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SOLIDIFICATION OF SAVANNAH RIVER PLANT HIGH-LEVEL WASTE

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SOLIDIFICATION PROPOSAL

The Department of Energy, in accord with recommendations from the Du Pont Company, has started construction of a Defense Waste Processing Facility (DWPF) at the Savannah River Plant (SRP). This facility will immobilize the large quantity of high-level radioactive waste now stored at the plant plus the waste to be generated from continued chemical reprocessing operations. The SRP is presently the nation's primary source of tritium, weapons plutonium, heat source plutonium, and several other radionuclides for defense, space, medical, and energy applications. SRP was built by the Du Pont Company for the U. S. Atomic Energy Commission in the early 1950's and the plant is still operated by the Company for the

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U. S. Department of Energy (DOE). The plant comprises a large, remote land area with extensive support facilities and a single operating contractor; it is an excellent location at which to begin this nation's first large-scale immobilization of high-level radioactive wastes.

The DWPF will provide an immobile waste form suitable for emplacement in a federal repository to be constructed as described in the Nuclear Waste Policy Act of December 1982. Together these facilities will assist in the demonstration of a safe, effective nuclear waste disposal policy for the United States. The DWPF is the result of over ten years research, development, and demonstration at the Savannah River Laboratory, and cooperation with similar efforts overseas.

The DWPF has been authorized for construction starting late in 1983, and will be producing canisters filled with radioactive waste immobilized in borosilicate glass by January 1990. The DWPF project which will cost \$910 million will provide facilities to take radionuclides from existing waste tanks at SRP, primarily metal oxide sludges, and immobilize them in glass which is then poured into stainless steel canisters. These canisters will be sealed, cleaned, and stored prior to emplacement in a federal repository. Facilities will also be provided to immobilize residual waste salts (primarily sodium nitrate) as grout for onsite disposal, to remove sludge from tanks, and to remove cesium-137 from the waste salts in the tanks.

SRP HIGH-LEVEL WASTES

The radioisotopes produced at SRP are generated in large nuclear reactors by irradiating the appropriate target materials with neutrons from uranium fuels and then chemically separating the products of the irradiations in two onsite reprocessing plants. The high-level radioactive waste from this chemical separation contains the radioactive fission products generated in the reactors, some unrecovered uranium, trace quantities of plutonium and other irradiation products, most of the reprocessing chemicals and inert target materials, and large amounts of sodium nitrate (Figure 1). The sodium nitrate is generated during neutralization of nitric acid waste solutions with sodium hydroxide; alkaline waste is stored in carbon steel tanks at SRP.

In the 30 years of SRP operations, some 72 million gallons of high-level waste have been generated; waste generation is expected to continue at the rate of 1.5 to 4 million gallons a year depending on operating demands. This waste (Figure 2) has been evaporated to about 30 million gallons which is now stored in large underground tanks. The principal radionuclides in the waste are strontium-90 and cesium-137. Some 10% of the waste is a sludge generated from precipitates of the hydroxides of iron, manganese, and aluminum; the sludge contains most of the strontium and small amounts of actinides not recovered in the reprocessing plant (Figure 3). The remainder of the waste is liquid and saltcake, which consists primarily of sodium nitrate, sodium nitrite, sodium

aluminate, and sodium hydroxide; this waste fraction contains most of the cesium (Figure 4).

A waste tank replacement program has provided 27 new, double-wall, stress-relieved tanks in separate underground concrete vaults (Figures 5 and 6). Some of the original tanks built in the 1950's have experienced stress corrosion cracking. At present, the waste is being moved from the old tanks to the new tanks. This program will be completed by about 1988. Were the DWPF not built, additional new tanks would have been required in this and the next decade for storage of waste generated by continuing plant operations.

No injury or radiation exposure to the public has ever resulted from SRP high-level waste storage. However, storage of radioactive waste in tanks is not considered an acceptable permanent method of disposal. The concerns are three-fold. First, even though risks to the public are minimal in an operating waste tank complex, there is a possibility of leaks, accidents, natural disasters, or sabotage causing onsite radioactive contamination which would be extremely expensive to clean up. Second, if the tanks were ever placed in a prolonged storage mode, the probability of releases of radioactive waste would increase. Third, continued tank storage means repetition of the very costly process of building new tanks and transferring the waste when the present tanks eventually become unsatisfactory. The useful lifetime of the present tanks is probably greater than 20 years, but it is reasonable

to assume that a complete set of new tanks would be required in the period of 2000-2020.

