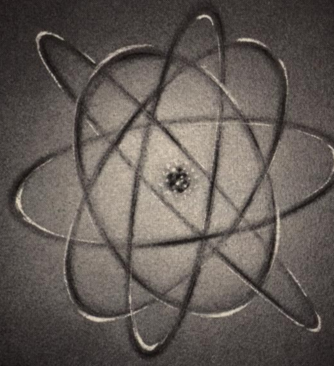

RADIOACTIVE WASTE MANAGEMENT

ACCENT ON VALUE

CHEMICAL PROCESSING DEPARTMENT

HANFORD ATOMIC PRODUCTS OPERATION

THE MANAGEMENT OF RADIOACTIVE WASTES IN HANFORD SEPARATIONS PLANTS

INTRODUCTION

The Hanford plant is operated for the Atomic Energy Commission by the General Electric Company primarily to produce plutonium (a radioactive element found in only trace quantities in nature). Substantial quantities of other radioactive elements are also produced as a result of fission and other reactions in the nuclear reactors. In the Chemical Department, the desired products are isolated and purified for a wide variety of beneficial uses. Virtually everything else that leaves the chemical processing plants is considered to be a radioactive waste.

The primary objective of waste management is to assure that radioactive materials will not constitute a hazard to important life forms in the vicinity. Many of the radioactive materials can be absorbed by plants and animals, and their radiations can damage the living tissues. Large expeditures of funds and technical effort are made to assure that significant quantities of these materials are kept isolated from important life forms.

During the nineteen years of experience in storing and disposing of radioactive wastes at Hanford, an active program of environmental monitoring has been maintained to assure that the minute quantities of radioactive materials released by the chemical processing operations are, and remain, within acceptable limits. This brochure provides a brief description of some of the storage and disposal methods now in use and the handling concepts now being developed for future use.

CURRENT WASTE MANAGEMENT PRACTICES

The isolation of radioactive materials is now accomplished by storing nearly all of the fission products in underground tanks in the processing area. In conducting this confinement operation, the Chemical Processing Department is fortunate in having more favorable climate and ground conditions than exist in most places. So little rain falls in the area that rain water evaporates from the ground rather than percolating downward to the ground water. More than two hundred feet of soil lies between the processing areas and the ground water below. This soil is so dry that many thousands of gallons of water can be added to the soil at a given point near the surface without drainage to the ground water. This capillarity, or blotter effect, would tend to catch and hold small volumes of liquid waste that might inadvertently escape to the ground.

Very small quantities of radioactive materials can be safely dispersed in the environment. The release of radioactive materials is monitored by highly sensitive instruments and is carefully controlled to insure that the quantity released is within established limits. In addition, the environment is monitored regularly for possible contamination. Over 400 wells have been drilled within the reservation, to depths as great as several hundred feet. These wells are sampled and analyzed regularly to measure radioactive materials in the ground water. Local water supplies are monitored regularly for radioactivity. Samples of air and of vegetation, taken from both on and off the Hanford reservation, are tested regularly for possible contamination. Animal life is checked periodically for radioactive material content. These surveillance activities routinely verify that the intake of radioactive materials by people in the Hanford environment is well within the most favorable range suggested by the Federal Radiation Council.

