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SEVEN YEAR WASTE PROGRAM

CHEMICAL PROCESSING DEPARTMENT

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December 15, 1958

HANFORD ATOMIC PRODUCTS OPERATION
Richland, Washington

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SEVEN YEAR WASTE PROGRAM
CHEMICAL PROCESSING DEPARTMENT

INTRODUCTION

This seven year program covers liquid and solid radioactive waste disposal from the several facilities in the Chemical Processing Department. There is also a very active separate program on the gaseous effluents from the processing operations, both for the removal and the recovery of radioactive and chemical constituents in these gaseous streams.

The liquid wastes are classified as: (1) solvent extraction column wastes which contain appreciable quantities of radiomaterials and are stored in large underground tanks, (2) steam and process condensates which contain only trace quantities of radiomaterials; these condensate wastes are discarded into cribs from which the water percolates into the ground with the accompanying absorption of residual radiomaterials by the soil, and (3) process cooling water which is discharged to a swamp from which it seeps into the ground or is evaporated, though not normally contaminated, the process cooling water is monitored to detect possible equipment failure.

Solid wastes include failed equipment from the processing plants, filter cloths, wiping rags, etc. At the present time most of these solid wastes are buried in the ground.

To date, in the Chemical Processing Department, the waste storage and disposal operation has been handled primarily as an adjunct to the production plants with the principle research and development interests for these plants being directed toward improving the production processes for separation and recovery of product. These present waste disposal methods have been based on limited investigation of the several parameters of the waste disposal problem and were selected primarily because of their immediate availability, combined with a relatively high degree of safety and reasonable construction and operating costs. Observation and inquiry on the waste program have thus tended to operate only within a predetermined conceptual framework; i.e. storage in liquid form in underground tanks for the high activity level, relatively low volume per ton wastes; and disposal to ground of lower activity, relatively high volume per ton wastes. The possibility and probability of the superiority of alternate methods of waste disposal has long been recognized but the pressing problems of fuel separations and recovery precluded any but the most cursory examinations of these alternates. Initially the Hanford Atomic Products Operation was considered only as short term. Today, however, production planning is on a long range basis and it is apparent that fission product wastes will continue to be produced for years to come. It is thus timely to re-examine the total liquid and solids waste storage and disposal program for the separations area.

In comparing waste disposal methods, there is a tendency to limit the inquiry to only the initial construction and operating costs. Actually, on some waste programs, the continuing need for monitoring and storage replacements, years

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after the last waste has been discharged or handled, can exceed the total initial construction and operating cost incurred for the facility. Similarly, for methods of waste handling with significant attendant risk, the related actuarial or insurance protection charge can prove to be the controlling cost in long-term waste disposal considerations. The prudent choice of the final waste disposal practice will be made in recognition of the above cost facets coupled with the broad public responsibility aspects of this problem which will weigh against selection of disposal means having significant risks.

This proposed waste disposal program is based on the philosophy that the lowest total eventual cost for waste disposal will be one in which a good deal of emphasis is placed on reducing (1) the required future monitoring cost, (2) the required future storage replacement cost, and (3) the actuarial cost. In order to accomplish this it can be expected that the combined near-term construction and operating costs may be more than those incurred by present disposal practices. The actual knowledge of monies involved in future monitoring and actuarial costs is very limited. However, these areas of costs are getting closer scrutiny not only at Hanford but also throughout the world-wide nuclear program. A detailed study on this important phase of waste disposal is a vital part of this program.

OBJECTIVES

This proposed seven year waste program covers the development and implementation of waste disposal methods for the Chemical Processing Department, both as interim answers to present plant needs, and as long-term waste disposal methods which will minimize total long-term disposal costs. These total long term disposal costs include not only the usual construction and operating costs for the disposal facility but also cover the two additional facets of the waste disposal cost picture: these are (1) the combined monitoring and storage replacement costs to assure continuing integrity of disposal not only for the present but also for perhaps as long as several hundred years following the last operational use of the facility; and (2) some sort of insurance premium, which is termed actuarial cost, to cover the risk potential of either a catastrophe or some lesser unexpected hazardous condition developing at a future date.

The program objective for the high activity liquid wastes is to convert them to an inert solid form suitable for indefinite storage with no need for re-handling. With the current practice of storing high activity wastes in liquid form in underground tanks, re-handling will be necessary on a recurring basis during the hundreds of years the wastes must be contained. Re-handling of materials is not good practice in any field of production, and this is especially true for highly radioactive materials. The final storage place for the inert-solid, high-activity wastes will be constructed so as to provide long-term, high integrity storage and containment of these waste materials with no need for re-handling.

The program objectives for the liquid wastes currently deemed sufficiently low in activity for disposal to ground are to: (1) delineate the sources of activity contaminants for these waste streams, (2) markedly reduce the total activity in these streams by in-plant process, equipment, and/or operating modifications.

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(3) develop and install a pilot plant process for removal of the majority of activity from a specific stream, (4) evaluate the economic and public responsibility aspects for using the demonstrated process on other effluents, and (5) investigate the long range economic aspects of stepwise reductions in the water addition to ground in both the cribs, which receive process and steam condensate, and the swamps, which receive cooling water.

The program objective for the coating-type wastes is to convert these liquid wastes to essentially a solid form in the existing underground storage tanks. It is postulated that some of the older, high-activity waste which is currently stored as a neutralized solution may also be treated in a manner similar to these coating-type wastes.

The program objectives for the solid-type wastes are to: (1) recover product where economically feasible from filter cloths, wiping rags, etc. and (2) review in detail burial procedures for failed equipment and other solid-type radioactive wastes.

PROGRAM

The goals of the proposed program are based on the premise that the lowest total eventual cost for waste disposal will be one in which a good deal of emphasis is placed on reducing the required future monitoring cost, the required future storage replacement cost, and the actuarial hazards cost. This program, as is true for any forward-looking program, is based on current information. For each of the items there is an implied flexibility for the appraisal and future adaptation of these goals to the constantly evolving knowledge of the waste disposal parameters from both the safety and cost viewpoint.

The total expenditures including research, development, start-up, and capital costs for this program are shown in Figure I. Details on the research and development costs on a yearly basis are shown in the body of the report starting on page 20 and on Figure XXIII. The peak yearly research and development expenditure is in 1963 and is 25 percent of the historical yearly average cost for research and development as sponsored by the Chemical Processing Department, with the average yearly research and development cost for this seven year waste program being only 16 percent of the historical average.

On Figure I (Order of Magnitude Costs - Constant Worth Basis - 1959 Dollar):

<u>Item 1</u> - Expanded Leak Detection - $600,000.
A leak detection system is being evaluated for tank 113-SX. This item covers the possible extension of leak detection to 15 other tanks.

<u>Item 2</u> - Condenser Water - Purex - $200,000.
This provides an emergency water supply from deep wells to sustain controlled tank farm operation during emergency outages of the main water system and is to be ready for use in 1959.

<u>Item 3</u> - Interim Storage Tanks - $3,000,000.
This is to provide 5 - 150,000 gallon stainless steel tanks designed to

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provide a tank within a tank containment. It is proposed to build the first of these five tanks beginning in 1961 with the remaining four being constructed starting in 1962. There is a two-fold advantage in doing this: (1) initial storage of acidic wastes will be required starting about 1962 to provide well-aged wastes for testing purposes in the inert conversion facility starting in 1964; and (2) actual construction of one of these new type storage tanks will provide valuable information for construction of the balance of this tank farm.

Item 4 - Solid State Containment Facility for the High Activity Wastes. The development cost shown as $3,000,000 covers the research and development program for the solid-state process. The capital costs for $4,000,000 include the processing facilities which are to be located in B Plant. The start-up costs of $1,500,000 would be incurred during the years 1964 and 1965 after which the facility would be turned over to production. This program would be extended to treat the present tank-stored neutralized Purex waste sometime after 1965.

Item 5 - Five Year Storage Solid Waste - $4,000,000. This is to provide a storage facility by 1965 for the high activity solid wastes from the Purex Plant which will be adequate to meet production needs for at least five years and perhaps as long as fifteen years. The wastes may be aged in interim storage tanks or treated directly in the inert conversion facility. HW-58240 reports on the economic factors for handling current wastes.(1) Studies are continuing to delineate more clearly the relationships of thermal conductivity of the material, storage geometry requirement, heat liberation rates, etc. For example, the thickness of stored solid waste may be more than doubled if the wastes are aged 4 years prior to conversion to a solid form. The economic advantage of increased container size though must be balanced against the need for interim storage space.

Item 6 - Miscellaneous Items - $200,000.

The sub-total covering the high activity phase of the waste program is $16,500,000.

Item 7 - Process and Steam Condensate - $4,200,000. This covers all the liquid effluents from the production plants except for the cooling water. The $600,000 per year cost, or $4,200,000 for the next seven year period, is in line with the cost for cribs and auxiliaries assuming the present program of ground disposal is continued. However, the specific types of future disposal facilities for these intermediate wastes will probably be modified as results are obtained from the continuing technological and economic studies. As an example of what may evolve from these studies, it may be found that plutonium bearing wastes from the Finished Product Operation may be economically treated by concentration followed by either recovery of the plutonium or processing to an inert solid in the inert conversion facility.

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Item 8 - Prototype For Activity Removal - $600,000.
The initial judgment basis for selecting ground disposal to handle these intermediate level wastes has been altered by the maturing process technology and by the more stable production demands in the primary plants. In addition to technical and economic evaluations, the continuing studies in this area will include the program for pilot plant evaluation of an optimized facility for removal of the majority of activity from a specific stream. Significant results are expected from this program by 1962.

Item 9 - Plutonium Solids Waste Recovery - $350,000.
This is scheduled for FY-1959. A project proposal covering design only has been approved. An economic pay-out of 1-1/2 years, based on the value of the recovered plutonium, is estimated for this facility.

Item 10 - Cooling Towers - $1,750,000
This is for both the Redox and the Purex facilities with the initial cooling tower unit being scheduled for 1962 and the balance scheduled for 1965. In addition to improving the certainty of containment of activity already in the ground, the cooling towers will provide a water supply independent of the export water system.

Item 11 - Organic Disposal - $100,000.
This consists of a combustion chamber and off-gas filter for disposing of the waste organic liquid and is scheduled for 1963.

Item 12 - Miscellaneous Items - $600,000.

Item 13 - Solidification Program - $100,000 for development and $400,000 capital expenditure. The initial unit is scheduled for installation in 1963 with subsequent facilities provided in 1964 which would be adequate to handle all the coating-type wastes currently in underground storage. Extension of this program to include the older, high salt content, high activity wastes already in storage would be after 1965.

The grand total for this seven year waste program is $24,600,000 and the yearly average is $3,500,000.

The total waste disposal cost per unit of production for this future seven year period is essentially the same as the cost for the current period. However, in addition to meeting the production needs for waste disposal during this next seven year period this program will provide major additional benefits.
(1) It will provide process facilities to convert high activity wastes to an immobilized solid state as contrasted to the present method of holding these wastes indefinitely in the highly mobile liquid state.
(2) It will provide storage facilities for the high activity wastes from the Purex operation which will be adequate to at least 1970 and perhaps to 1980.
(3) It will provide a leak detection system for both the Redox and Purex tank stored high activity wastes.

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(4) It will provide an improved integrity of interim storage for Purex high activity wastes by means of secondary containment through the tank-within-a-tank design concept.
(5) It will result in a markedly reduced demand on the vadose zone and thus preserve this natural resource as an extremely valuable secondary line of radioactive material containment for future Chemical Processing Department operations. The vadose zone is that portion of the ground lying above the water table.
(6) It will provide a separate cooling water system by means of wells and cooling towers for both the Redox and Purex operations which will be completely functional even if all export water to the 200 Areas is interrupted.
(7) It will provide economic plutonium recovery from solid wastes which previously had been disposed to ground.
(8) It will provide an economic means for organic disposal.
(9) It will convert millions of gallons of mobile liquid coating-type wastes to a relatively immobilized state in the existing storage tanks.

These goals for waste management within the Chemical Processing Department are in harmony with the long range objectives and philosophy of the Atomic Energy Commission as set forth by Joseph A. Lieberman, in his Geneva paper on waste management, and also, with the objectives and philosophy of the National Academy of Sciences, as set forth in their report on the disposal of radioactive waste on land.(2)(3)

HIGH ACTIVITY WASTE

On the high activity wastes, the initial storage was in liquid form in underground tanks such as the one shown on Figure II. There were as many as 10,000 gallons of liquid to be stored for each ton of uranium processed. The specific radioactivity of these solutions was much lower than present day high activity wastes and essentially no boiling took place in these tanks. An air-cooled, reflux condenser, as shown on the top of the tank, was adequate to take care of heat generation in excess of that lost to ground.

The trend, in the production plants, has been for smaller and smaller quantities of radioactive waste per unit of material processed with the result that, at both Redox and Purex plants, the wastes have sufficient fission product concentration to support self-boiling for several years. Considerable savings in capital investment for storage space have resulted from allowing these boiling wastes to self concentrate. The principle design modifications to handle these boiling wastes has been to provide a condenser and off-gas system for removing the water vapors from the tank, and to provide internal circulators to reduce hot zones with their accompanying "bumping" problem.

Figure III shows a typical tank currently being used to store these boiling radioactive wastes. Note that circulators and a condenser are provided. The 75-foot diameter of the original tank design is retained and the volume increase is obtained by essentially doubling the depth of the liquid. This increased depth of stored liquid provides the potential for a markedly larger accumulation of energy in the stored wastes. The internal circulators have proven very successful in dissipating the energy stored as heat in the solution. However, an additional hydrostatic head limitation has been placed on the use of these storage tanks to preclude the possibility of generating sufficient internal pressure to result in structural damage to the tank.

Figure IV shows a typical plan view of a high level waste storage facility. The wastes from the production plants are routed to the individual storage tanks through the diversion box shown at the upper left of the figure. Tank nozzles for temperature, liquid level, liquid transfer, agitators, and spares are provided for each storage tank. There is a common vapor line header for removal of the water vapor or low pressure steam. The vapor passes through a cyclone for gross separation of entrained liquid and then into a condenser with the non-condensibles going to a particulate filter prior to being discharged to the stack. There is a by-pass on the off-gas system with a seal pot to provide both a vacuum relief as well as a pressure safety system for the tank farm.

Figure V shows a typical section view of the same waste storage facility. At the present time the Purex tank farm contains six, one-million gallon storage tanks and the Redox SX Farm contains fifteen one-million-gallon storage tanks. There are 9 other tank farms with a total of 108 underground storage tanks similar in design to the one shown in Figure II. At the present time, there are 15 full tanks of this type containing 10,537,000 gallons of high activity wastes of a non-boiling nature, 60 full tanks containing 27,882,000 gallons of waste of a medium activity level and high inert salt content such as coating-type wastes, and 33 tanks of this type which are essentially empty or contain only a small heel of material.

Figure VI shows the trend in final high activity waste volume per ton of uranium processed. The T Plant high activity wastes in 1950, shown at the left of the graph, contained 10,000 gallon per ton. By 1953 this had been reduced to about 4500 gallons per ton which was about the minimum waste volume attained for the bismuth phosphate operation. The Redox plant start-up was in 1952 with an initial waste volume per ton of about 4,400 gallons or roughly equivalent to the best performance of the bismuth phosphate plant. By 1953, this had been reduced to 3,200 gallons per ton followed by subsequent volume reductions down to the present time when the waste is running about 500 gallons per ton.

The initial Purex operation was in 1956 and the waste volume was about 900 gallons per ton during the start-up period. This was reduced to about 300 gallons per ton in 1957, with indications that the volume per ton will be significantly less than 100 gallons as a result of the present planned program. Thus the waste volume from one ton of uranium has been reduced to only a few percent of what it was in 1950 which has resulted in a tank utilization improvement of better than 50 to 1. The design requirements have

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become markedly more demanding for the storage system in order to contain these much more highly concentrated radioactive wastes. However, costs for the complete tank farm are up by only a factor of about 4 (including the inflation factor) as contrasted to the tank utilization improvement of 50 to 1.