The long-term solution to the nuclear waste problem at SRP is to remove the waste from tanks and immobilize that waste in a high integrity solid form. The solidified waste would then be transferred to a federal repository.

WASTE FORM

Borosilicate glass has been selected as the waste form for the design of the DWPF. Extensive research done by Du Pont and others, both nationally and internationally, on alternative forms for immobilizing waste make it clear that borosilicate glass provides a desirable combination of properties. These are: (1) adaptability to a wide range of waste compositions, (2) capability of being produced on a large scale in a radioactive environment, (3) acceptable production cost, (4) long-term integrity, and (5) low aqueous solubility. Possibly, other waste forms (e.g., crystalline ceramics) could provide slightly lower aqueous solubility or have other individual advantages, but they are harder to produce, appear to be significantly less cost-effective than borosilicate glass, and provide at best only slight improvement in immobilization of defense wastes.

France is already producing borosilicate glass containing high-level waste, and England, Germany, Japan, and Sweden are seriously considering this waste form. The most consistent objections raised against borosilicate glass forms have involved high temperature

leaching and hydrothermal reactions at temperatures above 200-250°C in a repository. These conditions are not applicable to the dilute, low heat-generating (less than 500 watts/canister) SRP defense waste forms. At spacings of about 20 feet in any acceptable repository geology, the canister surface temperature can be maintained below 100°C. At such temperatures there is little difference between the leaching characteristics of borosilicate glass and crystalline ceramic waste forms.

PROCESS

The DWPF design has been developed under DOE direction over the past four years by a multidisciplined Du Pont task force, composed of personnel from the Savannah River Laboratory (SRL), the Engineering Department, and the Savannah River Plant staff.

The process for waste immobilization (Figure 7) consists primarily of converting the sludge fraction of the waste into a glass form in a vitrification operation. Onsite aboveground storage will be provided for the glass canisters until a federal repository is developed for final emplacement, thus separating the immobilization project from the selection of a waste repository. The salt fraction of the waste is to be dissolved in the waste tanks and decontaminated by the addition of sodium tetraphenylborate which precipitates cesium as its tetraphenylborate salt. Cement and a sand/clay mixture will be added to the decontaminated salt and the resultant saltstone will be placed in an engineered burial site

at SRP. Radioactivity removed from the salt solution is added to the glass. The DWPF will convert the 30 million gallons of sludge, salt, and liquid presently in the waste tanks into approximately 1 million gallons of glass with essentially all of the radioactivity, and 16 million gallons of salt with only 0.0001% of the radioactivity.

Technology for borosilicate glass production is well developed; the preferred glass production process is a slurry-fed, joule-heated ceramic melter. The DWPF process operations have been demonstrated with actual radioactive waste in small-scale equipment in shielded cells at the Savannah River Laboratory (Figure 8). The glass process has also been scaled up to production size in a pilot plant which demonstrated equipment operability with nonradioactive simulated process solutions (Figure 9). Continuing development has provided the basis for and verification of design and operating features for the DWPF.

New technology makes it possible to treat the separated sludge and soluble salt streams in the waste tanks. To reduce the amount of borosilicate glass produced, about 75% of the aluminum compounds will be dissolved from the sludge with caustic in those waste tanks where the aluminum concentration is high; other sludge will be simply washed with water to reduce the dissolved salt content. Sludge will be removed from the tanks with special slurring pumps developed for the waste transfer program. Following the slurring and removal operations, the sludge heel in the tanks is washed to

remove soluble salts and the remaining sludge is chemically dissolved. Removal of greater than 99.9% of the waste from existing SRP waste tanks and required aluminum removal have been successfully demonstrated over the past two years (Figures 10, 11, and 12).

Decontamination of salt was likewise demonstrated in waste tanks at SRP this year. Over 500,000 gallons of dissolved salt were decontaminated by a factor of 50,000 and representative samples were removed and cast as saltstone for longevity tests. This process simplification resulted in project savings of nearly \$500 million by avoiding construction of a large, shielded remotely operated ion exchange process.

