Shielding Requirements Curves

These curves have been developed from the same data as Fig. 2, using a gross gamma radiation level of 0.1 mr/hour at the ground surface above the tanks. In the design of various tank sizes and geometries, the structure requirements of a particular secondary container will alter the required thickness of the concrete shell. Fig. 3, is provided to aid the structural design engineer to select the required combinations of earth shielding and either plain concrete (Sp. Gr. 2.3) or concrete (Sp. Gr. 3.5) made with borytes aggregate which together will provide sufficient shielding to meet the design requirements.

For example, if the secondary container required 24 inches of plain concrete (Sp. Gr. 2.3) for structural support, then the additional thickness of earth would be 57 inches to reduce the gamma radiation level to 0.1 mr/hour at the surface of the ground. The resulting earth cover would then be five feet for design purposes.

FIG. 3. SHIELDING REQUIREMENTS CURVES

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Fig. 4 - Preliminary Schematic Flow Diagram

This diagram illustrates the principle conceptual functions for the interim storage requirements for high-level, radioactive liquid wastes which would be neutralized and concentrated sufficiently to support self-boiling. These functions and requirements have been defined with the text as a bases for an engineering study to determine the economical tank geometry and size, and tank farm layout. Conclusions attained from this study could provide the bases for the scope design of a new tank farm.

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(50 GAL/TU FINAL STORAGE VOLUME)
 FIG. 4 PRELIMINARY SCHEMATIC FLOW DIAGRAM

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