In addition to the handling of the water vapor from these highly concentrated wastes, the self boiling phenomena introduces a second problem. For the older waste, an inventory is kept for all tanks and this is considered adequate as a means of detecting any leakage of wastes from these tanks. However, with the boiling wastes, it is impossible to detect even large quantities of waste loss to the ground by inventory differences. For example, a typical self boiling tank at Purex can boil off 300 to 500 thousand gallons per month. Tank 103-A in the Purex farm has boiled off about six million gallons of water in a two year period. Assuming that all material fed into the tank and all liquid removed could be accurately measured, a 1 percent gross error on tank 103-A alone would be 60,000 gallons during this period. It is for this reason that a supplementary leak detection system is being considered for the present high activity waste tanks in both the Purex and Redox tank farms.

Figure VII shows a section view of one type of leak detection system which is being considered for all tanks containing self-boiling wastes. The initial installation of this leak detection system is currently being made for tank 113-SX in the Redox farm. The system consists of a vertical caisson, shown on the left, 12 feet in diameter, sunk to a depth approximately 10 feet below the concrete footing of the storage vessel. A 4-inch schedule 40, stainless steel pipe, which serves as a monitoring channel, extends out under the tank. Figure VIII shows the spider arrangement of five monitoring channels that might be used for each tank for the Redox farm. Each caisson is located so as to serve three underground storage tanks. Only nine of the fifteen SX tanks will contain self-boiling wastes.

Figure IX shows a similar arrangement for the Purex tank farm consisting of two caissons, with accompanying monitoring channels, for the six storage tanks of the present Purex farm. In addition to the protection provided from the hazards viewpoint by being able to detect a leak, this detection system may allow extension of the useful life of the storage tanks by improving the predictability of storage integrity.

Figure X shows the tank concept currently visualized for the new Purex tank farm scheduled for about 1963. Considerable engineering effort is yet required before the storage tank design can be formalized. These tanks will be used for interim storage to allow the wastes to age prior to converting them to an inert solid state. This horizontal design concept provides a tank within a tank and does not rely upon the environs for containment of any high level activity. This design incorporates sufficient flexibility to provide for the addition or removal of wastes, cooling water, condensibles, and air from the annular space. The function of the heel pit shown above the tank on the left, is to transfer the stored waste to solidification facilities or to transfer the wastes in the event of failure of the inner tank.

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Stainless steel is specified for both the inner tank and as a liner for the outer annulus in order to provide for the storage of acidic wastes. One contemplated flow sheet on waste treatment consists of a formaldehyde kill of the nitric acid prior to discharging the wastes from the processing plant. The effluent stream from the formaldehyde kill process would be about one-half normal in nitric acid. This relatively small concentration of nitric could be further reduced by partial neutralization if the economic balance on corrosion and service life of the interim storage tanks justified this step.

Heat removal from the sludge layer would normally be provided through natural convection air currents. Cold air is taken into the annulus from the top of the structure as shown at the far right of the figure. Three replaceable circulators are also included in this 150,000 gallon storage tank. Total overall length of the structure is 130 feet and the diameter of the inner tank is 16 feet.

Figure XI shows the product which has been made from simulated wastes in a test spray column in the 300 Area. Note the spherical shape of these particles and the almost complete absence of dust. These particles are 150 to 200 microns in diameter and may be remelted and cast into larger shapes for packaging.

Figure XII shows a schematic arrangement for this conceptual spray column process for converting high activity waste to an inert solid form. The spray column shown on the left is 4 feet in diameter by 12 feet long. Liquid feed, at 75 gallons per hour, is mixed with 30 cfm of air for atomizing. Heat is introduced to the product from the heated side walls to effect calcination of the atomized spray feed before it contacts the vessel side wall. The temperature of the product leaving the bottom of the spray column is 750° centigrade. It is cooled to about 200° centigrade and then fed into a cyclone separator. The hot waste product is sent to the packaging operation at the rate of 100 pounds per hour or about 12 percent by weight of the entering feed. The air and steam vapor from the wastes pass through the cyclone separator to a heat exchange surface in an evaporator. The non-condensibles are withdrawn from this heat exchanger by means of a venturi scrubber jet activated by the condensate pumped under high pressure into the jet nozzle. A knock out separator is used to de-entrain the majority of the condensate which flows back to the evaporator for re-evaporation. The non-condensibles are filtered and passed to the stack. It must be kept in mind that this spray column is a <u>concept only</u>, albeit a promising one. Most commercial types of spray driers are heated directly by the flow of hot combustion gases inside the vessel. The proposed unit, by heating the feed material by radiation from the heated vessel walls, eliminates many of the problems attendant with handling the off-gases in a spray column operation. There would be no moving parts in this spray column with the possible exception of the feed nozzle.

There are several other promising methods for calcining radioactive wastes and several of these have received and are still receiving considerable attention both at Hanford and off-site. Phillips Petroleum at Idaho Falls is concentrating on the fluidized bed calciner. This process consists of injecting the

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liquid wastes into a heated agitated bed of feed particles. As is true of all calcining operations, the off-gases and vapors from the fluidized bed require both a means for particle cleanup as well as a scrubbing, condensing, and filtering operation prior to release to the atmosphere. A pilot plant calciner is now under construction at the National Reactor Testing Station with scheduled completion being early in 1960. Progress on this test facility, as well as work at other sites, will be followed closely for potential application to the problems at Hanford.

A third possible type of calciner is a rotary kiln. Studies on waste calcination in a screw type kiln have progressed to the pilot plant stage at Brookhaven National Laboratory.

A fourth possibility for calcining the high activity wastes is to utilize the heat of radioactive decay to evaporate the wastes to dryness and also to aid in heating the resulting dried product to sintering or fusing temperatures. This process has been studied at Oak Ridge. Heat balances are critical. However, by insulating the waste container, process cycle times can be markedly reduced and a process may be made practical by this means.

Several organizations, both in the United States and the other countries are investigating the ultimate disposal of radioactive wastes by mixing and sintering with earthen materials. Atomic Energy of Canada Limited and the United Kingdom Atomic Energy at Harwell are evaluating these. Personnel at MIT, ORNL, and Los Alamos are also investigating ceramic glazes, shales and glass respectively. However, the principle effort to date has been directed toward utilizing materials containing high concentrations of alumina and silica. The waste and solids are mixed in either batch or continuous equipment to form a thick slurry which is fired at about 1,000° centigrade to form a fused ceramic mix. Either internal heat, due to fission product decay, or heat from an external source is utilized to achieve the required sintering temperature. Flux may be added to reduce this temperature, or, in the case of a Purex type waste, the sodium nitrate may serve as a flux.

This program for conversion of high activity waste to an inert solid will require the major portion of the development effort for the entire Hanford waste program. The basis of current consideration is that those inert conversion processes which merit intensive development will be delineated in calendar year 1959. Intensive laboratory development will be carried on from 1960 to 1962, along with intensive pilot-plant studies. These pilot plant studies will be enlarged in scale and carried through June 1964. The installation of a full scale prototype in B Plant is targeted for completion by June 1964.

Figure XIII shows a schematic layout of this inert conversion facility in the B Plant. This layout is highly tentative in nature and is merely to give an indication of the magnitude of the operation. Ten cells are shown as being required for the waste receiving system, the feed tank, the spray dryer and cyclone cell, plus the necessary scrubbing, filtering, and treatment facilities. Note that the solid packaging operation is shown only as a line below the

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cyclone. It is visualized that at least two additional cells in B Plant will be required for this packaging operation. Following packaging the inert solid wastes will be sent to a new solid storage vault which is to be constructed as part of this program.

This inert conversion plant will not interfere with the program for recovery of specific fission products. Feed for the specific fission product recovery process would come from the interim storage tanks and the waste residue from the specific fission product recovery operation would be sent to the waste receiving tank of this inert conversion plant.

DISPOSAL TO GROUND

The major part of ground disposal is the liquid wastes currently deemed sufficiently low in activity for disposal to the Vadose Zone; the Vadose Zone being that portion of the ground lying above the water table. The program objectives on this disposal are to (1) delineate the sources of activity contaminants for these waste streams, (2) markedly reduce the total activity in these streams by in-plant process, equipment, and/or operating modifications, (3) develop and install a pilot plant process for removal of the majority of activity from a specific stream, (4) investigate the long range economic aspects of processes for removing radiomaterials from plant liquid effluents, and (5) investigate the long range economic aspects of stepwise reductions in the water addition to ground in both the cribs, which receive process and steam condensate, and the swamps, which receive cooling water.

The Chemical Processing Department is especially favored by the great thickness and relatively good ion-exchange properties of the Vadose Zone sediments. In the past six years, emergency production commitments have necessitated disposal of large quantities of radiomaterials to this area. Today, however, with production capabilities and demand being in relatively good balance and process technologies being quite mature, more conservative ground disposal practices can be economically utilized. The amount of activity being disposed to ground has already been reduced to less than 20 percent of prior levels; in-plant improvement programs are scheduled to reduce this to less than 2 percent of prior levels by 1960.

It is postulated that the maximum long-term benefit from this unique natural resource, the Vadose Zone, may be obtained by preserving it, where economically feasible, as an extremely valuable, secondary line of radioactive material containment for Chemical Processing Department operations.

Over the past fifteen years the production plants have gone through several stages. Initially the primary separations method was the batch precipitation process in the bismuth phosphate plants. This was followed first by the Redox solvent extraction operation and subsequently by the Purex process. Each of these processes have been subject to further evolvement as the basic flow sheets have been modified and the plants made more productive and more efficient. The result has been that, in the waste streams emanating from each of these facilities,

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there have been numerous changes in concentration, flow, and activity level.

Early in 1945 it was recognized that, in the next few years, millions of gallons of trace activity liquid wastes would be discharged from the recovery plants. It was also recognized that a firm design basis for treatment facilities would not exist until the plant operations had been refined to the point that the streams would have a relative stability of chemical constituents and activity level.

To plan for disposal of these low-activity waste streams, through technological programs for removal of the activity, would have required essentially a freezing of the flowsheet within the plants. This would have been inadvisable since it essentially would have limited the in-plant technology to the existing levels. It is well known, of course, that, due to the freedom for technological change, which was not inhibited by pre-set disposal practices for the waste streams, numerous capacity increases and marked cost reductions have been effected within each of these processes.

In 1945 there were essentially three avenues open for disposal of the low activity waste stream; (1) storage in tanks as is, (2) concentration prior to storage, and (3) disposal to ground. It was decided at that time to initiate a program for capitalizing on the known high ion exchange properties and great thickness of the vadose zone underlying the separations area. In the intervening years, this disposal to ground program for the low activity liquid wastes has been highly successful and has resulted in major savings as contrasted to the cost which would have been incurred for either underground storage per se, or concentration coupled with underground storage based, at least, on the technology known in 1945.

The low NGS plutonium program which was initiated in 1953 is a good example of the relative importance of disposal-to-ground methods. This low NGS plutonium program markedly increased the uranium throughput for the separations plants. The production demand requirement was so critical that, in addition to establishing new production loads for uranium throughput at T Plant and Redox, considerable funds were expended at B Plant to return it to operating condition.

With total stored wastes from the T Plant operation in excess of 4,500 gallons per ton, underground storage space was critical during this period. Just how tight the situation was is indicated by the schedule for underground storage tank 101-C. The metal waste was removed from tank 101-C on May 15, 1953 and on the same day tank 101-C started receiving wastes from the TBP recovery facility as no other storage space was available. A minimum of one year's time would have been required to provide additional underground tank storage.

A related program to the low NGS plutonium was the program to recover uranium for feeding back into the AEC complex. During this period, there was a critical shortage of uranium resulting from the fact that the low NGS program required only a low exposure level in the reactors and thus high uranium throughput. Every reasonable means was taken to increase production throughputs and uranium

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recovery in the metal recovery operation. This was done both to feed uranium back to the production stream and also to clear underground storage tank space to provide for the new uranium wastes from the T Plant operation on the low NGS program.

Initially, disposal of the wastes from the TBP recovery operation had been back into tanks which had originally contained the uranium bearing wastes. These wastes, by concentrating within the plant, had been held to about 5,500 gallon per ton of uranium recovered or about 1000 gallons more volume than that associated with each ton of T Plant uranium waste. This volume was further reduced to about 4,000 gallons by concentrators located at the BX and TX tank farms. However, the solids contained in these doubly concentrated wastes was so great that they could not be pumped. It was necessary to have this second-stage concentrating facility located within the immediate vicinity of the storage tanks. Timing-wise, the two tank farms equipped with waste concentrating facilities, BX and TX, were filled with highly concentrated wastes just as the low NGS plutonium program was starting. Thus, if the low NGS plutonium and uranium recovery programs were to go ahead at the rate required, new means would have to be found for disposal of the wastes from the TBP recovery operation.

It was indeed fortunate for the overall weapon's program that scavenging of tank stored metal recovery wastes had been receiving some Laboratory attention. The scavenging process consists of adding precipitate forming chemicals to the waste solution to form a precipitate which has an affinity for certain elements including the long half-life fission products. These are carried down with the precipitate leaving the supernatant liquid relatively free of undesirable radiomaterials. This process offered the hope that the immediate, emergency need for underground storage space could be met by disposing of the supernatant liquid to ground. Development efforts were put on an accelerated schedule. Successful prototypical testing of waste scavenging in the metal recovery plant was accomplished in time to allow planned disposal of large quantities of waste supernatant liquid to the ground on a specific soil retention basis as required by production demands. By this means, sufficient underground tank storage was made available so that there was no hold up on either the low NGS program or the uranium recovery program.

This emergency need for uranium in the AEC complex was also reflected in numerous changes in the TBP recovery operation to increase throughput, with quite major changes in flowsheets and methods of waste disposal being made frequently. It is a noteworthy achievement that all during this period there was not one instance where the disposal to ground practice did not result in reasonable and safe containment of the wastes and its associated activity.

Figure XIV shows the activity disposed to the ground since plant startup. The activity is expressed as kilo curies of gross beta. From plant startup until July 1953 the total activity disposed to ground was 100,000 curies as shown by the bar on the left. The succeeding bars on the chart represent the activity disposed to ground for each fiscal year. The Metal Recovery operation started in November 1952 and was completed in March 1957. Note the sharp reduction in activity disposed to ground in fiscal year 1958.

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Today the technology of the plant processes is at a quite advanced state and production capabilities are in good balance with demands. Thus much greater assurance can be provided for predicting stream composition and activity levels both for the present and for the longer range future. This fact, coupled with the rapidly developing technology on activity removal from liquid waste, such as mineral bed processes, makes the re-assessment of the total waste disposal to ground program especially appropriate at this time.

A two pronged approach to this problem is proposed: (1) sources of activity contaminants for these waste streams will be delineated, and, by in-plant process, equipment and/or operating modifications, the total activity in the waste streams will be reduced; and (2) an optimized process will be established for treating a specific waste stream to remove the majority of activity. There is an active program underway on the in-plant phase of this program and the markedly reduced activity to ground forecasted for 1959 and 1960 will be accomplished by the in-plant reduction of total activity contained in the effluent streams. Note that the forecasted amount for 1960 is less than 10 percent that for 1958. These reductions will be made in both the primary plants on fission product removal and the finished product operation for plutonium removal from the plant effluent streams.

The pilot plant will be installed to remove the majority of the remaining activity in one of the effluent streams. This pilot plant will serve to optimize the design and economic parameters for this effluent cleanup facility. The ultimate goal for effluent cleanup will be established as additional knowledge is gained on the many economic, safety and political aspects of this problem.