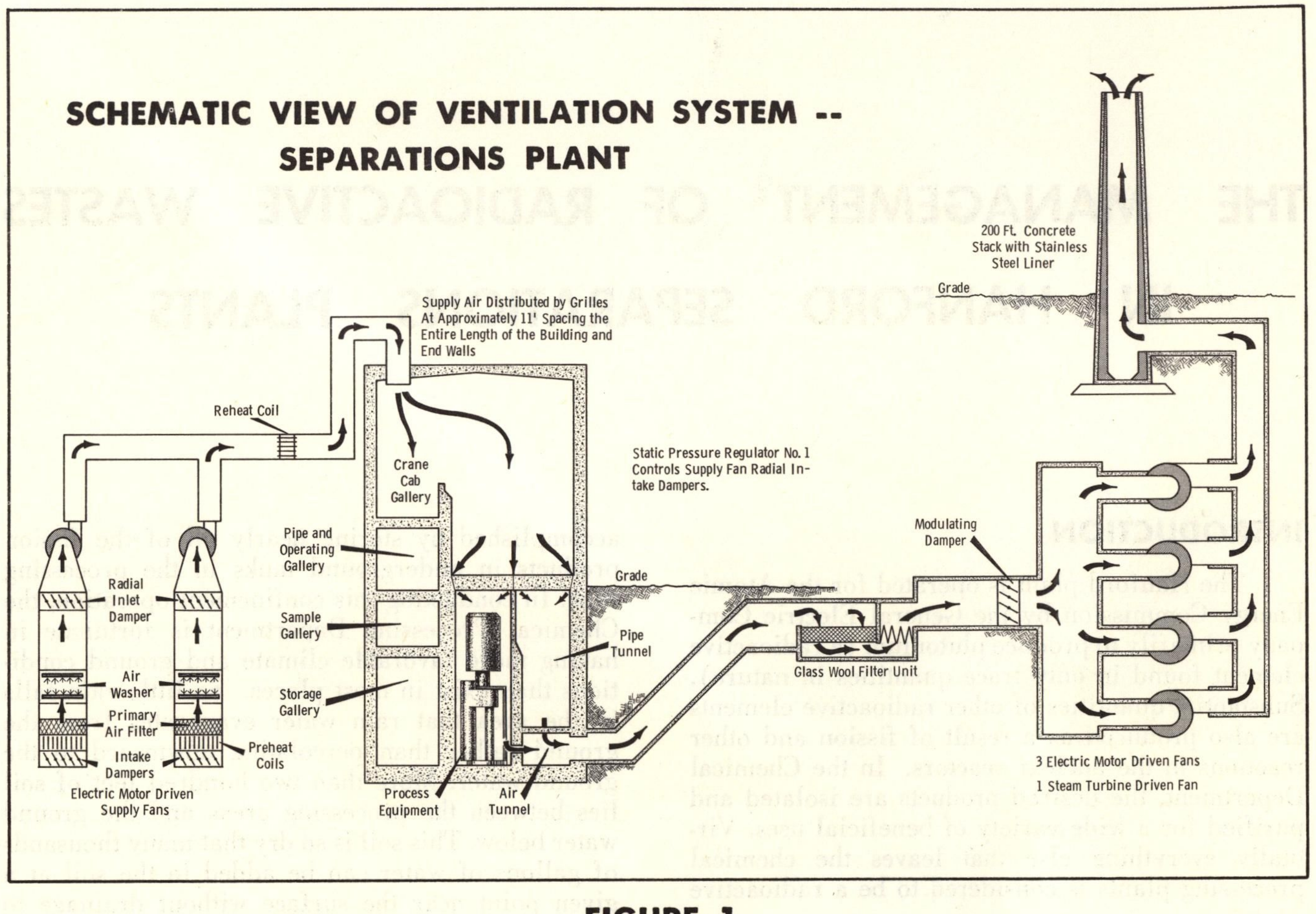

FIGURE 1.
WASTE GASES ARE CAREFULLY CLEANED

THE GASEOUS WASTES ARE CLEANED AND DISPERSED

The ventilation system pictured schematically in Figure 1 is typical of those used in all process buildings. Filtration of the effluent air is required because small particles of dust or mists picked up from process operations can contain radioisotopes and might contaminate the atmosphere and surrounding areas. The filters retain virtually all particles having a diameter greater than about one micron. The particles which pass through the filters are so small that they tend to remain dispersed in the atmosphere. Less than one millionth of one percent of the radioactive wastes generated is released to the atmosphere with these particles.

Some radioactive elements can exist as a gas and therefore may require special treatment. Iodine, for example, is readily vaporized; one isotope, iodine-131, is sufficiently radioactive to require careful control. Special absorbers are used to retain this radioisotope in the plant facilities. The small quantities of iodine-131 which do escape to the atmosphere decay rapidly, thereby preventing a long-term accumulation in the environment.

SOLID WASTES ARE BURIED

Solid materials that become contaminated with radioisotopes are buried when their useful life is ended. Laboratory wastes, such as broken glassware, wiping tissues, etc., are placed in cardboard boxes for burial. Large plant equipment that has failed beyond repair is placed in wooden or concrete boxes for burial. The boxes of radioactive materials are placed in the bottom of trenches up to 20 feet deep. See Figure 2. The packaged materials are then covered with earth. Most of the containers are crushed by the weight of the soil, but the contaminated materials are retained in the ground.

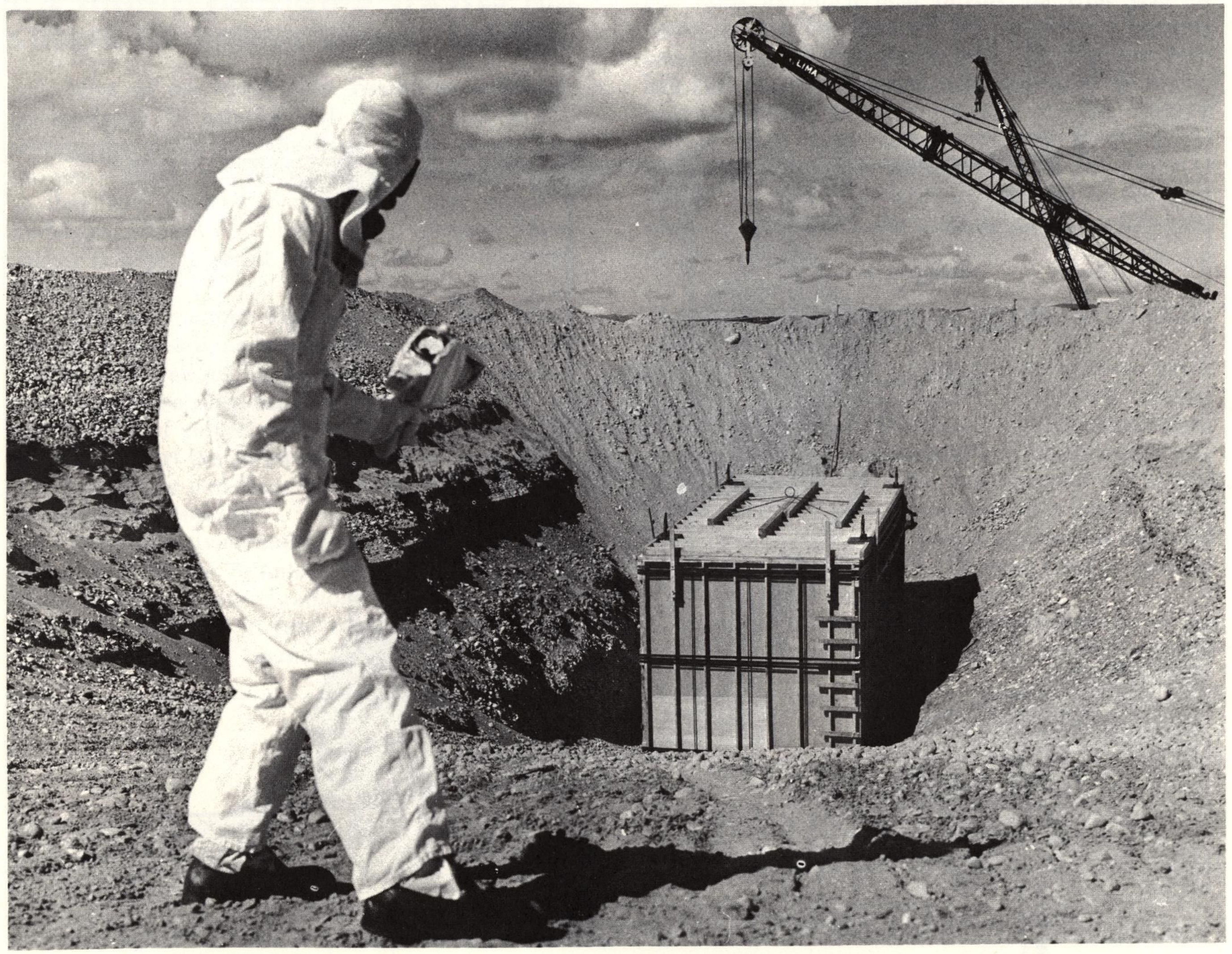

FIGURE 2.
SOLID WASTES ARE BURIED

Some of the failed equipment is so large and so radioactive that boxing and transporting it to a burial site is not practical. These materials are stored in special railroad tunnels adjoining the chemical processing plant. After years of storage, the radioactive materials will decay enough that the equipment could be removed for burial or reclamation if desired.