DWPF DESCRIPTION

The glass waste form will be produced in the DWPF by melting the sludge fraction of the waste and a borosilicate-type glass frit at temperatures of about 1150°C. The concentration of waste oxide will be about 28%. The glass production process employs a slurry-fed, joule-heated ceramic melter. The alkaline feed solution is to be adjusted with formic acid upon receipt in the process cell to establish reduced valence states of the sludge components. The glass will be cast and sealed in stainless steel canisters 2 feet in diameter and 10 feet long (Figure 13). About 500 canisters will be filled annually.

The vitrification of waste in the DWPF (Figure 14) will be performed in a heavily shielded concrete cell structure located

within a concrete building that is 88 feet high, 114 feet wide, and 413 feet long (Figure 15).

The basic design concept involves a system of barriers to separate the radioactive materials from operating personnel and the environment. Building walls restrict the spread of radioactive contamination and provide shielding to reduce radiation levels to less than 0.5 mrem/hr in occupied zones of the building. The ventilation system will establish differential pressures throughout the facilities to provide for air flows from zones of lesser to zones of higher contamination potential. The main processing cell is ventilated independently from the remainder of the facilities, and will operate at the lowest static air pressure within the facility.

Process cell operations are to be controlled from outside of the cell and all equipment is to be removed remotely from the cell for decontamination before repair, replacement, or disposal. Process piping and service connections will be made to the process equipment with connectors operated remotely from the cell crane. Nonradioactive chemical feed systems, instrumentation, and service systems are located outside the cell in controlled operating areas. The design concept is similar to that of the operating existing SRP reprocessing facilities which over 30 years of operation have demonstrated an availability of 85%.

Removal and installation of process equipment within the shielded building will be by remotely controlled crane with

closed-circuit television for viewing crane operations. Glass melting and canister filling and cooling will be in one segregated section of the building while canister decontamination and initial monitoring will be in a second segregated section; all of these facilities are to be serviced by the cell crane. Canister sealing and final monitoring will be in an isolated, remotely operated, contact maintenance cell. A five-inch-diameter cap will be pressed and sealed in filled canisters using a resistance upset welding technique whereby the entire joint area is welded simultaneously. All remote operations in the canister handling facilities are to be monitored by closed-circuit television; viewing windows will also be provided at critical locations. Canisters are to be moved from one cell section to a less contaminated section or between cells through tunnels which act as airlocks.

One of the major differences from commercial glassmaking is in the nature of off-gases emitted from the melter. The melter feed contains 50% water in addition to both radioactive volatile and semivolatile materials. Contamination is removed as condensables and particulates in a series of scrubbers, mist eliminators, and high efficiency filters with an overall effectiveness of 10^8 (Figure 16). In addition, the off-gas lines from the melter are kept clear of splatter and entrained molten glass by a film cooler on the throat of the exit line (Figure 17). This device cools glass particles to below their softening temperature and helps sweep them away from the pipe walls.

Service areas and operating corridors are to be housed within a seismic- and tornado-resistant structure in the space immediately surrounding the process cells (Figure 18). This space is minimized to reduce investment in the high cost structure. The process cell ventilation air will be exhausted through a single-stage sand filter of a seismic-resistant design which is expected to operate at greater than 99.9% efficiency based on similar filters installed in the two SRP reprocessing plants. Exhaust air from the operating corridors within the seismic-resistant processing building, which has a potential for some contamination, will be filtered through a single-stage high efficiency particulate air (HEPA) filter system. This latter exhaust air system will be housed in a facility of standard construction because only small amounts of radioactivity are expected to be present.

All raw material preparation facilities, nonradioactive support facilities, and personnel facilities will be of standard construction.

The waste glass canisters will be moved in a shielded transporter and stored in a natural-convection air-cooled storage building (Figures 19 and 20) until they are transferred to a federal waste repository. The storage building is a modular design that provides for storage of two years of canister production, with the intent of expanding this space as necessary until a federal repository become available. The canisters are to be decontaminated to the level which permits shipping packages of radioactive materials on public roadways.