One of the dangers which is recognized in ground disposal is that something new or different will be put on top of the soil column already saturated with radioactive materials. For example, oxalate put into the soil would complex plutonium and make it mobile. Likewise solvents in general can be expected to upset the soil balance. Any complexing agent such as many of the cleaning fluids make for bigger molecules which then migrate relatively easily through the soil structure. A vital part of this waste program is to physically isolate all abandoned crib or disposal areas to preclude inadvertant addition of foreign materials to the soil in these areas.

Ground disposal practices at Hanford have been based on controlled disposal. Figure XV shows a cross section of the general Hanford area with the 200 East and West Areas located near the highest elevation. The top light zone is the fluviatile, high permeability, sand and gravel structure. The next lower darker zone is Palouse soil and the black band lying directly beneath this zone is a calcium carbonate layer termed caliche. The vertical lines on this section represent monitoring wells.

Figure XVI shows a schematic cross section representing the various waste disposal facilities with the volumes and gross activity that have been sent to ground from each of these facilities up to February 1958. The swamps have received 1.2 X 10^{11} liters containing 2,000 curies of fission products. The 18

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trenches have received 10^8 liters of solution containing 640,000 curies of fission product. The 70 cribs have received 1.2×10^{10} liters containing 1.7×10^6 curies of fission product. The disposal of activity to trenches has been mainly from the waste scavenging program for the metal recovery operation and has been on a specific soil retention basis, which is limited to about 100 gallons per square foot. In cribs, disposal of as much as 20,000 gallons per square foot has been accomplished.

Figure XVII shows the type of spread of water beneath a crib. Note that in the upper section there is little spread of water due to the fact that this section is a high permeability, coarse sand and gravel area. As the water encounters the first darker zone, which consists of a much lower permeability material, the water tends to spread laterally. In actual practice the spread may be several fold more than is indicated on this figure. After passing through this low permeability layer, the water gradient is again almost vertical until it encounters the next low permeability zone where it again spreads laterally. The different shades beneath the crib are to denote the boundary layers for different isotopes.

Figure XVIII shows the penetration by the various radioactive materials. With most separation's type waste, the soil shows the highest affinity for plutonium followed closely by the rare earths. Strontium tends to move somewhat further through the soil and cesium penetrates the furthest of the long, half-life isotopes. Ruthenium, which has a relatively short half-life, leads all the other radioactive materials in movement through the soil, and is thus the first radioisotope detected in well samples taken adjacent to and beneath crib disposal sites. However, as shown by the slide, the nitrate ion precedes even the ruthenium ion and is used as a good lead indicator of crib utilization and life.

Figure XIX shows the contours on the water table as of January 1944. Note that the maximum water table elevation is at 390 feet beneath both 200 West and 200 East Areas. If all water disposal to ground ceased in these two areas it can be expected that the water mounds would slowly recede and the water table would eventually approach that shown on this figure.

Figure XX shows the water table as of February 1958. Note that the water mound in West Area rises to 470 feet or 80 feet above the natural water table. In contrast to this the water mound beneath East Area is at 410 feet or only 20 feet above the natural ground water yet in volumes disposed to ground, significantly more water has been discharged in East Area than in West. This difference in water mound contours is a good indication of the relative permeability of the soils beneath these two areas.

Test well samples in a limited area under the disposal sites have shown 1.5×10^{-7} micro curies per cc (μc/cc) gross beta contamination detected in the ground water. The maximum permissible concentration for drinking water has generally been taken as 1×10^{-7} μc/cc for gross beta from an unknown mixture of fission products. The permissible concentration for ruthenium is 1×10^{-3} μc/cc or 10,000 times greater than for mixed fission products. Since ruthenium is the leading isotope to reach the ground water, the activity detected in the ground water under the disposal sites is only a small fraction of the permissible limit. It is postulated that all activity currently in the vadose zone can be essentially immobilized by eliminating the outflow of water from these areas. As part of this waste disposal program it is planned to investigate the long range economic aspects of stepwise reductions in the water additions to ground in both the cribs and the swamps. The final balance which will be considered in this study will be to utilize cooling towers in place of swamp disposal and specific treatment facilities in place of cribs.

Figure XXI indicates the long-range future water table balance that may exist. The process and steam condensates would be decontaminated and recycled to the processing plant. The condenser cooling water would flow through the cooling towers for heat removal and be recycled to the plant. Make up water, to replace that lost through evaporation and in cooling tower blowdown, would be from deep wells located near the processing plant. The activity in the blowdown stream from the cooling towers would be removed either by the natural soil or by a treatment facility. This long-range future water balance may possibly show an actual water flow into the area beneath the processing plants as contrasted to the present large water flow away from these areas. The potential for migration of the activity already in the ground would be markedly reduced by a future water table balance such as this. Economic factors, coupled with the developing knowledge of the hazard cost or insurance risk factor, will guide the decisions on this ground disposal balance.

The process and steam condensate effluents represent only a small part of the total volume yet contain 99.9 percent of all activity being disposed to ground. Processes which might be used in place of ground disposal for decontaminating these effluent streams include: (1) multi-effect evaporation, (2) precipitation filtration, and (3) solid fixation.

(1) On multi-effect evaporation, the total activity content of these streams has been steadily decreasing and this program proposes marked future improvements also as is indicated in Figure XIV. These reductions in total activity reduce the decontamination factor required for satisfactory clean-up of these streams. A single-effect evaporation of the feed stream coupled with multi-effect vapor utilization can result in reasonable costs for this method. The low total salt content of these streams will cause only a small boiling point rise and thus allow good temperature differential optimization.

(2) On precipitation-filtration, ORNL reports favorably on the use of the lime-soda process for removal of fission products from aqueous streams. Construction was completed in 1957 at Oak Ridge on a plant capable of treating 500,000 gallons per day.

(3) Solid fixation includes specific mineral formation such as calcite removal of strontium from phosphate containing solutions. For example, the $CaCO_3$ structure of the soil mineral calcite reforms as a

calcium phosphate mineral called apatite. Any ions, such as strontium, which are similar to calcium are included as part of the new crystal structure. Preliminary studies have shown that Pu^{+4} will also enter into this reaction. It is hypothesized that all radioactive "bone-seekers" will behave similarly.

Similar work is in progress on cesium removal. Hydrated alkali alumino-silicate minerals of the zeolite family such as clinoptilolite ($RO.Al_2O_3.10SiO_2.6H_2O$) and analcite ($RO.Al_2O_3.4SiO_2.2H_2O$) have the common property of cation exchange under appropriate conditions, and were found to be highly selective for cesium even in the presence of much larger concentrations of extraneous interfering cations such as Na^+ or Ca^{++}. These natural zeolites have many advantages over synthetic exchange resins including greater stability, higher cesium selectivity in most cases, comparable exchange capacities, and the further possibility of fusing them for permanent fixation.

COATING-TYPE WASTES

The term coating-type wastes is to include not only the standard wastes from removal of fuel coats but, also, all separation's wastes of a significant activity level, but yet sufficiently low in activity concentration so that solidification on a massive scale would not result in undue temperatures. In addition, these wastes normally have a high inert salt content. The program objective on coating-type wastes is to install a prototypical unit for converting these liquid wastes to essentially a solid form in the existing underground storage tanks followed by subsequent production operation of this process.

At the present time there are 60 full underground storage tanks containing 27,882,000 gallons of this type waste. There are also about 250 gallons at Purex and 150 gallons at Redox of coating type wastes produced per ton of Uranium processed. There are 32 empty underground storage tanks available for storing this type waste. Production schedules indicate that this storage space will handle the production requirement for coating-type wastes to at least 1965.

There are also 15 full underground storage tanks containing 10,537,000 gallons of neutralized high activity wastes of a non-boiling nature. These particular high activity wastes are several fold larger in volume per ton than present day production plant wastes from either Purex or Redox and also contain several times the inert salt content. However, in addition to being much lower in specific activity at time of processing than present day wastes, there has also been a significant decay period for the radiomaterials with resultant further reduction in specific activity: thus, future processing of these to an inert state may follow techniques found acceptable for treating the coating type wastes. The high activity wastes from the power fuels recovery program is also expected to have a high inert salt content and may possibly be handled in a similar manner. It is expected that, due to their high specific activity, current neutralized Purex wastes will have to be processed through an inert conversion plant in a manner similar to that for the acidic Purex wastes.

Figure 22 shows a schematic lay-out for a submerged combustion type facility which is being considered for reducing the liquid content of the coating type wastes. It is proposed to accomplish this within the underground storage tank itself. The process would consist of a submerged burner with the combustible gas and the necessary oxygen being fed to the burning chamber and the hot combustion gases being discharged beneath the liquid level. The immense vapor volume of the underground storage tank would serve as an initial de-entrainer with more complete decontamination of the off-gas being effected in the filter prior to the condenser. It is estimated that a volume reduction of two-thirds can be effected on these wastes, leaving a residue consisting of about 90 percent solids. Since there is no immediate production need, initiation of this test program will be delayed until about 1963.

SOLID-TYPE WASTES

At the present time almost all solid-type wastes are disposed of by burial. As part of this seven year waste program, the solid-type waste disposal methods will be reviewed. Where economically feasible, product will be recovered from miscellaneous contaminated materials such as wiping rags, filter cloths, etc. by means of a continuous-belt, muffle-type incinerator. The pay-back period for this facility is estimated as 1½ years, based on value of the product recovered. In addition, recent work on recovery of failed equipment is showing considerable promise. This work will be extended to consider major equipment pieces. Burial techniques will also be reviewed in detail.

RESEARCH AND DEVELOPMENT

Figure XXIII shows the research and development costs for this seven year program which are included as part of the total program cost on Figure I. The estimated number of research and development personnel are shown on the right in Figure XXIII and the order of magnitude costs are shown on the left in thousands of dollars. The circles joined by lines refer to personnel. The fiscal years are shown at the base. Personnel requirements are estimated to peak during 1961-1962 and to drop off during the later years of this program. During the peak period about 80 percent of this research and development effort will be on the inert conversion process for the high activity wastes. Dollarwise, the cost of this research and development program through 1962 will be mainly for laboratory and pilot plant studies. In 1963, $300,000 is programmed for larger scale prototypical equipment.

The current research and development effort sponsored by Chemical Processing Department totals about 3.3 million dollars in both FY-1958 and FY-1959. This program includes power fuel reprocessing, by-product recovery, waste containment, radiation control, exploratory research, and plant support research and development. Historically, similar programs have maintained a yearly average research and development expenditure of about 3 million dollars. Since 1945 the primary research and development programs have been the Bismuth Phosphate Process (1945-1946), the RG Line (1949), the RM Line (1952), Redox, Phase I (1952), Uranium

Metal Recovery (1952-1957), Uranium Oxide Plant (1952-1957), Expansion of 234-5 Facilities in 1955, Redox Phase II (1956), Purex Phase I (1956), and Purex Two-Cycle (1958).

The peak yearly research and development expenditure for this proposed waste program is $720,000 in 1963. This peak yearly amount is about 25 percent of the historical yearly average for research and development effort in the Chemical Processing field, with the average yearly expenditure for the seven year program being $480,000 or only 16 percent of the total research effort. This level of research effort for waste disposal is considered as a realistic share of the continuing research and development program in the Chemical Processing field. Financing of this research program will be from the 2000 budget. Related information will be developed under the programs sponsored by the Divisions of Biology and Medicine and of Reactor Development.

ACKNOWLEDGMENT

This seven year waste program represents the integrated effort of personnel in the Redox, Purex, Finished Products, Research and Engineering, Financial and Facilities Engineering Operations of the Chemical Processing Department and the Chemical Research and Development Operation of the Hanford Laboratories Operation. Special acknowledgment is extended to L. W. Finch, E. Doud and G. L. O'Neill, each of whom made major contributions to the development of this program.

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(2) Joseph A. Lieberman, The Role of Waste Management in the Development of the Nuclear Energy Industry - A/conf. 15/P/2004 Second United Nations International Conference on the Peaceful Uses of Atomic Energy, July 7, 1958.

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FIGURE I

SEVEN YEAR WASTE PROGRAM

Constant Worth Basis - 1959 Dollar

HIGH ACTIVITY WASTES

	THOUSANDS		BENEFICIAL USE
1. Expanded Leak Detection	$ 600		1960
2. Condenser Water Purex	200		1959
3. Interim Storage Tanks	3,000		1 in 1962 Balance in 1963
4. Solid State Containment			
Development	3,000		See page
Capital	4,000		1964
Start-Up Costs	1,500		
5. Five Year Storage - Solid Waste	4,000		1965
6. Miscellaneous	200		
Total		$ 16,500	

WASTES TO GROUND

	THOUSANDS		BENEFICIAL USE
7. Process and Steam Condensate	$ 4,200		As needed
8. Prototype for Activity Removal	600		1962
9. Plutonium Solids Waste Recovery	350		1960
10. Cooling Towers	1,750		1 in 1962 Balance in 1965
11. Organic Disposal	100		1963
12. Miscellaneous	600		
Total		$ 7,600	

COATING-TYPE WASTES

	THOUSANDS		BENEFICIAL USE
13. Solidification Program			
Development	$ 100		
Capital	400		1964
Total		$ 500	
HIGH ACTIVITY TOTAL		$ 16,500	
GRAND TOTAL		$ 24,600	
YEARLY AVERAGE		$ 3,500	

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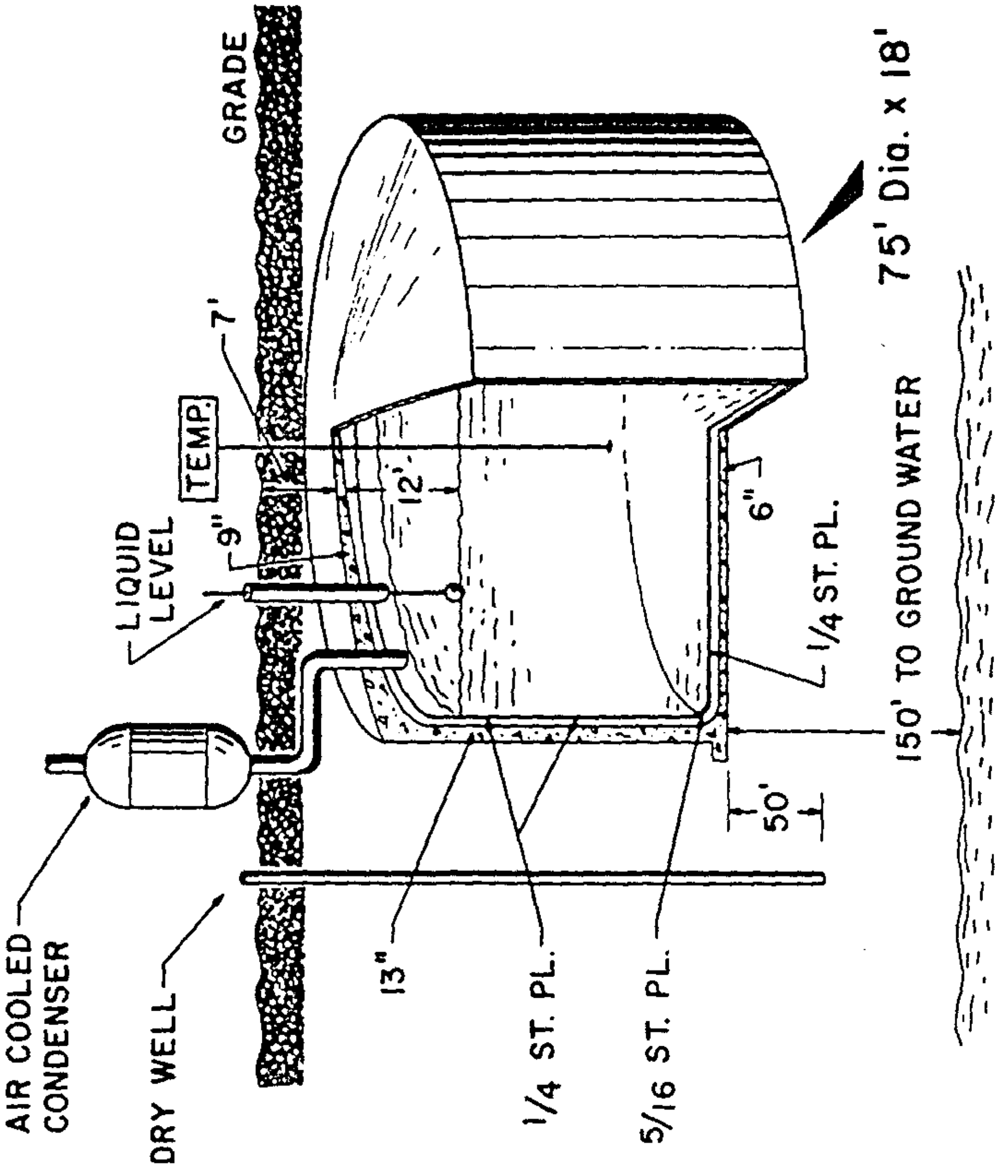