LOW-LEVEL LIQUID WASTES ARE PERCOLATED THROUGH THE SOIL

The processing of irradiated fuels requires the use of large volumes of water which may become contaminated. Most of this water is used for cooling purposes and never contacts radioactive materials unless a leak develops in the cooling coils or jackets of the processing equipment. (Instruments quickly detect such failures, and corrective action is promptly taken.) This cooling water is discharged to natural depressions in the soil, thus forming a pond or swamp. The water percolates through the soil to the ground water below.

Smaller volumes of waste water are formed when process solutions are concentrated in steam-heated evaporators. These condensates contain less than one ten-thousandth part of the radioactive materials processed. While this quantity of radioactivity is very small, it must be kept isolated from living organisms. This contaminated water is therefore routed to cribs, or porous structures, built below the surface of the ground. From these cribs, the water percolates through the ground to the

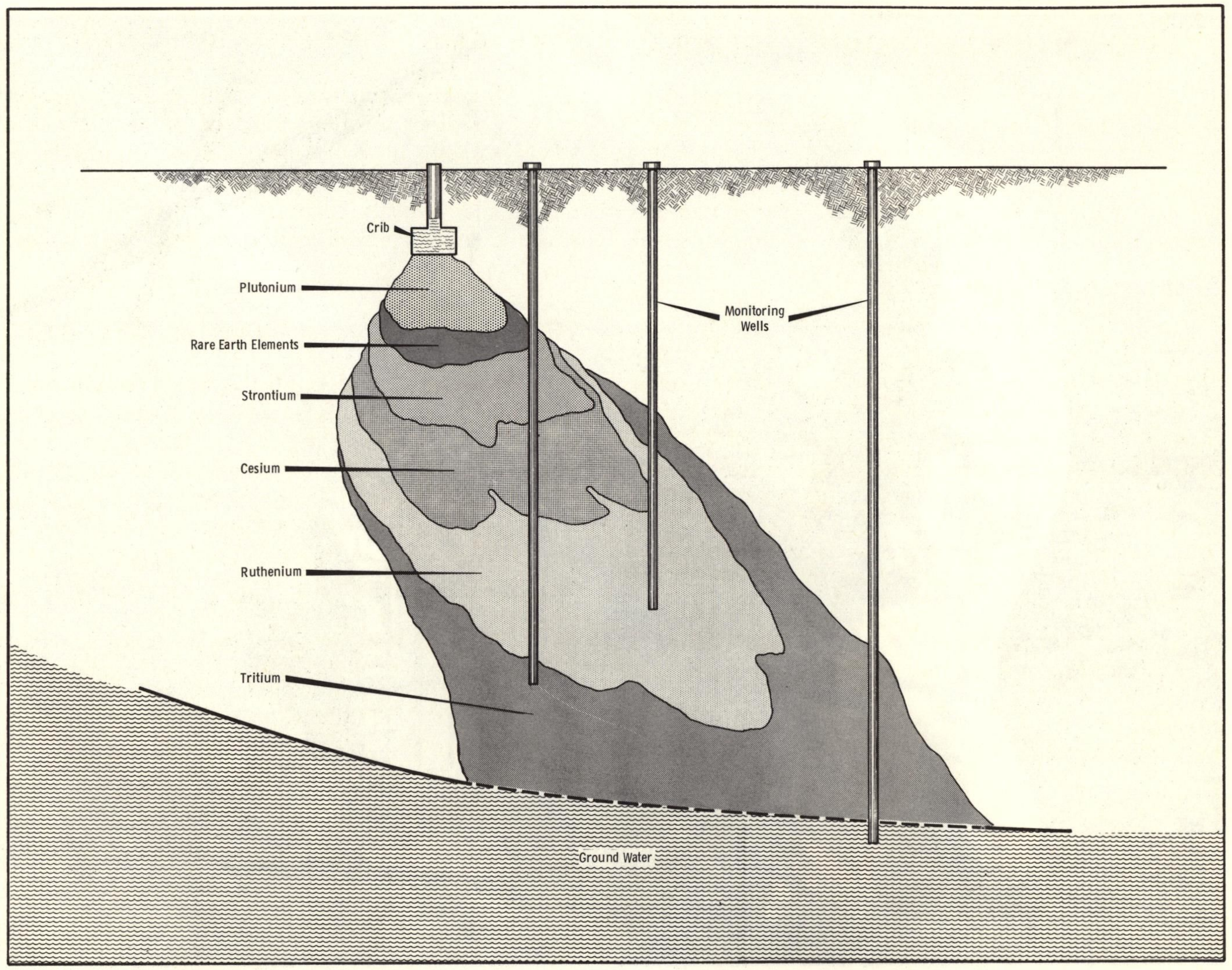

FIGURE 3.
SOME ISOTOPES ARE ADSORBED AND HELD IN THE SOIL

ground water. Most of the radioactive materials are adsorbed and held in the ground because of the ion-exchange properties of the soil. As indicated in Figure 3, different elements are adsorbed and held in the soil with different efficiencies. Plutonium is very tightly held, migrating only a few feet. The rare earths migrate a little faster, and strontium and cesium faster yet. Since some of these elements can be concentrated in animal or plant life, and since some of their radioactive isotopes have long half-lives, precautions must be taken to keep these isotopes away from plants or animals. A disposal site is therefore abandoned before the cesium or strontium reaches the ground water in significant amounts. The most hazardous isotopes are thereby held in the soil above the water table, under conditions which keep them isolated from important life forms.

The few radioisotopes that are not held by the soil are reduced to insignificant concentrations by the time they reach a point of potential contact with animal life. The ground water requires several years to travel the 10 miles or more to the edge of the Hanford reservation. During this transit time, ruthenium isotopes (with half-lives of one year or less) decay to concentrations well below their permissable limits. Tritium has a longer half-life (approx. 12 years), but because of dilution and very low initial concentration, it is also reduced far below permissable limits before it leaves the reservation.

THE HIGH-LEVEL LIQUID WASTES ARE STORED IN UNDERGROUND TANKS

Most of the radioactive fission products are accumulated in solutions of process wastes containing the chemical salts used in the separations process. These waste streams are made alkaline so they will be less corrosive and are routed to underground tanks for storage. The volume of these wastes is kept as small as practicable to minimize storage costs.