Salt decontamination will be performed largely in existing waste tanks with the addition of minimal external facilities. After saltcake in waste tanks is dissolved with water and removed to a processing tank, chemicals are added to precipitate residual radionuclides, mainly cesium-137 and strontium-90, along with any potassium which may be present. The resultant solids, which contain 99.998% of the initial activity, are concentrated from 0.5% to 10% by cross-flow filtration across sintered stainless steel tubes. These solids can be treated to remove organics and combined with sludge to form glass. The decontaminated filtrate will be mixed with Portland cement and a sand/clay mixture to produce a high-integrity grout referred to as saltstone. The saltstone will be buried onsite in an engineered landfill which minimizes the contact of the saltstone with surface water and ground water (Figure 21).

CONCLUSIONS

Both technology and engineering have been demonstrated for the conversion of nuclear wastes at SRP to an immobile borosilicate glass. This conversion will be accomplished in facilities that have minimal environmental impact and are safe to operate and maintain. Construction has now begun and should be complete by the end of 1988. Full-scale operation should begin in 1990. The existing wastes at SRP will be completely converted by about 2010.

This project will provide the public, and the leadership of this country, with a crucial demonstration that a major quantity of existing high-level nuclear wastes can be safely and permanently immobilized. This demonstration will both expedite and facilitate rational decision making on this aspect of the nuclear program.