Figure 2
533,000 Gallon Waste Storage Tank

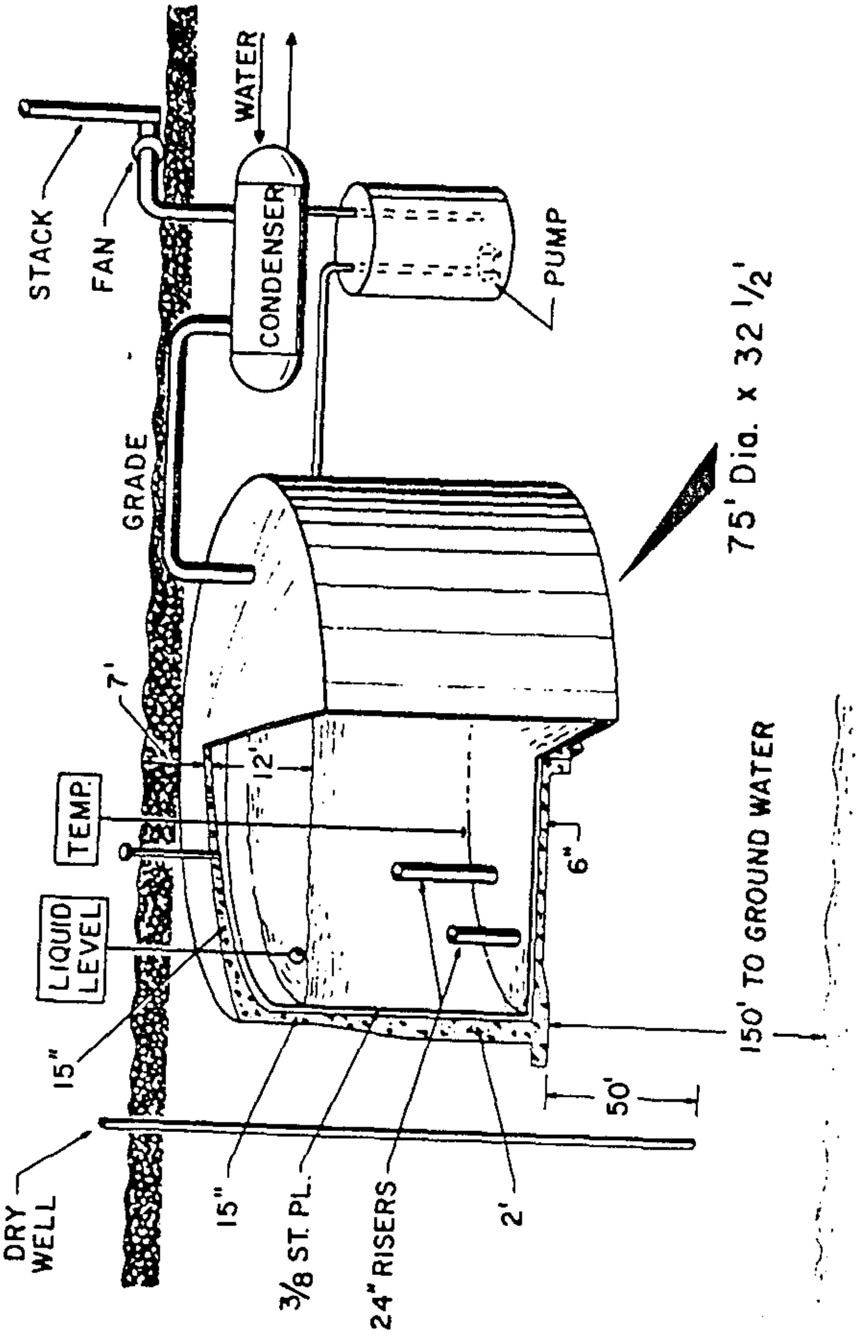
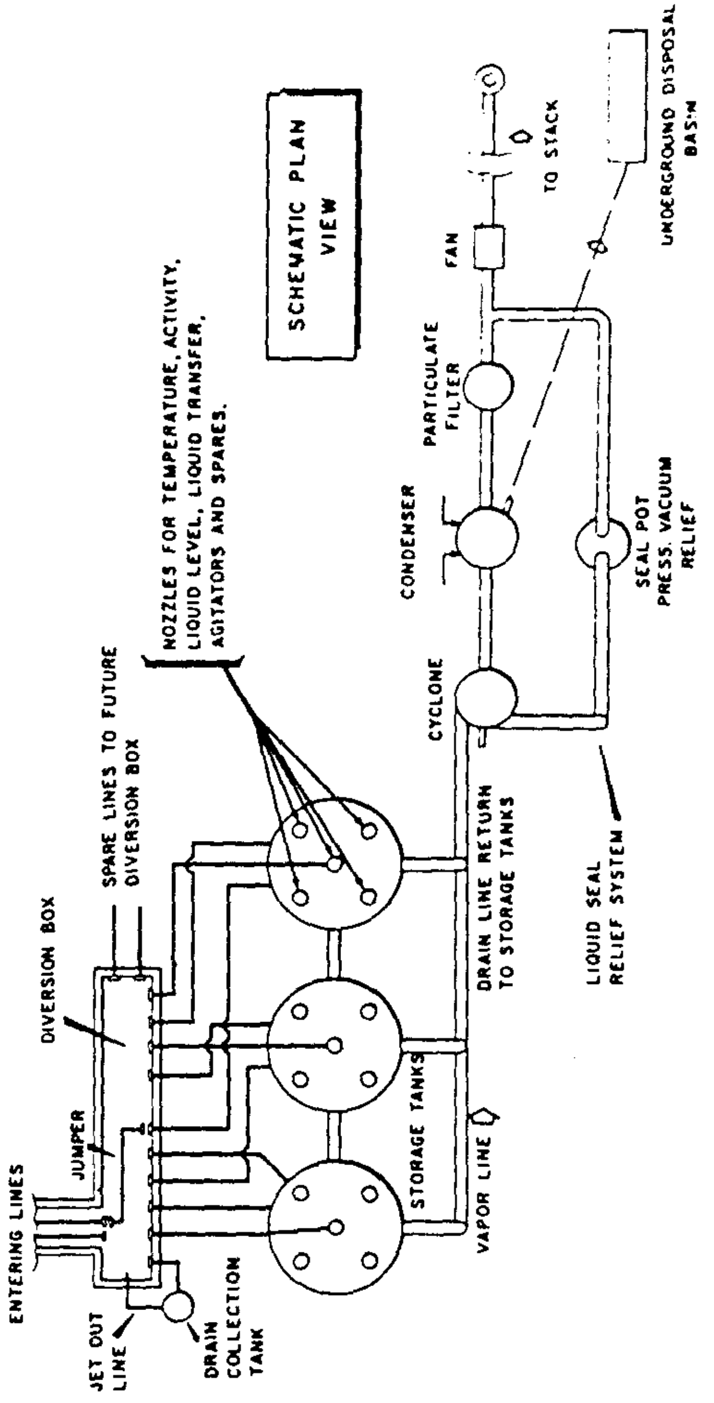
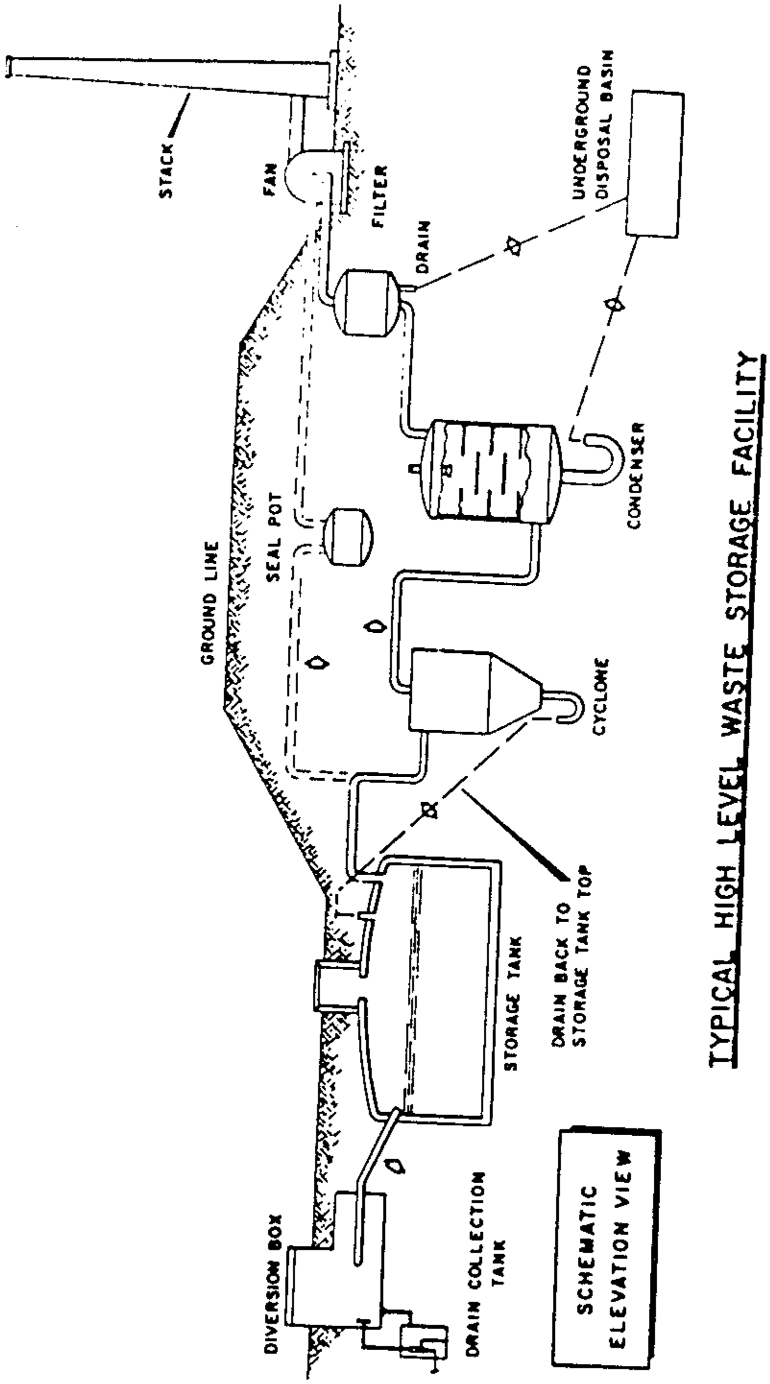


Figure 3
1,000,000 Gallon Waste Storage Tank



TYPICAL HIGH LEVEL WASTE STORAGE FACILITY

Figure 4



TYPICAL HIGH LEVEL WASTE STORAGE FACILITY

SCHEMATIC ELEVATION VIEW

Figure 5

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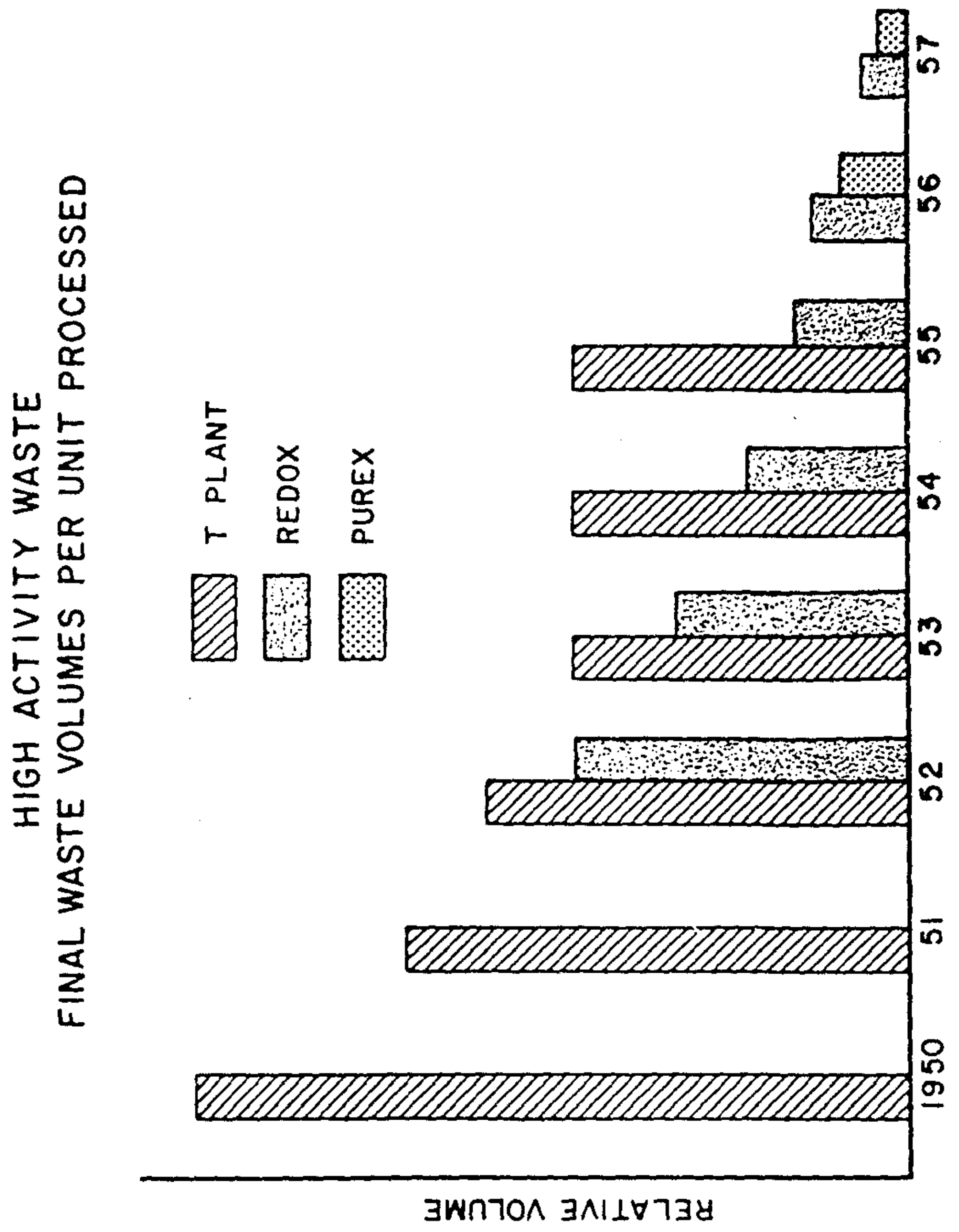