During the past eighteen years, 145 underground tanks, having a combined capacity of over 90,000,000 gallons, have been built at Hanford to contain these radioactive wastes. These tanks are constructed of reinforced concrete with an arched roof to support the seven to ten feet of earth cover. The tank walls and floor are lined with mild steel to contain the liquids.

As radioactive materials decay in these storage tanks, most of the radiated energy is converted to heat. Small amounts of radioactive isotopes release very large amounts of heat over a period of time. For example, one pound of pure strontium-90 generates as much heat during its first year of decay as can be obtained by completely burning one-half ton of high grade coal. During one half-life (28 years), a pound of strontium-90 produces heat equivalent to about ten tons of coal. Since a storage tank may contain tens to hundreds of pounds of such heat producers, it is necessary to design the tanks for efficient removal of the heat while keeping the radioactive materials safely confined.

THE MOST ACTIVE WASTES SELF-BOIL

Over 99 percent of all the waste fission products processed are concentrated in a small fraction of the liquid wastes. The decay of the radioisotopes in these wastes generates heat at a high enough rate to boil the solutions in the underground tanks; this self-heating action will continue for many years. Heat from the decaying fission products is removed with steam just as heat is removed from a simmering teakettle on the kitchen stove.

Figure 4 illustrates the special features of the 21 underground tanks equipped to handle these

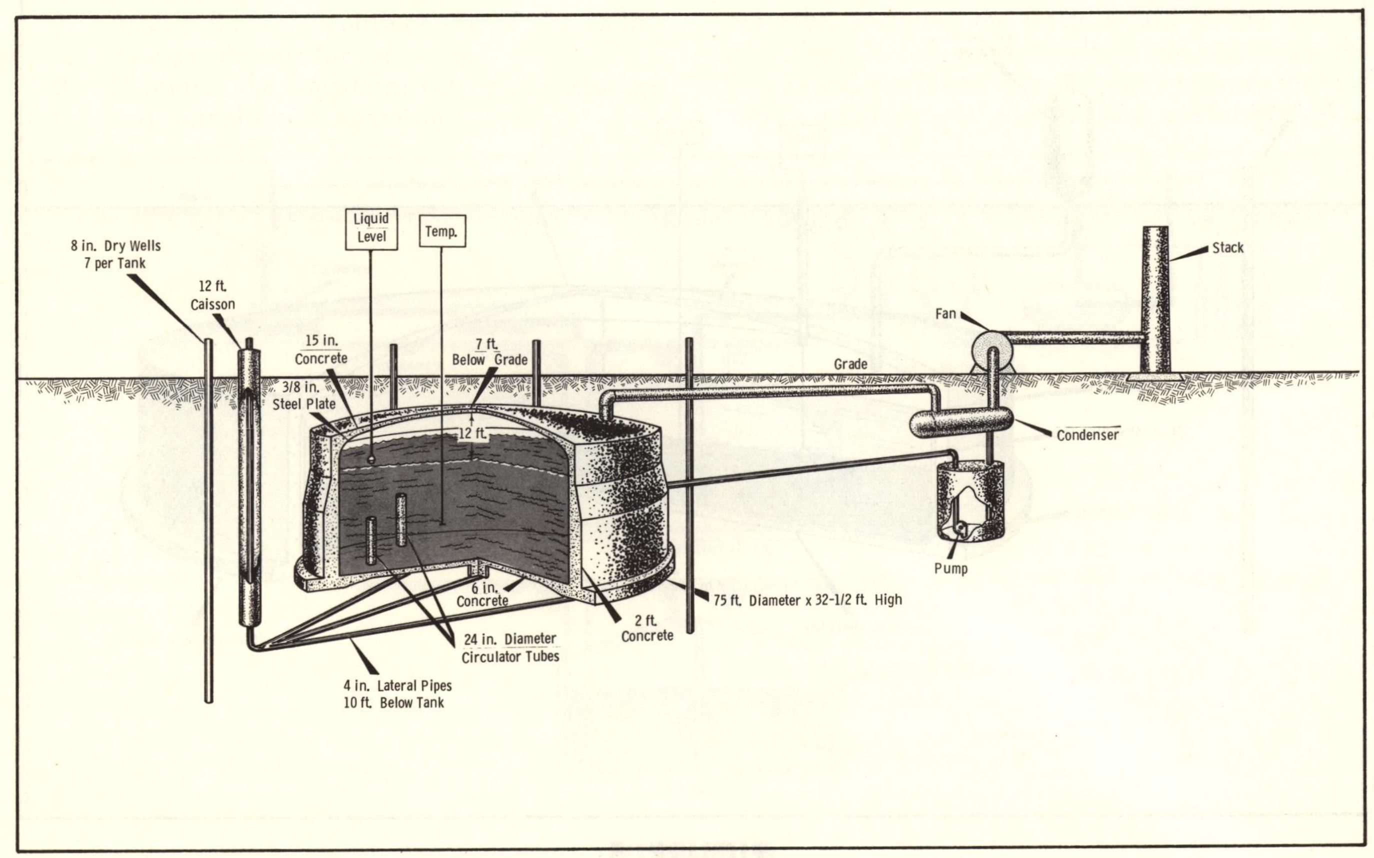

FIGURE 4.
THE MORE CONCENTRATED WASTES SELF-BOIL IN UNDERGROUND TANKS

self-boiling wastes. A vapor line carries the water vapor to a condenser (one in each of the two groups of tanks, or tank farms) where the water is condensed and drained to a pump tank. This water is then either pumped to a crib, thereby effecting self-concentration of the stored waste, or returned to the tank to prevent too much concentration. A fan maintains a slight vacuum in the condenser and underground tanks and exhausts any air in-leakage to the atmosphere through a filter and stack. Air-lift circulators are provided to assist in the uniform removal of heat. A special well and pump are provided to supply cooling water in an emergency, and a spare compressor is constantly ready to power the air-lift circulators if the normal air supply should fail. While these tanks are equipped with liquid-level measurement devices, it is difficult to detect a small leak by this means in a tank of self-boiling wastes. A network of vertical and horizontal pipes has therefore been installed around and under each of the tanks designed to contain self-boiling wastes. Traversing these pipes with a radiation detector provides a means of detecting radioactivity in the soil should a leak develop.