SOURCE OF RADIOACTIVE WASTES AT SAVANNAH RIVER

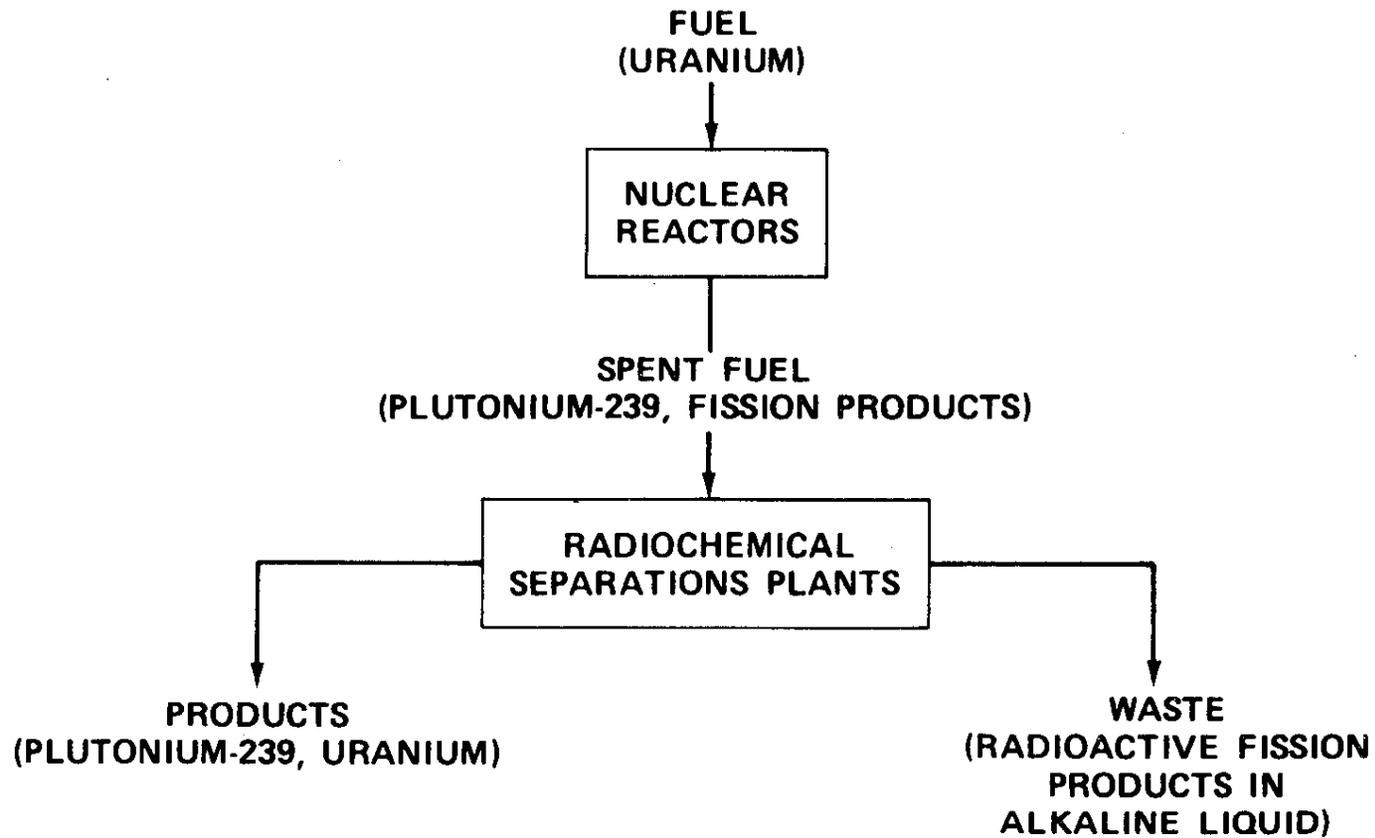


FIGURE 1.

LIQUID
AND SLUDGE

CONCENTRATED
LIQUID

SALT CAKE



RADIOACTIVE LIQUID WASTE

FIGURE 2.

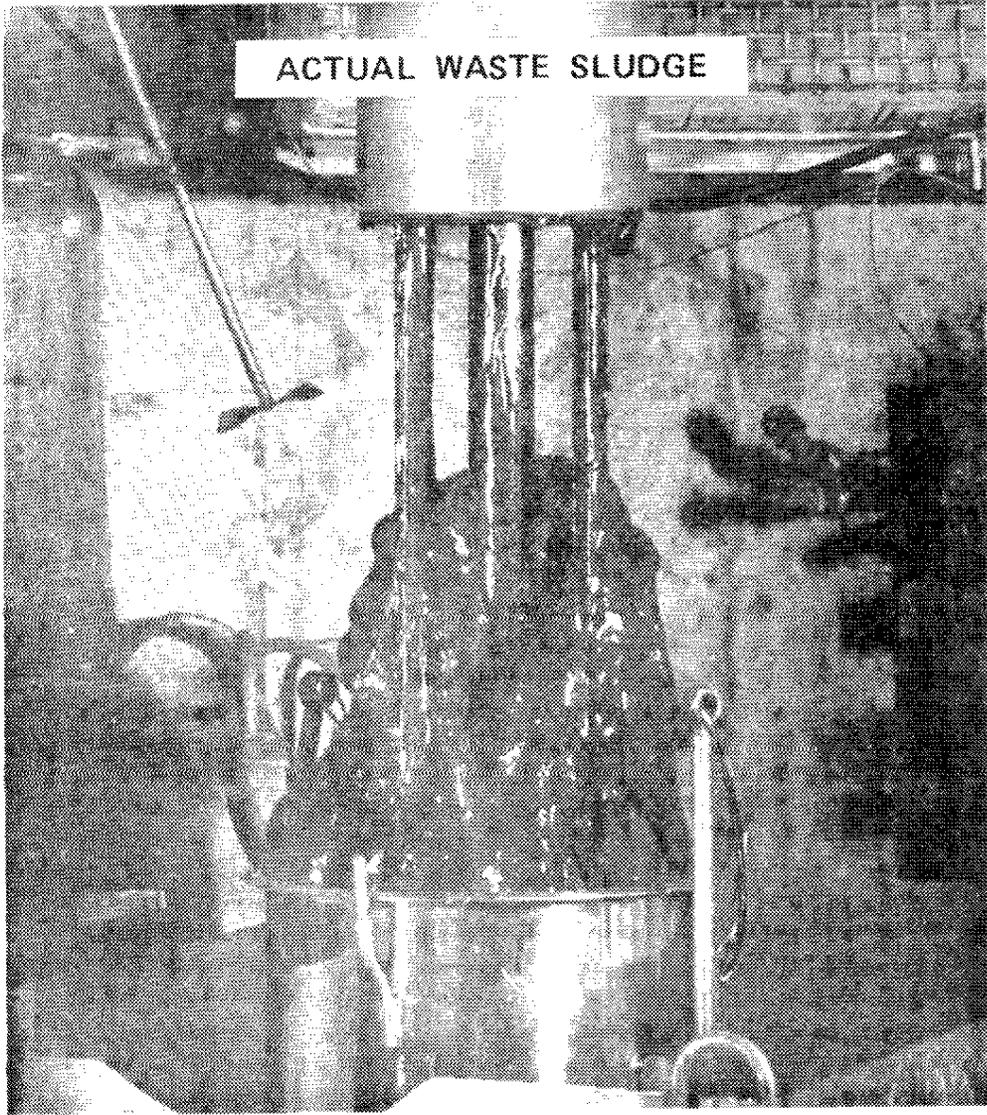


FIGURE 3.