Figure 6

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LEAK DETECTION SYSTEM

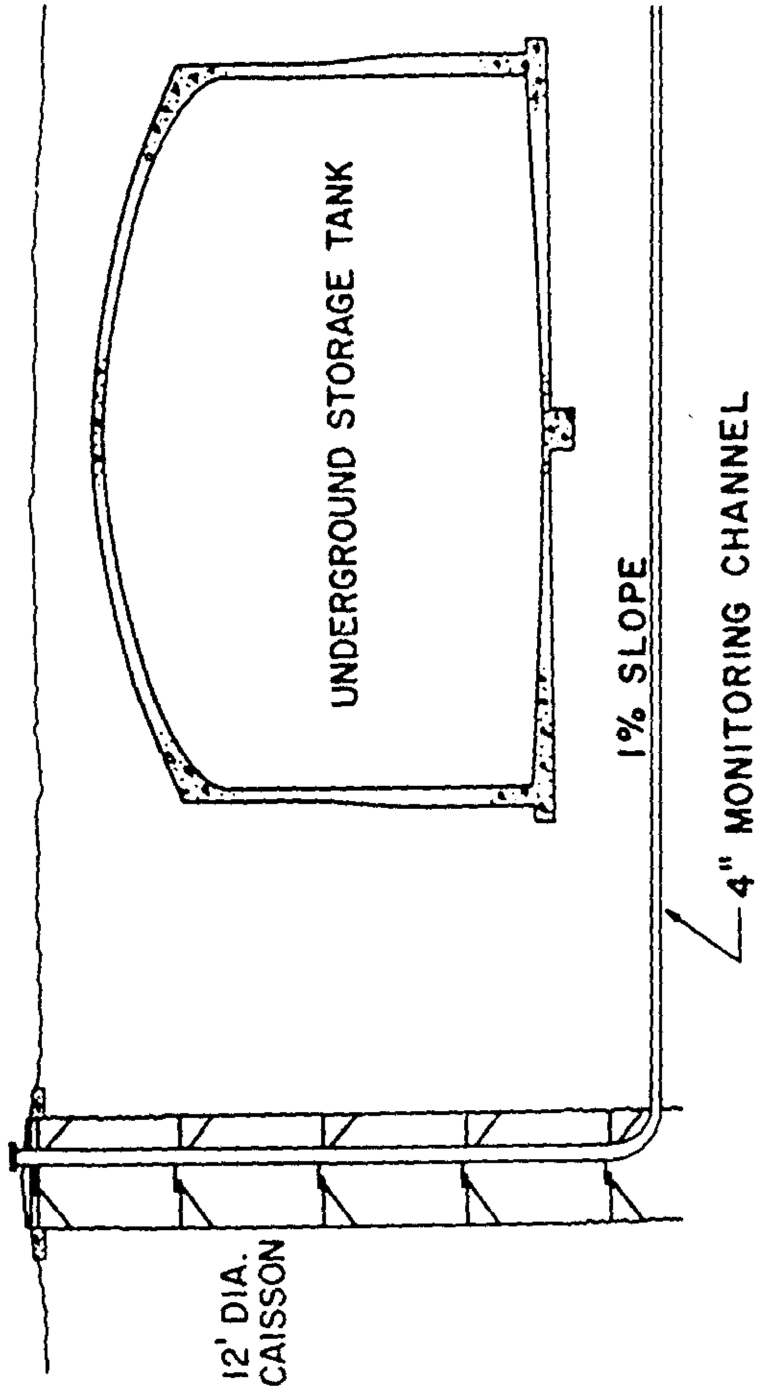

Figure 7

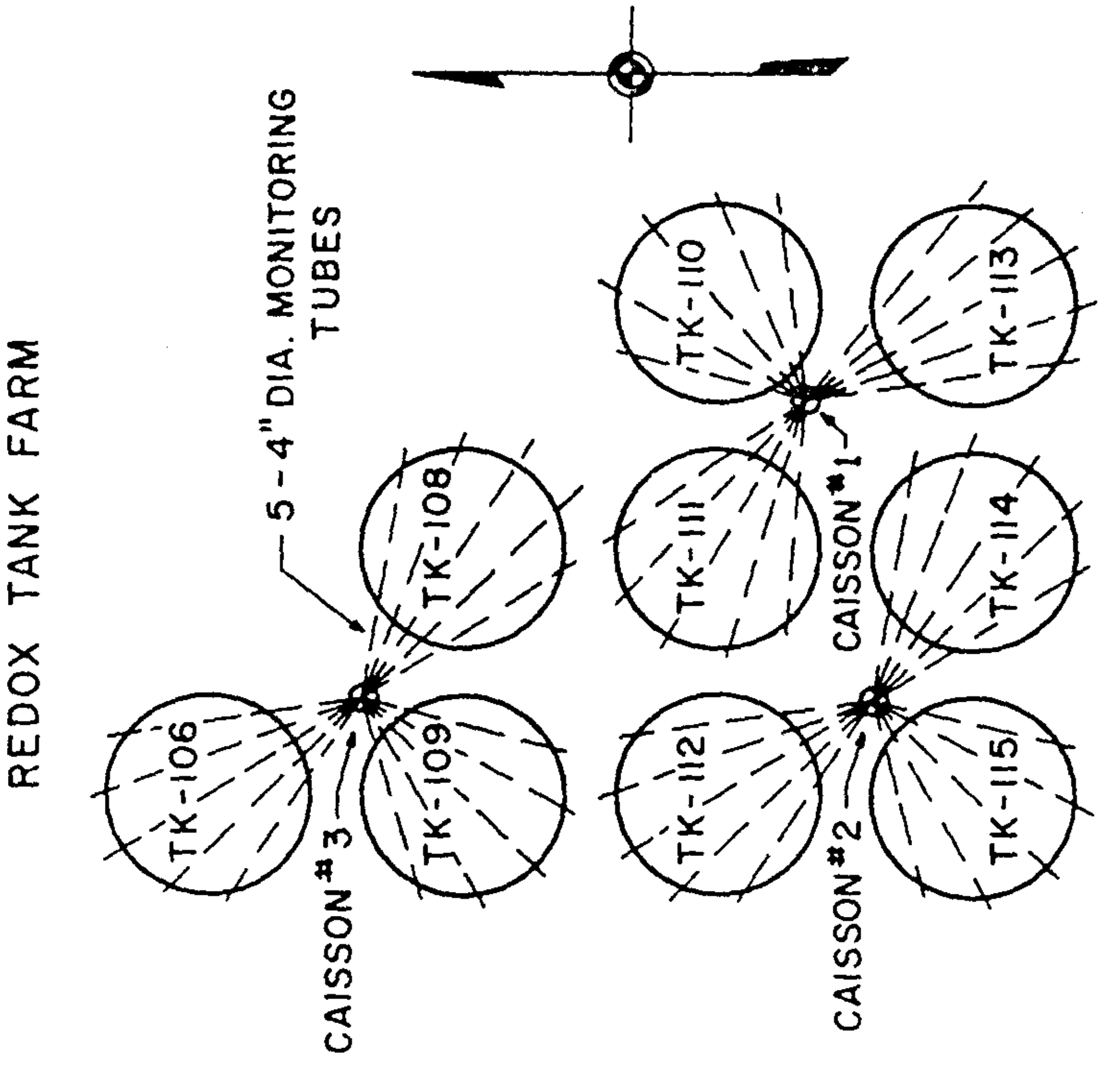
REDOX TANK FARM
5 - 4" DIA. MONITORING TUBES
TK-106
TK-108
TK-109
CAISSON #3
TK-110
TK-111
TK-112
TK-113
TK-114
TK-115
CAISSON #1
CAISSON #2

Figure 8

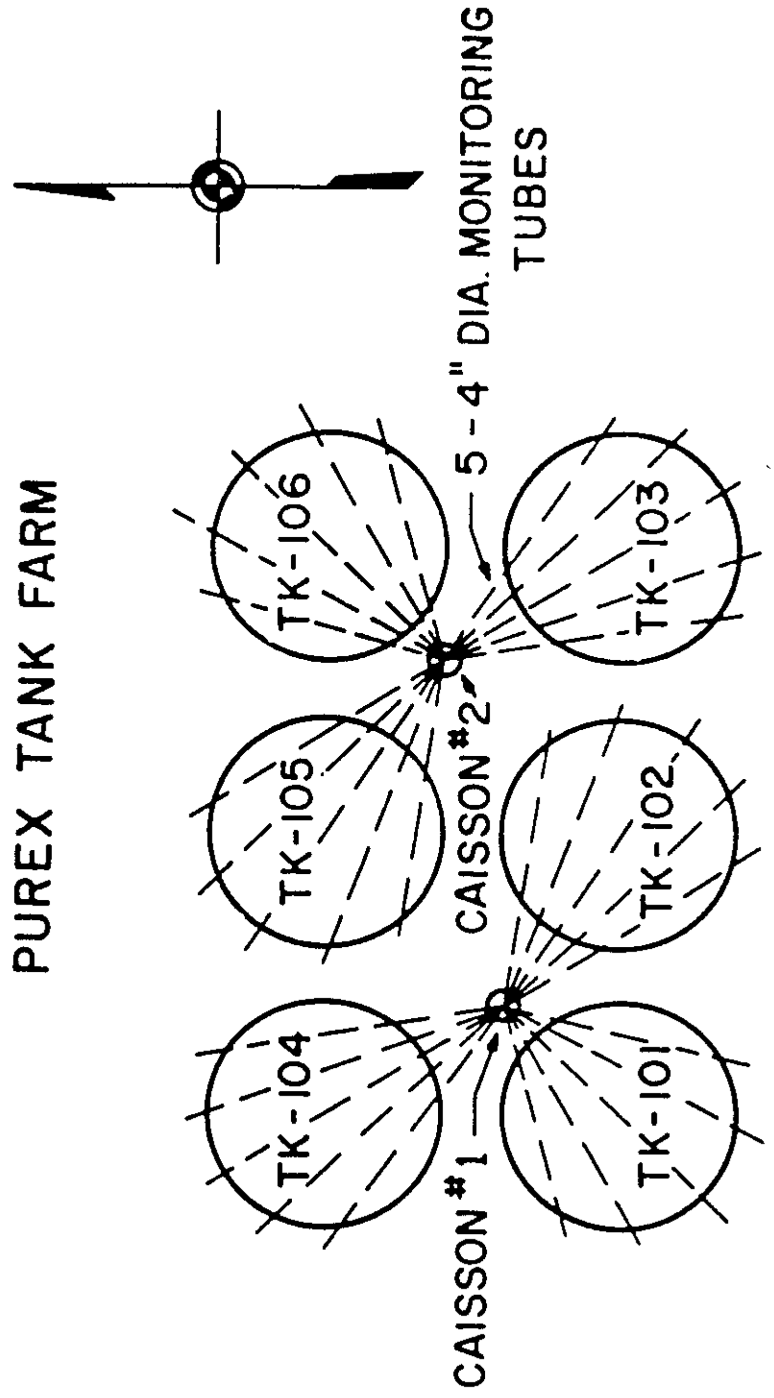
PUREX TANK FARM
TK-106
TK-105
TK-104
TK-103
TK-102
TK-101
5 - 4" DIA. MONITORING TUBES
CAISSON #2
CAISSON #1