THE LESS ACTIVE WASTES DO NOT SELF-BOIL

Most of the liquid wastes in storage do not self-boil; they contain relatively small amounts of radioactive isotopes, mixed with large amounts of non-radioactive chemicals. The removal of heat from these wastes is a relatively minor problem; heat is conducted through the tank walls and the surrounding soil fast enough to keep the temperature of the tank contents well below boiling temperatures. The problem of storing these non-boiling wastes is simply one of assuring confinement. Figure 5 shows a sectional view of one of the tanks used for this purpose. The tanks are equipped with

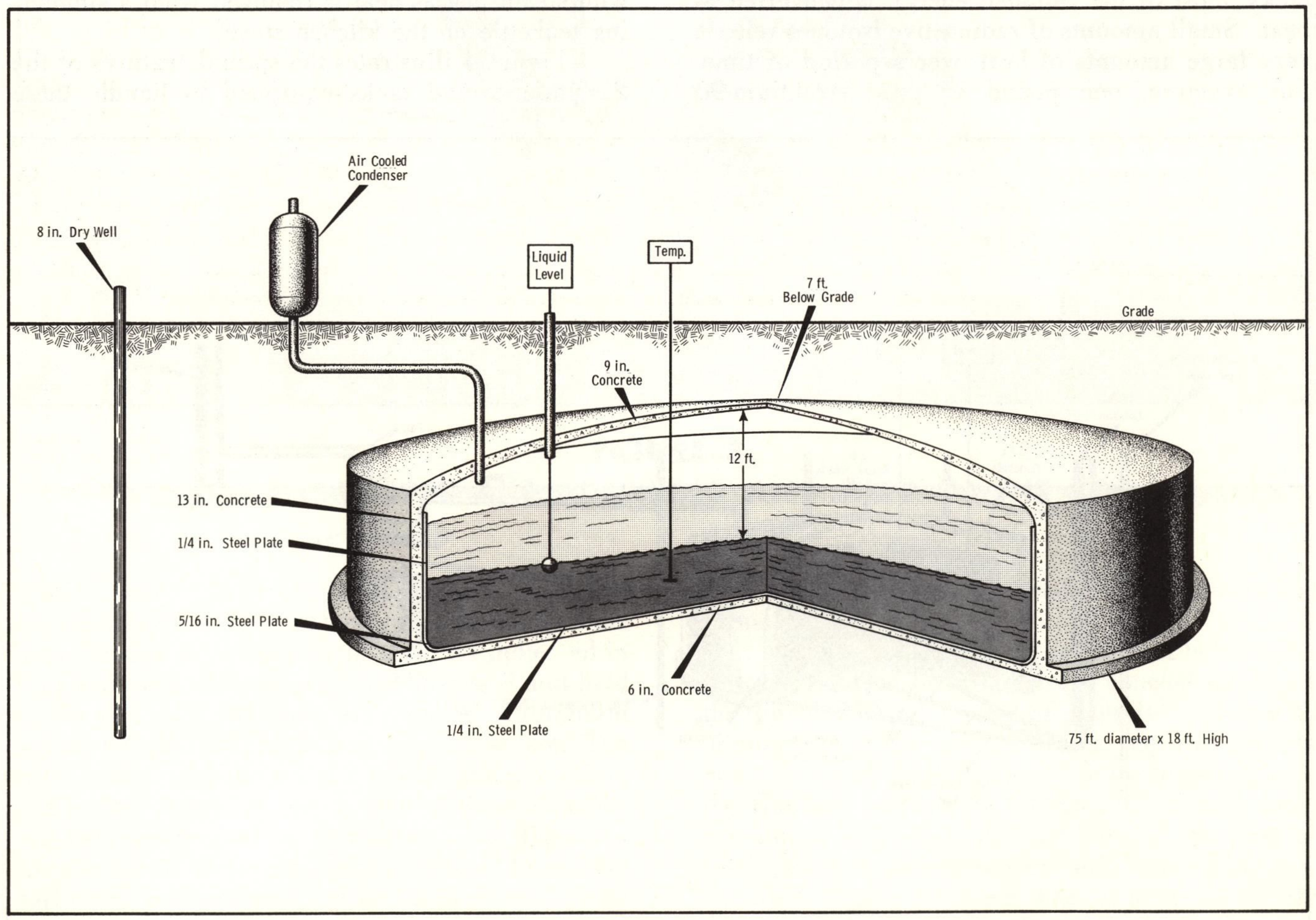

FIGURE 5.
THE LESS RADIOACTIVE LIQUID WASTES ARE STORED IN UNDERGROUND TANKS

devices to measure liquid level and temperature, and these instruments are read at regular intervals. Vertical pipes are placed in the ground around the buried tanks. Ionization chambers lowered in the pipes can detect radioactivity in the soil, thereby providing a secondary method of detecting a leak should one develop.

It is recognized that neither type of waste storage tank will last forever. In the few instances that tanks have developed leaks, the leaks were first detected by liquid-level measurements and subsequently verified by observing radioactive materials in the soil. In each case, the contents of the tanks were pumped to another vessel before excessive amounts had leaked. Because of the ability of the ground to hold liquid, none of the leakage has reached the water table.

IMPROVEMENT PLANS

During the past few years, the Chemical Processing Department has formulated a program to achieve significant improvement in the handling of high-level radioactive wastes. This program has three objectives:

1. Assure the safe confinement of the radioactive materials for centuries.
2. Minimize the long-term cost of the storage and surveillance operations.
3. Make selected isotopes available for beneficial use.

The improvement program is essentially approved by the Atomic Energy Commission and is now being implemented. In this program, the wastes that generate very little heat will be evaporated to salt cakes in the existing underground tanks. The self-boiling wastes will be separated into two fractions, one containing the long-lived heat producers for storage in separate high-integrity containers, the other fraction containing all other materials to be solidified eventually as "low-heating" wastes.

LOW-HEATING WASTES WILL BE SOLIDIFIED

A detailed design is now being developed for the first unit to concentrate "low-heating" wastes (those containing relatively small amounts of radioisotopes) to a salt cake. This unit is expected to be operative in the fall of 1964.