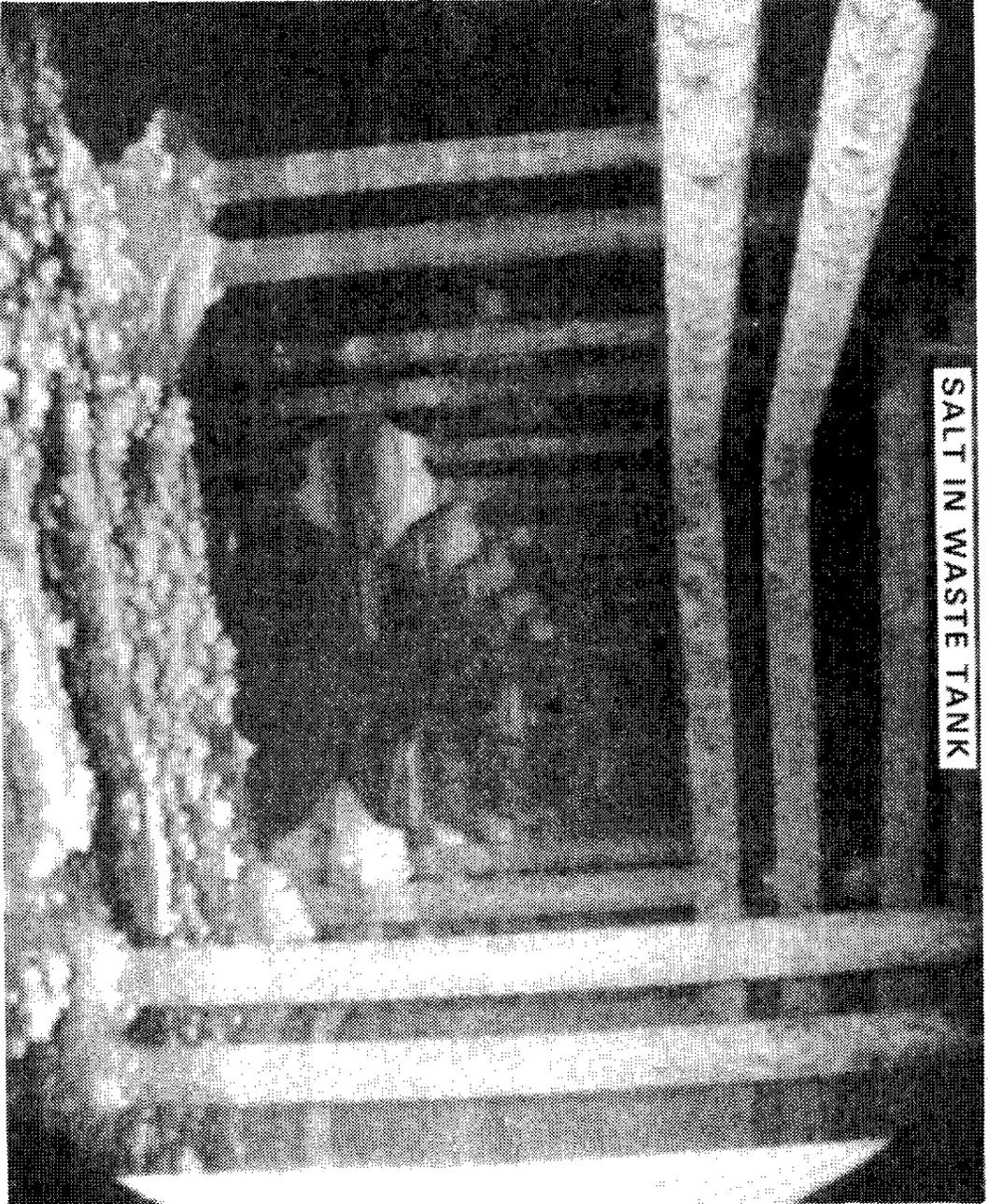


FIGURE 4.