Figure 9

INTERIM STORAGE VESSEL

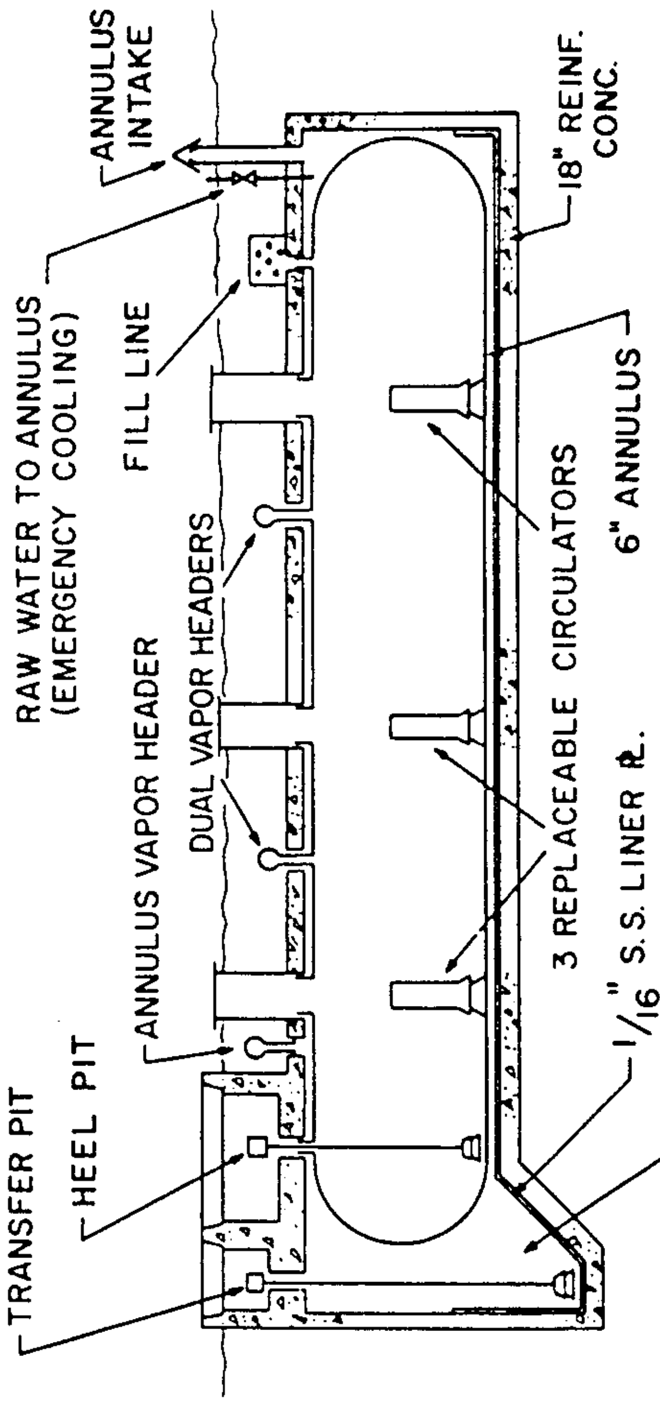

Figure 10

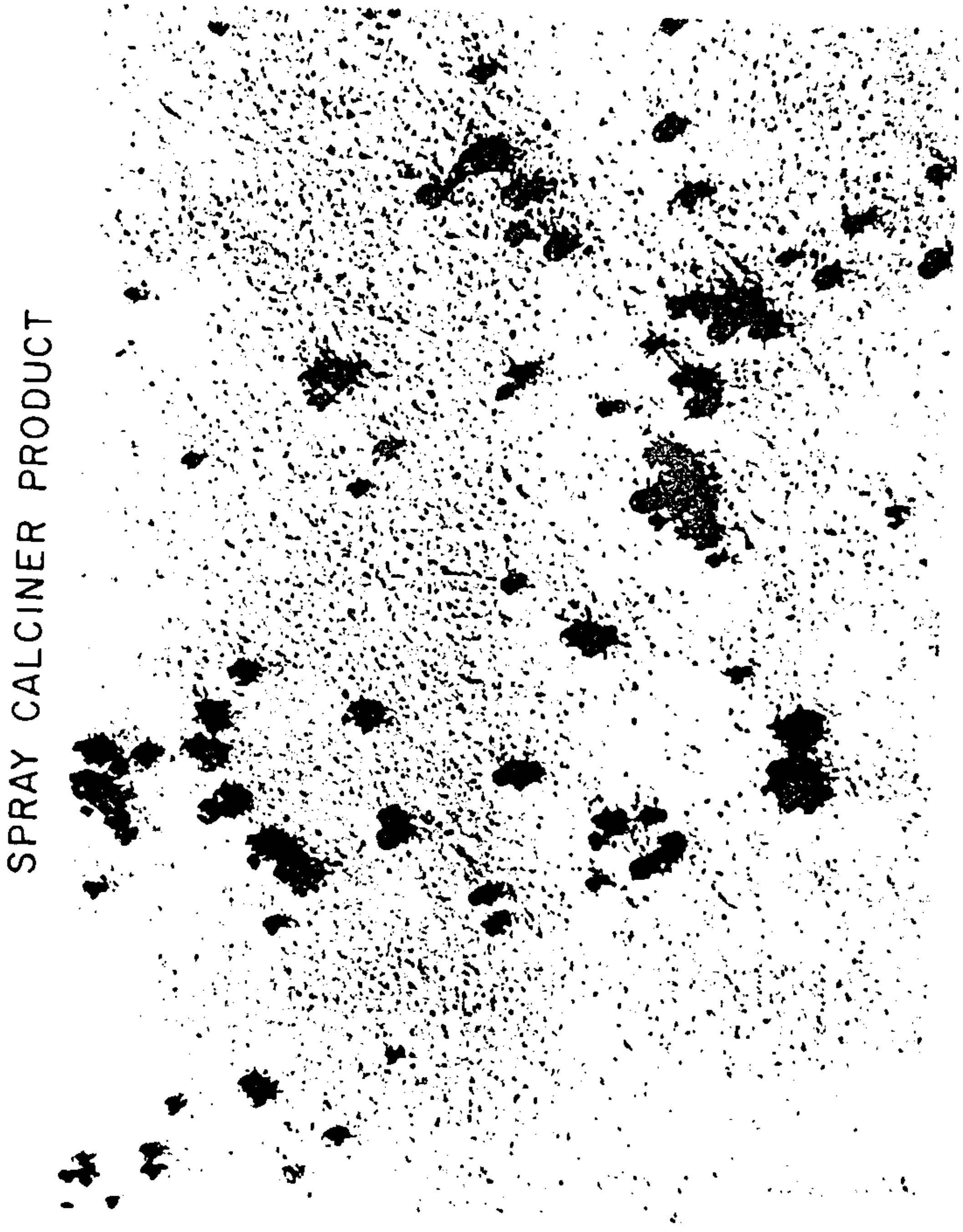

Figure 11

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SPRAY CALCINER

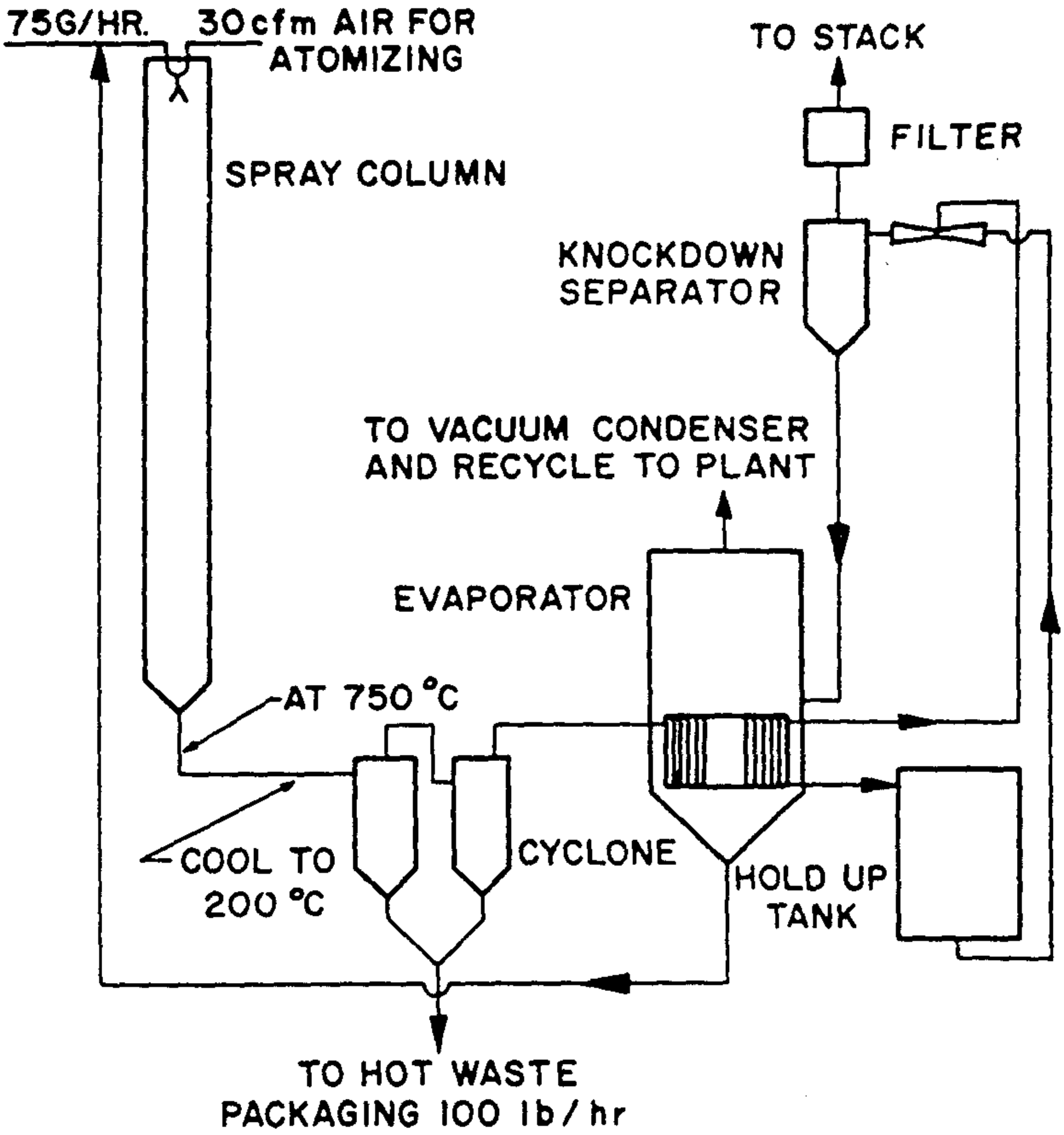

Figure 12

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SOLIDIFICATION

HIGH ACTIVITY WASTE INERT CONVERSION PLANT LAYOUT

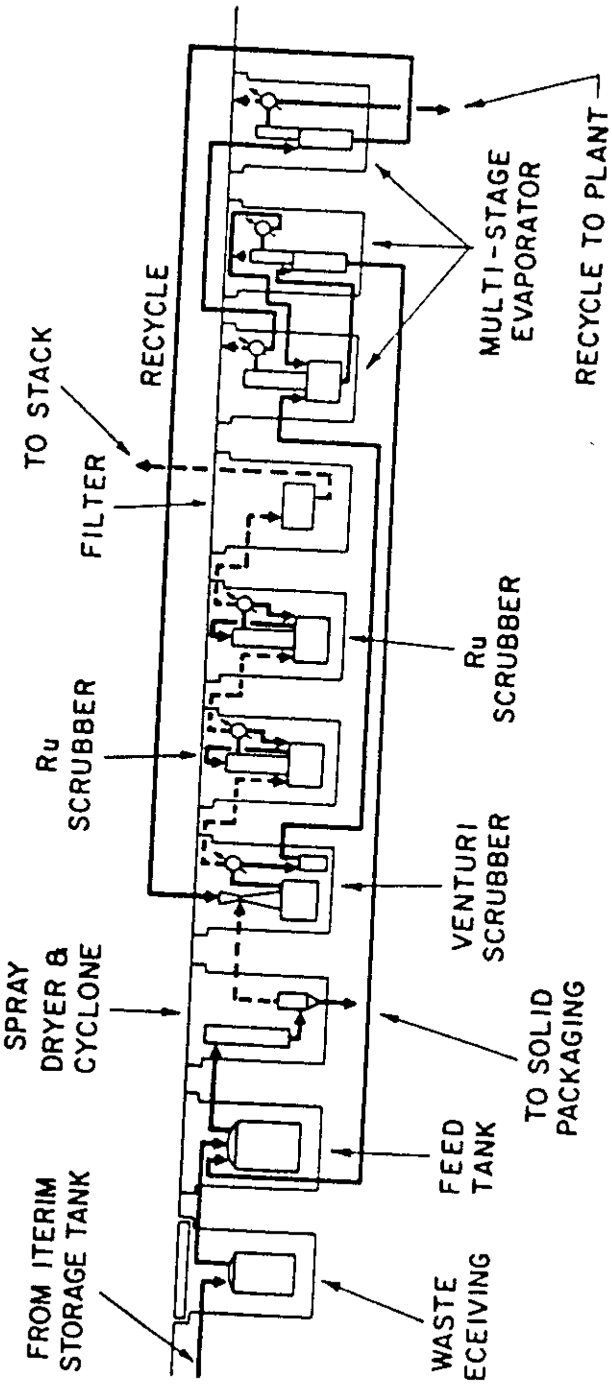


Figure 13

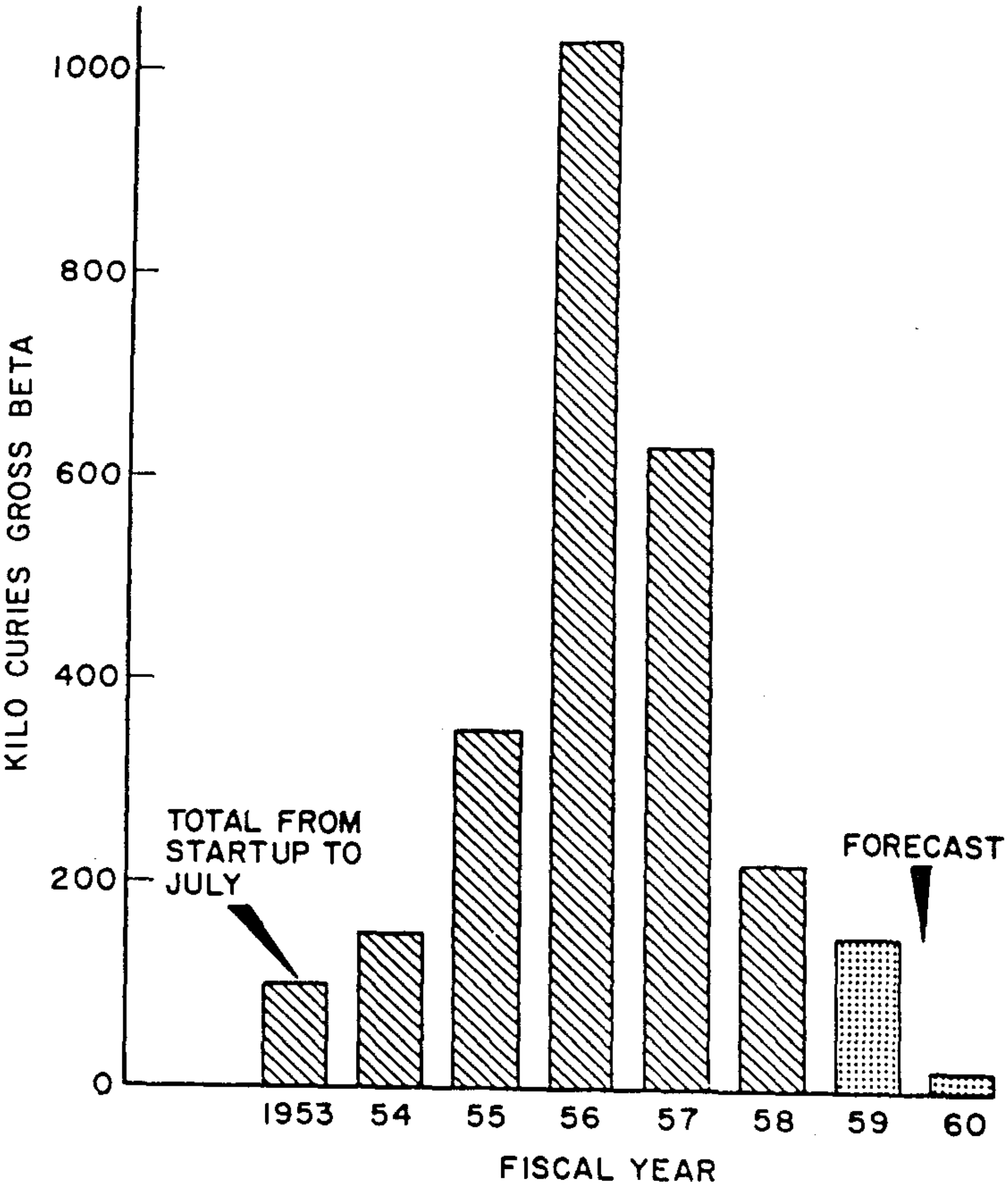

ACTIVITY DISPOSED TO GROUND

Figure 14

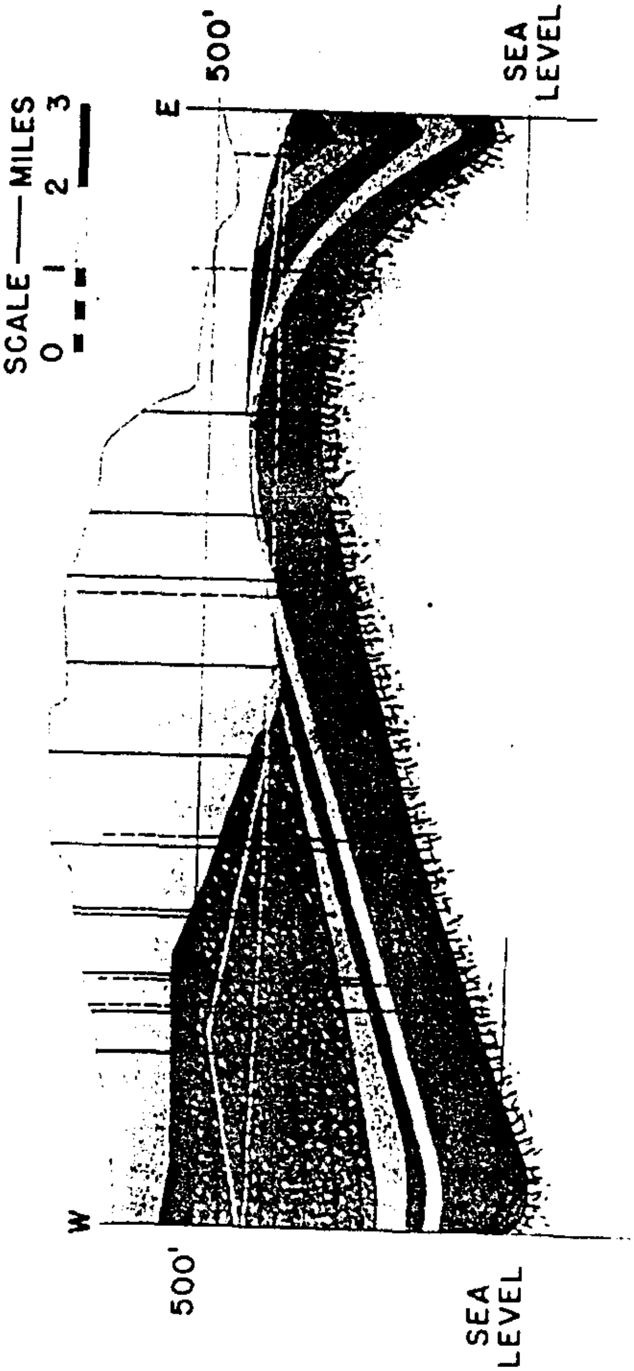
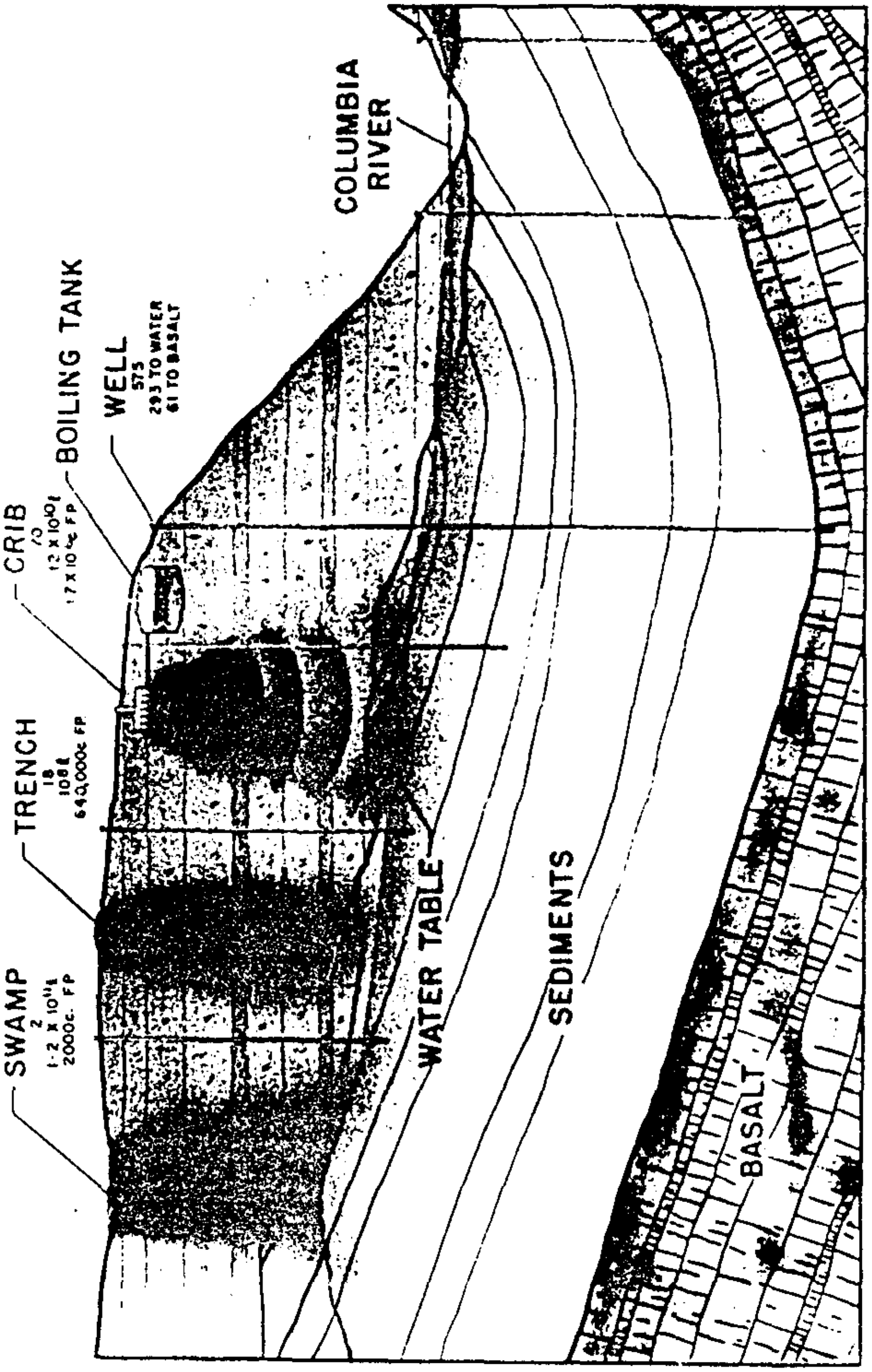