The proposed system is schematically portrayed in Figure 6. In operation, heated air is blown through a pipe and released near the bottom of the tank. The air rises inside a three-foot-diameter tube, inducing the waste liquid in the tube to rise also. In this way, the rising air both heats the liquid and circulates it in the tank to assure that the tank's contents are concentrated uniformly. The

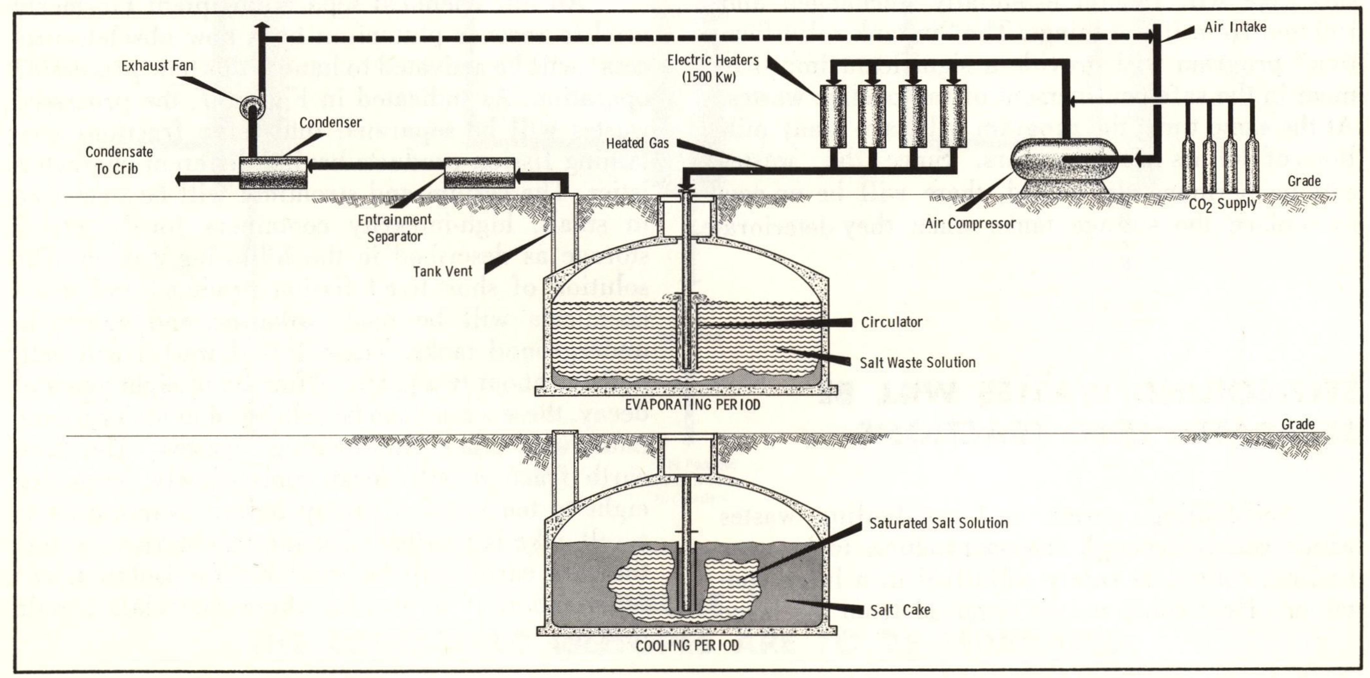

FIGURE 6.
THE LOW-HEATING WASTES ARE TO BE EVAPORATED TO SALT CAKES IN THE UNDERGROUND TANKS

air and vaporized water pass through an entrainment separator to minimize the escape of radioactive materials, and the water vapor is condensed. The condensate is routed to an underground crib. The air is exhausted to the atmosphere, or it can be recycled to the tank if air contamination should prove to be a problem.

Laboratory and pilot plant data indicate that this concentration system will work well in the 75-foot-diameter underground tanks, and that enough water can be removed so that the concentrate will crystallize to a massive salt cake upon cooling. Some crystallized salts are expected to accumulate on the bottom of the tank during the concentration operation. When the introduction of heated air is stopped, a salt cake is expected to form slowly on all sides of the mass as it cools. Since these wastes will continue to generate small amounts of heat from radioactive decay, it may require several years for the mass to cool sufficiently to form a solid salt cake.

Once the salt cake has formed, there need be no concern about the continuing integrity of the tank. When the tank liner eventually corrodes away and the concrete structure loses its strength, the salt cake will remain essentially unchanged and isolated from living things. The "in-tank solidification" program will provide a significant improvement in the safe confinement of radioactive wastes. At the same time, the program will save many millions of dollars in future years. Since the wastes will be permanently buried, there will be no need to replace the storage tanks when they deteriorate.

SELF-BOILING WASTES WILL BE SEPARATED INTO FRACTIONS

Self-boiling wastes, and non-boiling wastes which contain enough fission products to be near boiling, cannot be safely solidified in a large container. Heat could not be removed from the large solid mass rapidly enough, and the salt temperaturs could potentially climb to several thousand degrees Fahrenheit. It is therefore necessary to develop another method for handling the more concentrated wastes.

Self-boiling wastes initially contain many different heat-producing radioisotopes, most of which decay to very low levels in a few years. Most of the heat generated after ten years of decay is produced by only two long-lived isotopes — strontium-90 and cesium-137. These two isotopes alone produce enough heat to keep the wastes boiling for a few decades. However, if these two isotopes were removed from the waste, the remaining portion could be safely solidified after a few years of decay.

Scientists in the Hanford Laboratories have developed a solvent-extraction process than can extract the cesium and strontium from the waste solutions. Another group of fission products called the "rare earths" is also extracted and can be readily separated from the cesium and strontium in a separate operation. This process is call CSREX (pronounced ces' ar ex), standing for Cesium, Strontium, Rare Earth eXtraction.

An idle chemical separations plant (formerly used to recover plutonium by a now obsolete process) will be activated to handle this new processing operation. As indicated in Figure 7, the processed wastes will be separated into three fractions containing fission products having different characteristics. The cesium and strontium will be packaged in small, high-integrity containers for long-term storage as described in the following section. The solution of short-lived fission products and waste chemicals will be made alkaline and stored in underground tanks. These liquid wastes will self-boil for about two years. After six to eight years of decay, these wastes can be solidified in underground tanks with other "low-heating" wastes. The rare-earth fraction will decay more slowly, requiring eight to ten years of decay before evaporation to a salt cake is feasible. During this storage period, the rare earths will be available for isolation and purification if a use for these materials should develop.