Figure 15



HANFORD SITE CROSS SECTION SCHEMATIC SHOWING WASTE DISPOSAL FACILITIES

Figure 16

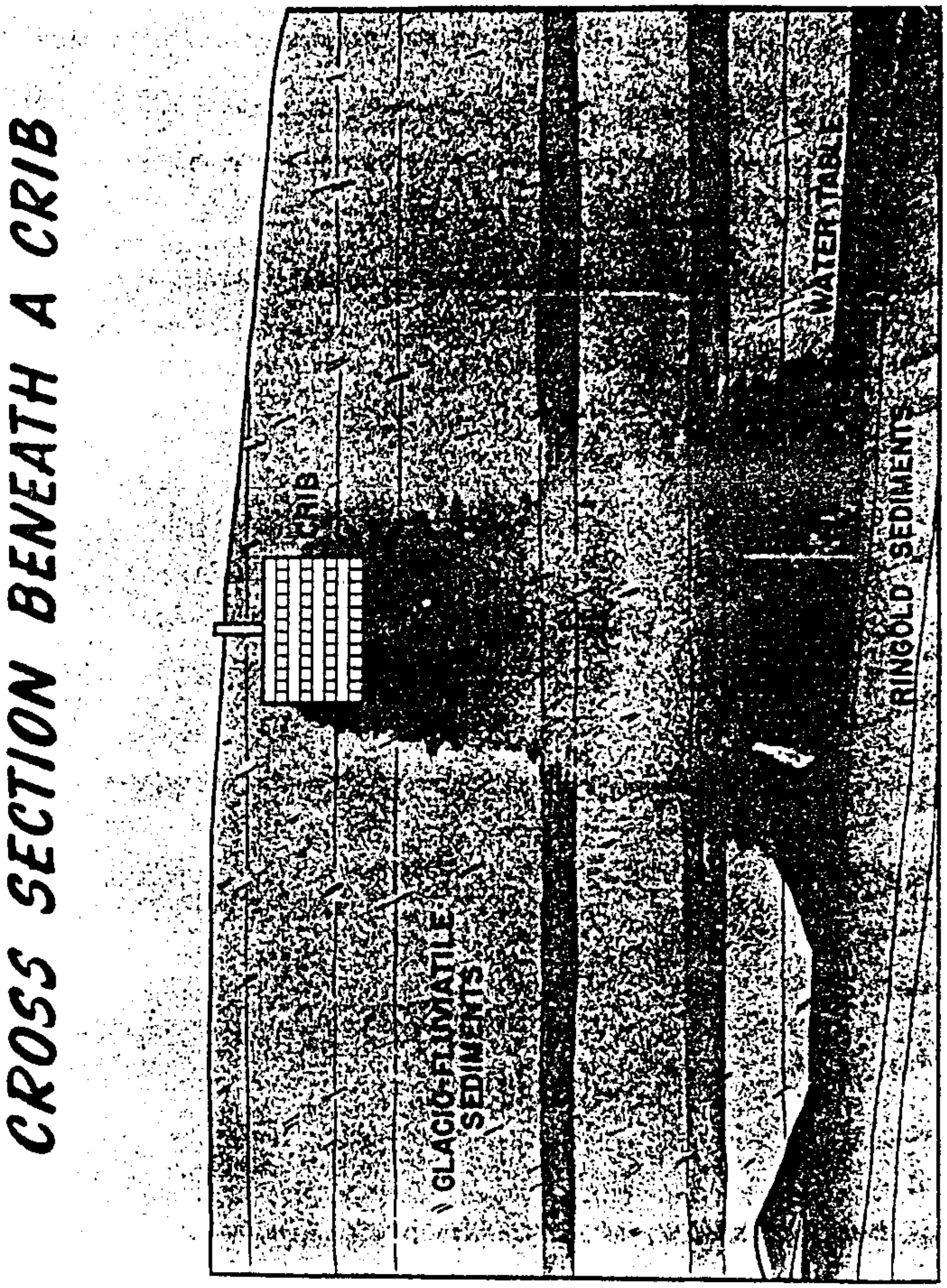

CROSS SECTION BENEATH A CRIB
CRIB
GLACIO-FLUVIATILE SEDIMENTS
WATER TABLE
RINGOLD SEDIMENTS

Figure 17

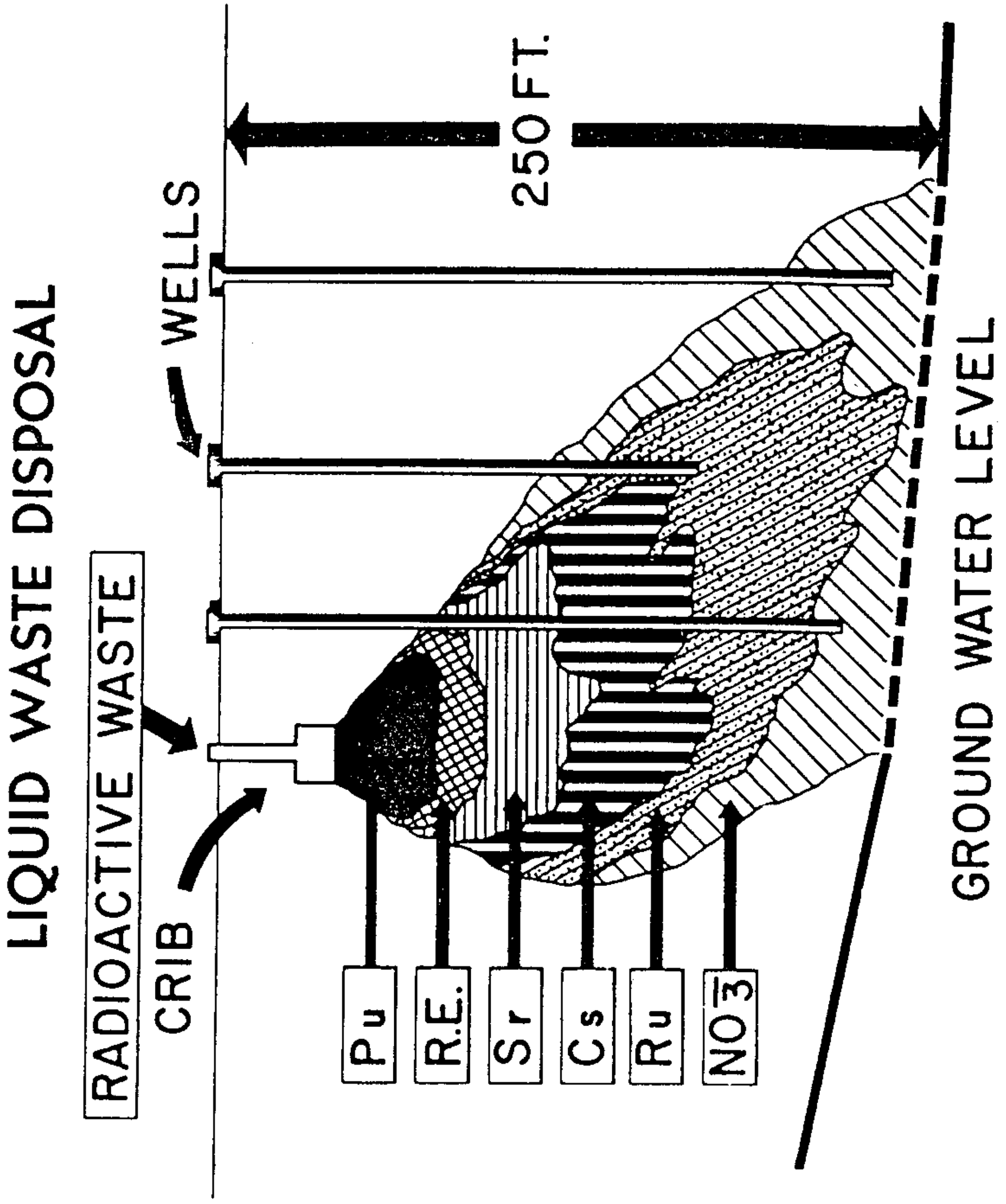

LIQUID WASTE DISPOSAL
RADIOACTIVE WASTE
CRIB
WELLS
250 FT.
Pu
R.E.
Sr
Cs
Ru
NO_3^-
GROUND WATER LEVEL

Figure 18

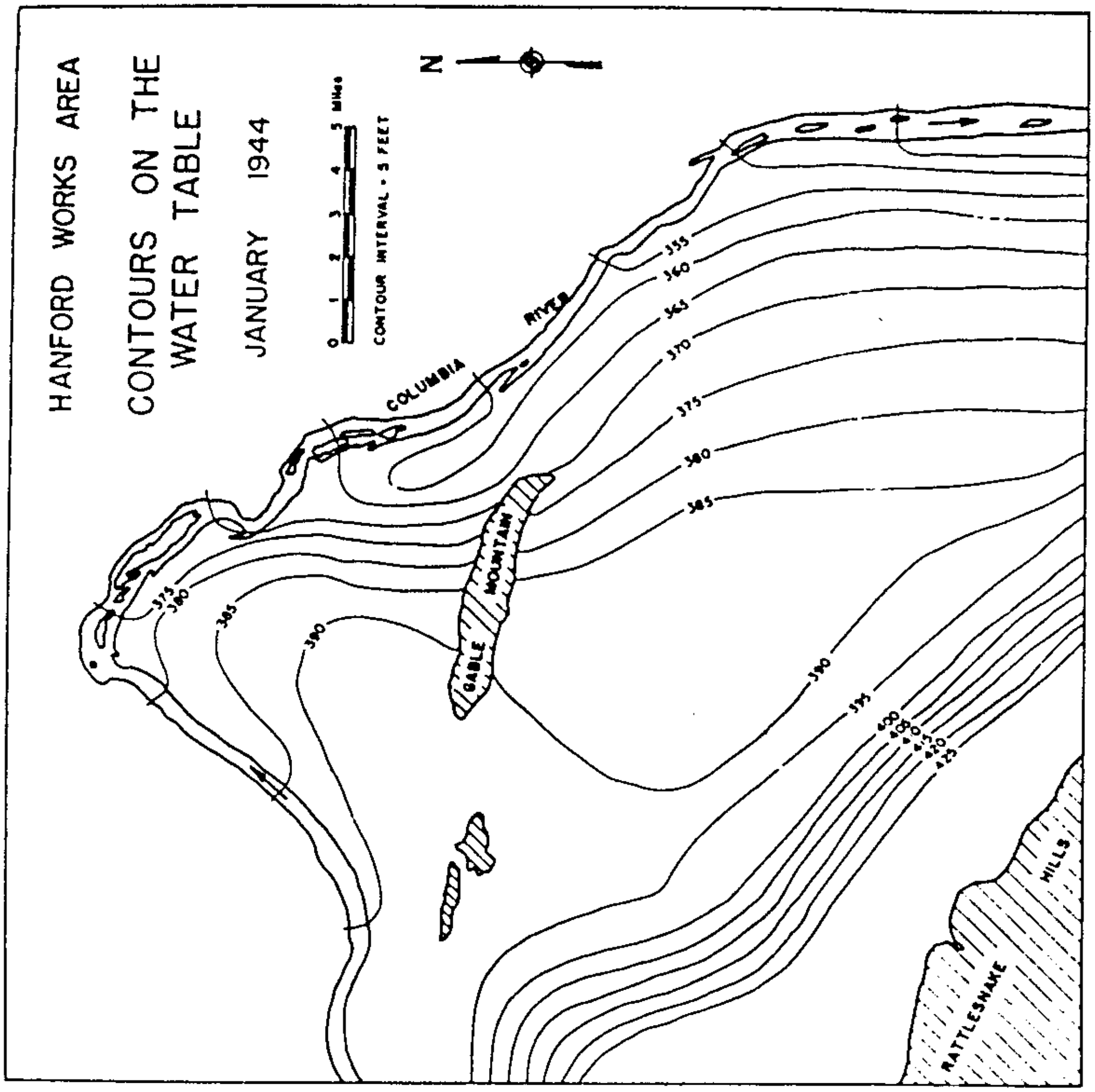

HANFORD WORKS AREA
CONTOURS ON THE WATER TABLE
JANUARY 1944
0 1 2 3 4 5 Miles
CONTOUR INTERVAL - 5 FEET
N
COLUMBIA RIVER
GABLE MOUNTAIN
RATTLESNAKE HILLS
355
360
365
370
375
380
385
375
380
385
390
390
395
400
405
410
415
420
425