Somewhat different procedures have been developed to process the self-boiling wastes now in storage. These wastes contain large quantities of salts that cannot be processed through the CSREX process economically. Cesium will be extracted from the large volumes of salt solutions by using a simple ion-exchange process very similar to a home water softener system. The cesium and strontium will be separated from the insoluble salts by leaching and precipitation techniques.

In each case, the separated cesium and strontium will be packaged, as described in the following section, and the residual salts will be concentrated to salt cakes with the "low-heating" wastes.

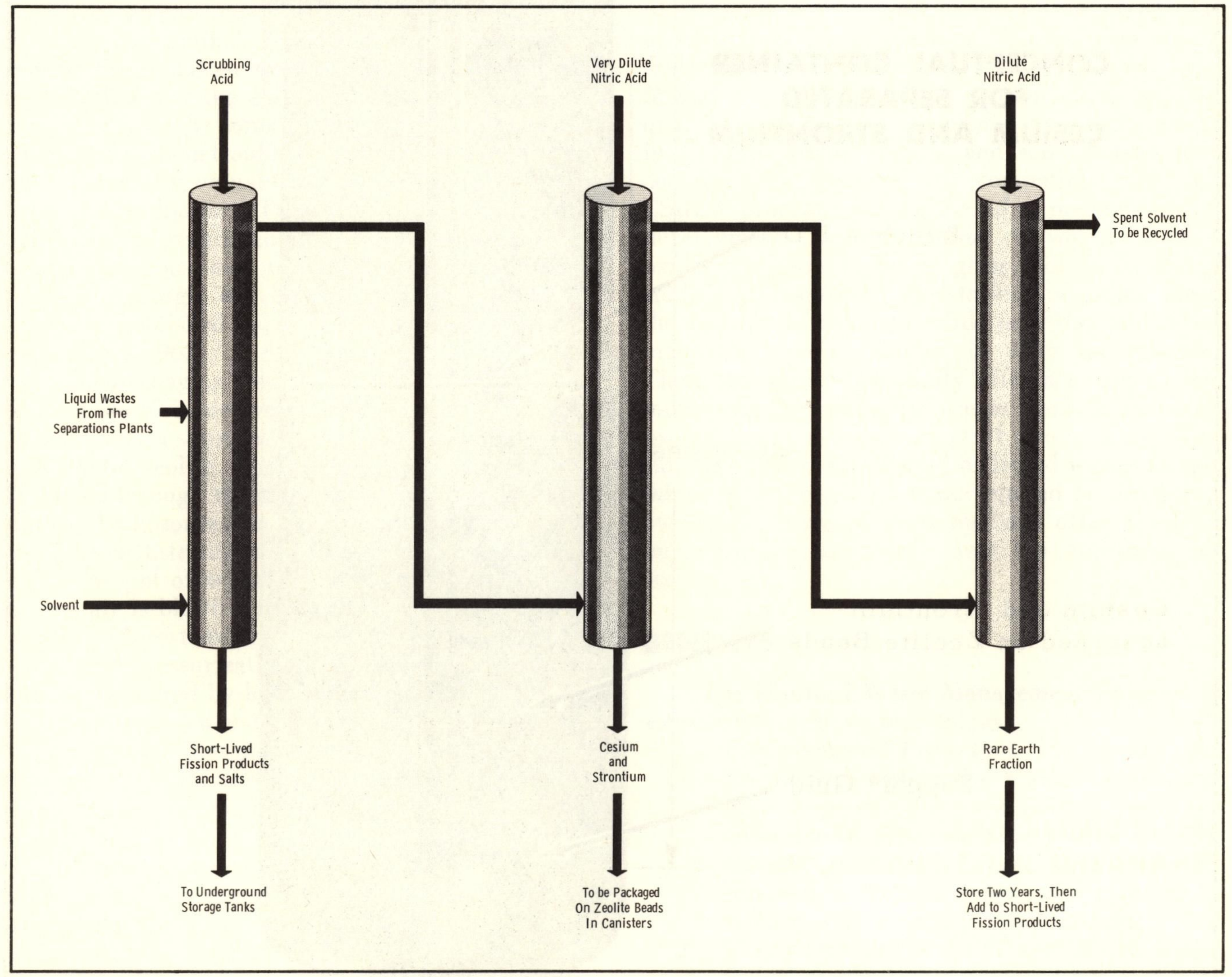

FIGURE 7.
THE LONG-LIVED ISOTOPES ARE TO BE SEPARATED

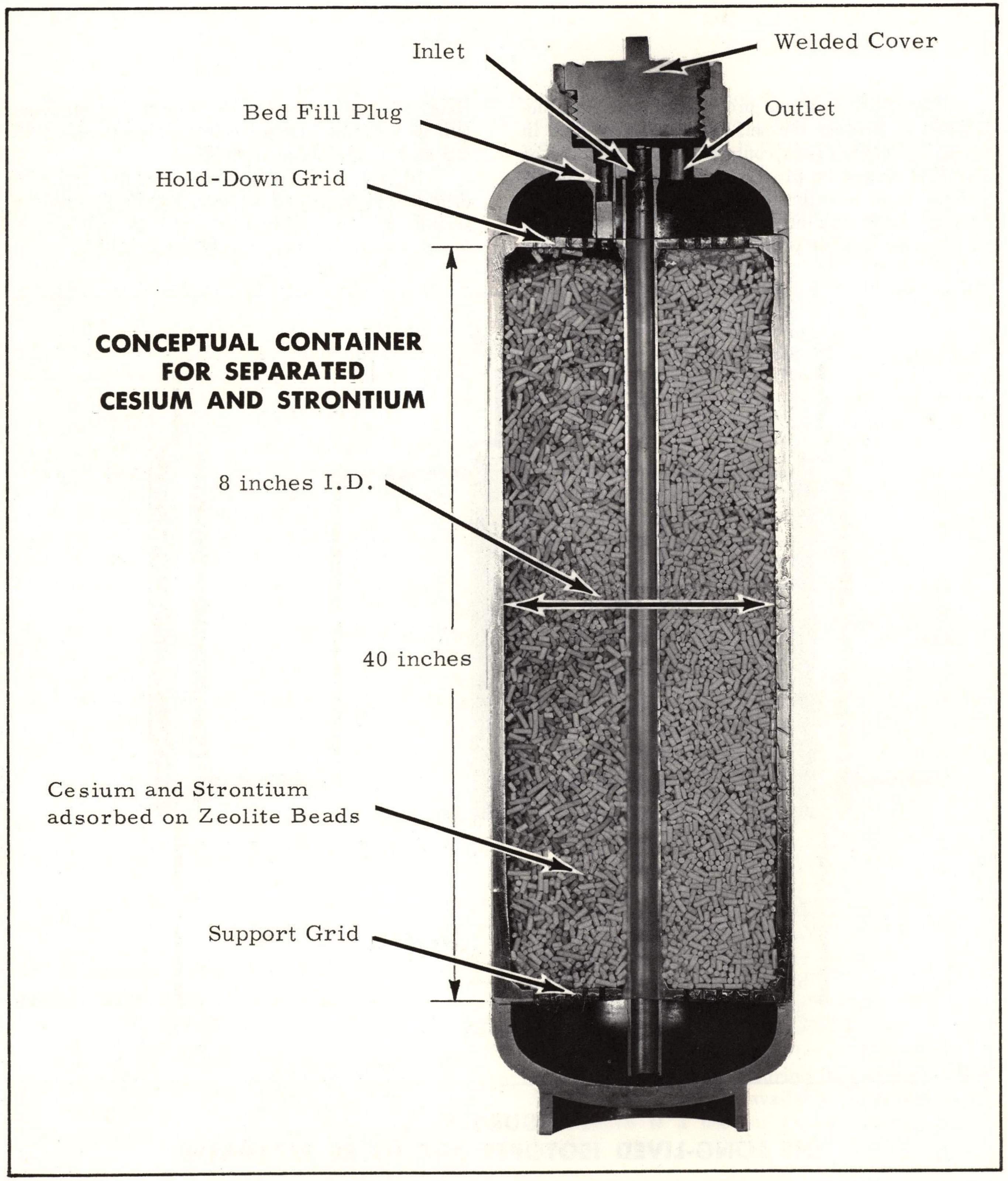

FIGURE 8.
THE LONG-LIVED ISOTOPES WILL BE ADSORBED ON ZEOLITE BEADS AND STORED IN CANISTERS

CESIUM AND STRONTIUM WILL BE PACKAGED

The isolated cesium and strontium will require special handling because of the unique characteristics of these materials. Cesium-137 and strontium-90 are both very active heat and radiation producers, both have long half-lives, and both are toxic to humans. The storage containers for these isotopes must provide a very high level of assurance against failure for any reason and must be capable of dissipating large amounts of heat. The container contents must be physically and chemically stable, and unlikely to be dissolved or dispersed even if the container were to fail. In addition, it is desirable that the cesium and strontium be recoverable from the container, since the radioactivity which makes them troublesome as wastes also makes them potenially valuable as long-lived sources of heat and radiation. These isotopes are already being used in experimental devices to power remote weather stations and navigational aids, sterilize foods and drugs, etc. See "Fission Products", a brochure published by General Electric Company September, 1962.

The packaging system which is being proposed to meet these requirements is depicted in Figure 8. In this plan, a stainless steel canister is filled with a synthetic zeolite. The cesium-strontium solution is passed through the canister; cesium and strontium can be absorbed on the zeolite in quantities up to 17 percent strontium or 35 percent cesium, based on the weight of zeolite. When the bed is loaded, heated air is blown through the bed to dry it thoroughly. More than trace amounts of water could cause later pressurization of the canister since water can be vaporized by heat or decomposed by radiation into hydrogen and oxygen gases. After drying, the canister will be completely sealed with a welded cap and carefully checked for a period of months to make sure there is no leakage, pressurization, or over-heating.

Present plans call for storage of the canisters under water in existing storage basins to assist in dissipating the heat. Each canister, if loaded as proposed, will generate about as much heat as a typical kitchen oven in constant use. Many years later when the heat generation rate has decreased considerably, the canisters might be moved to a dry storage area.