Figure 19

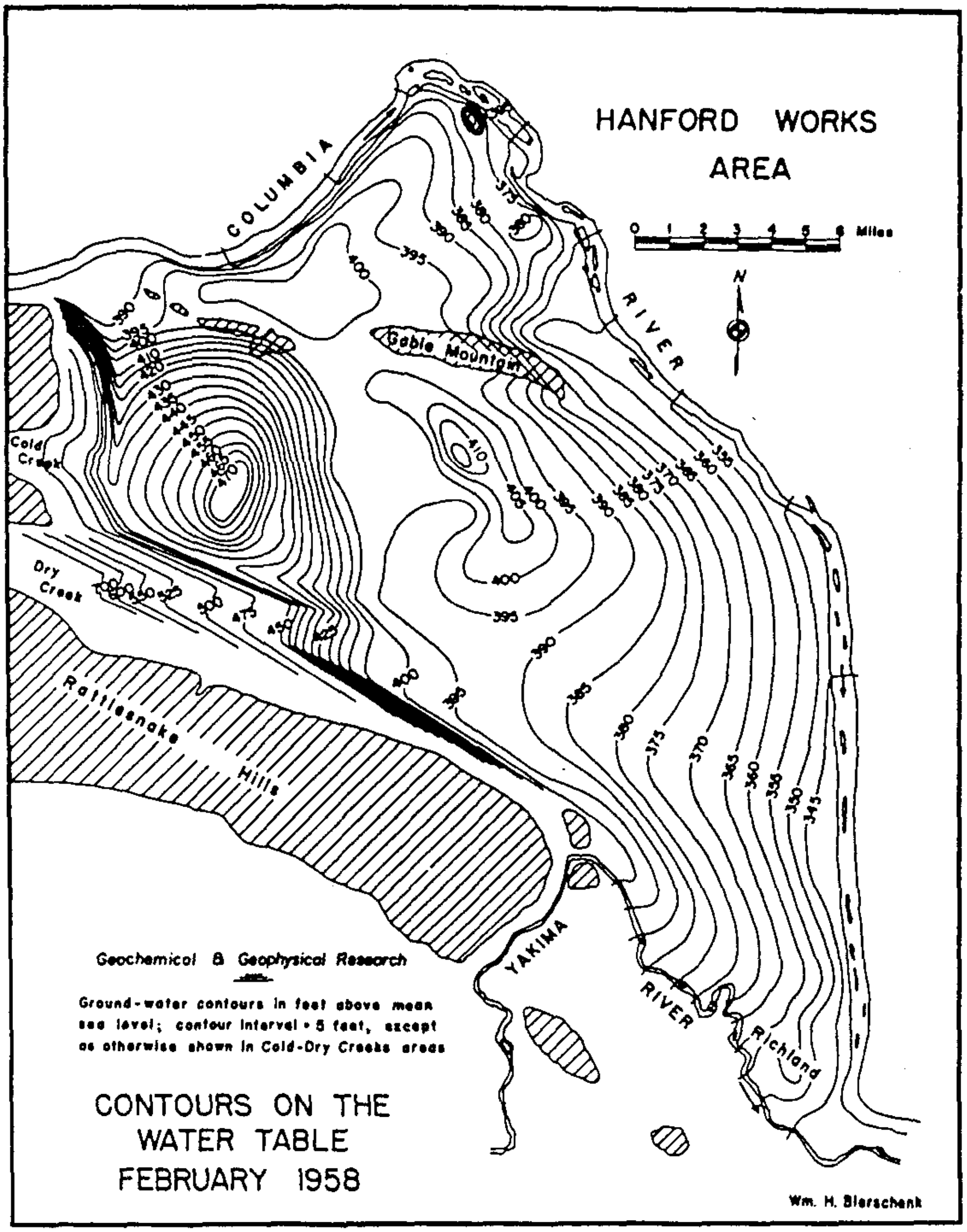

HANFORD WORKS AREA
0 1 2 3 4 5 6 Miles
N
COLUMBIA
RIVER
Gable Mountain
Cold Creek
Dry Creek
Rattlesnake Hills
YAKIMA
RIVER
Richland
Geochemical & Geophysical Research
Ground-water contours in feet above mean sea level; contour interval = 5 feet, except as otherwise shown in Cold-Dry Creeks areas
CONTOURS ON THE WATER TABLE FEBRUARY 1958
Wm. H. Bierschenk

Figure 20

WATER TABLE BALANCE WITH COOLING TOWERS

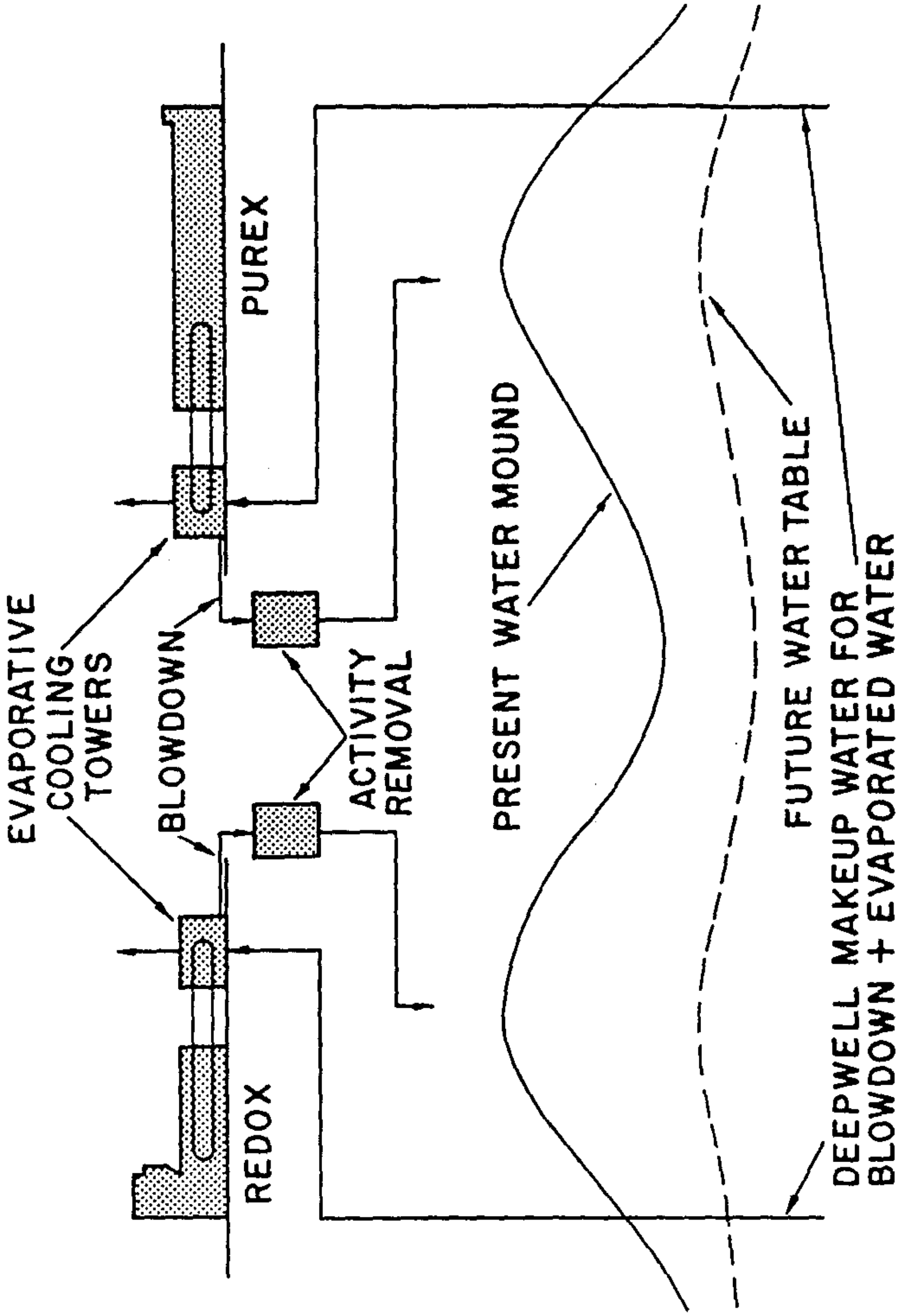


Figure 21

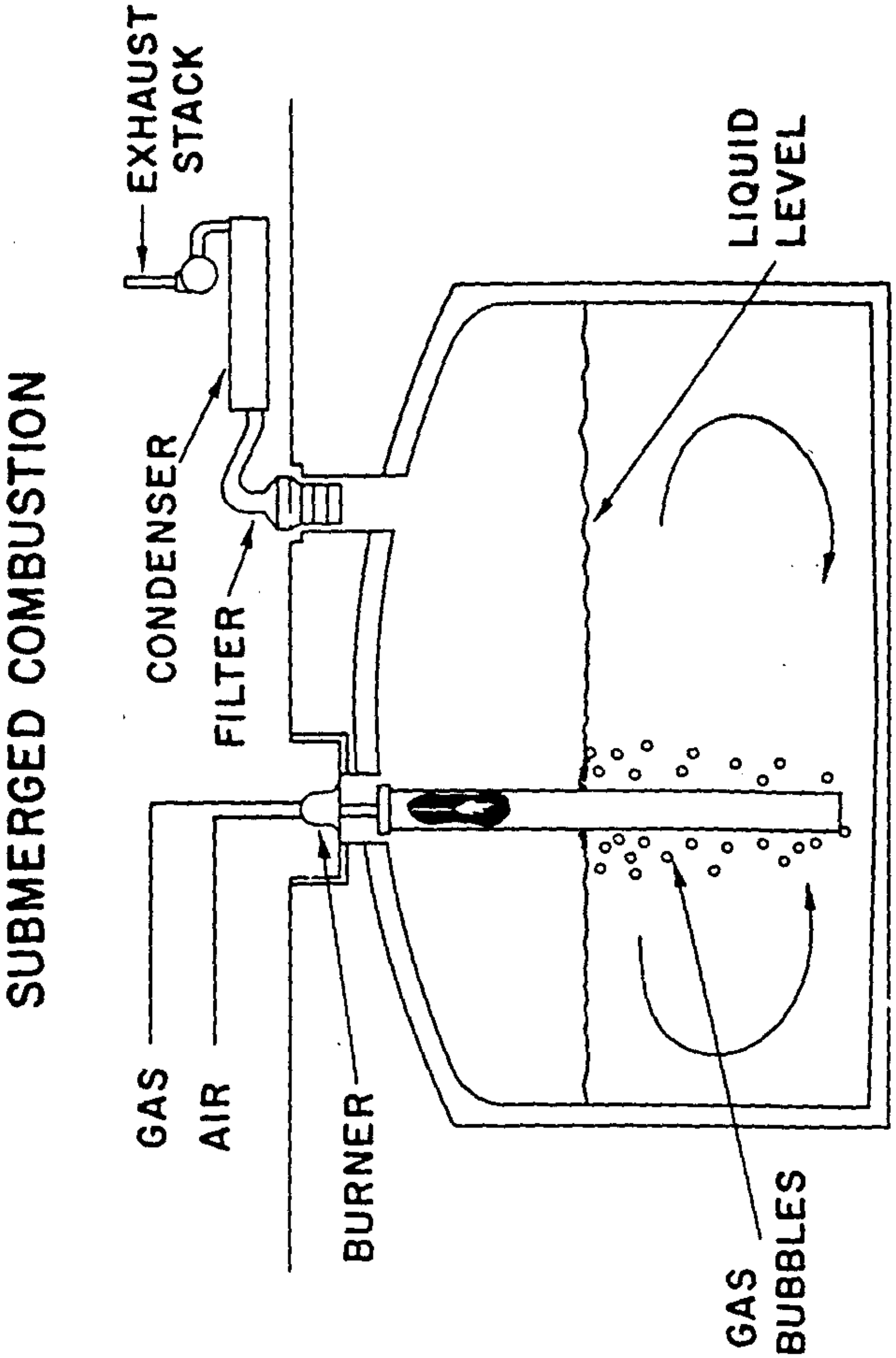

SUBMERGED COMBUSTION
EXHAUST STACK
CONDENSER
FILTER
GAS
AIR
BURNER
LIQUID LEVEL
GAS BUBBLES

Figure 22

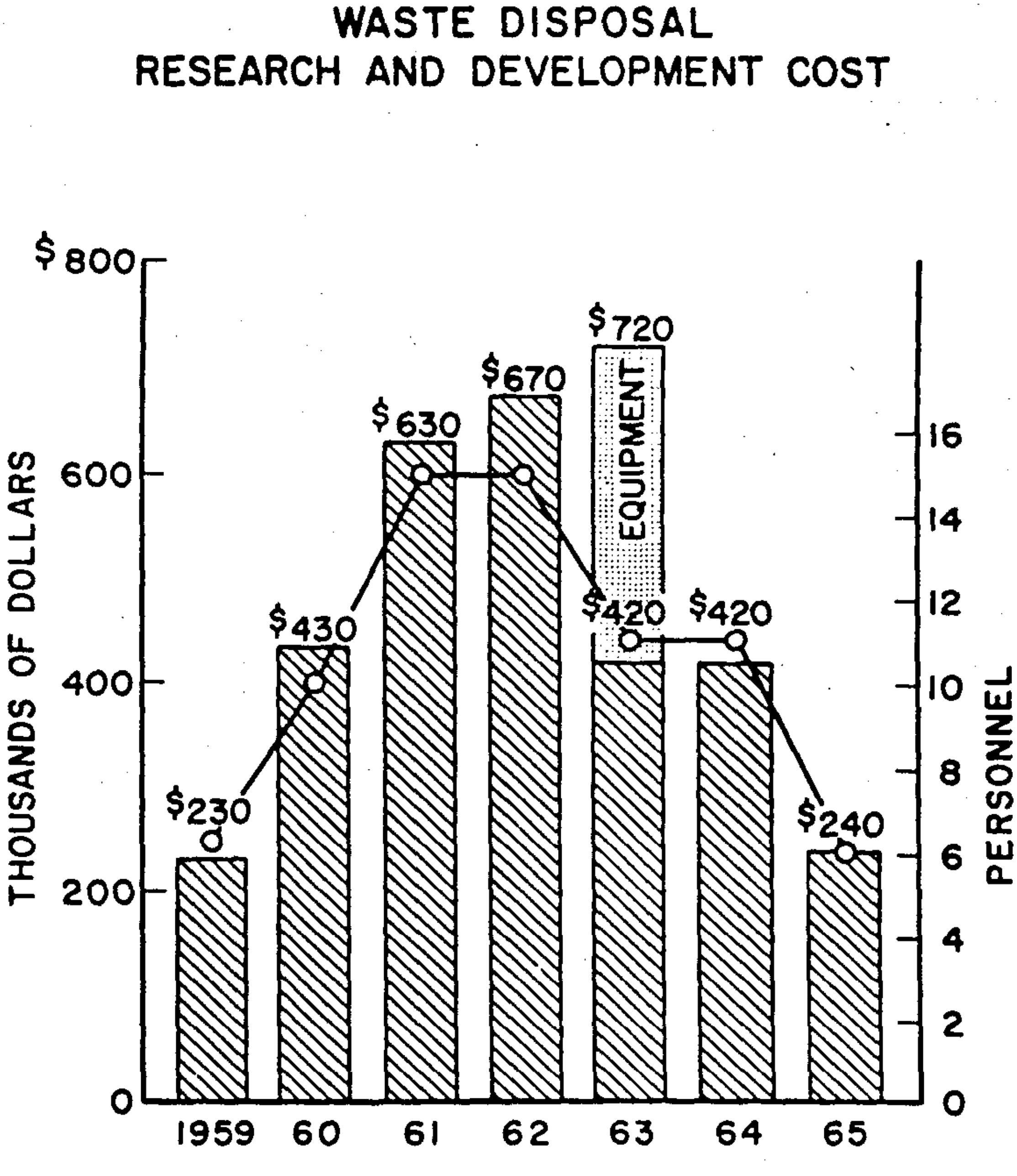

WASTE DISPOSAL
RESEARCH AND DEVELOPMENT COST
THOUSANDS OF DOLLARS
$800
600
400
200
0
PERSONNEL
16
14
12
10
8
6
4
2
0
$230
$430
$630
$670
$720
EQUIPMENT
$420
$420
$240
1959
60
61
62
63
64
65

Figure 23