These heavy-walled, corrosion-resistant, completely-sealed canisters are expected to provide long-term high-integrity storage with minimum risk and minimum attention. Even in the remote event of a canister failure, the strontium and cesium would not be dissolved or dispersed by natural forces to any significant extent. On the other hand, if it should be desirable to recover the strontium and cesium, either for beneficial use or for transfer to another package, the isotopes could easily be washed out of the zeolite with acid solutions.

ALTERNATIVES

Other ways of immobilizing high-level radioactive wastes are being developed at many sites, particularly with respect to the future processing of fuels from power reactors. Wastes from power fuels will be different from those discussed here in that they will generally contain much smaller quantities of process chemicals than are present in Hanford stored wastes, and some wastes will contain salts that are not compatible with the CSREX process. Among the other methods under development, the calcination of wastes to dry oxides and their fusion to glassy solids are being studied at Hanford Laboratories and other sites; the fixation of calcined solids in sulfur and other materials is being studied in other laboratories. These techniques potentially offer a means of immobilizing the radioactive wastes from the chemical processing of a wide variety of power fuels. The storage of radioactive solid or liquid wastes in salt mines or caverns deep under ground is also being evaluated. None of these methods offer Hanford any economic advantage over the approach described.

BENEFITS

The Hanford Waste Management Program is expected to achieve four things:

1. *Environmental Protection.* The careful confinement of radioactive materials will continue to protect plant and animal life in the environment from ionizing radiations. The minute quantities of radioactive materials released to the atmosphere will continue to remain well within acceptable limits.
2. *Waste Immobilization.* The radioactive materials currently being stored as liquids in underground tanks will be solidified. This solidification will result in increased certainty of long-term retention.
3. *Cost Reduction.* The costs of solidifying the high-level radioactive wastes will be less than the cost of continuing to store them in tanks. Many millions of dollars will be saved

because no new tanks need be constructed for the storage of "low-heating" wastes in the foreseeable future.

4. *By-Products Availability*. Potentially valuable by-products will be made available for beneficial use. While the need for these by-products is not yet established, the isolated materials are to be stored in such a way that they can be retrieved some later time.

The proposed program is believed to be the best means of achieving satisfactory long-term radioactive waste management at Hanford at reasonable cost. Similar methods may also be useful at other locations, although this program was specifically designed to fit Hanford's particular needs and advantages; other sites do not have the same large volumes of existing salt wastes, the same favorable climate and geology, and the same available facilities that exist at Hanford.

If the proposed Hanford program is approved and carried out as planned, it is expected that all existing liquid wastes will be segregated and solidified, and the processing of wastes will be on a current basis by about 1973.

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