NEW WASTE STORAGE TANK
(1,300,000-GALLON STRESS-RELIEVED TANK)

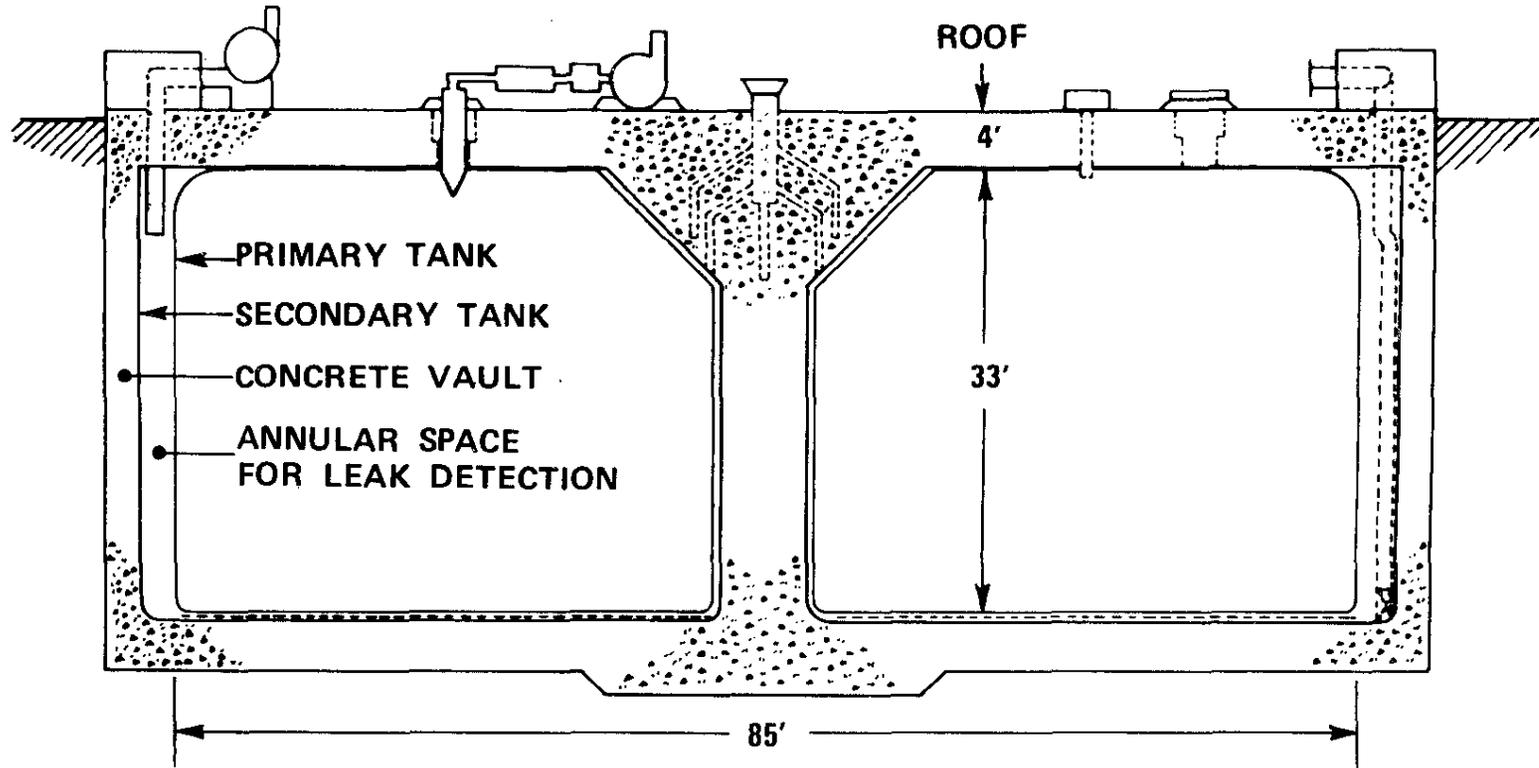
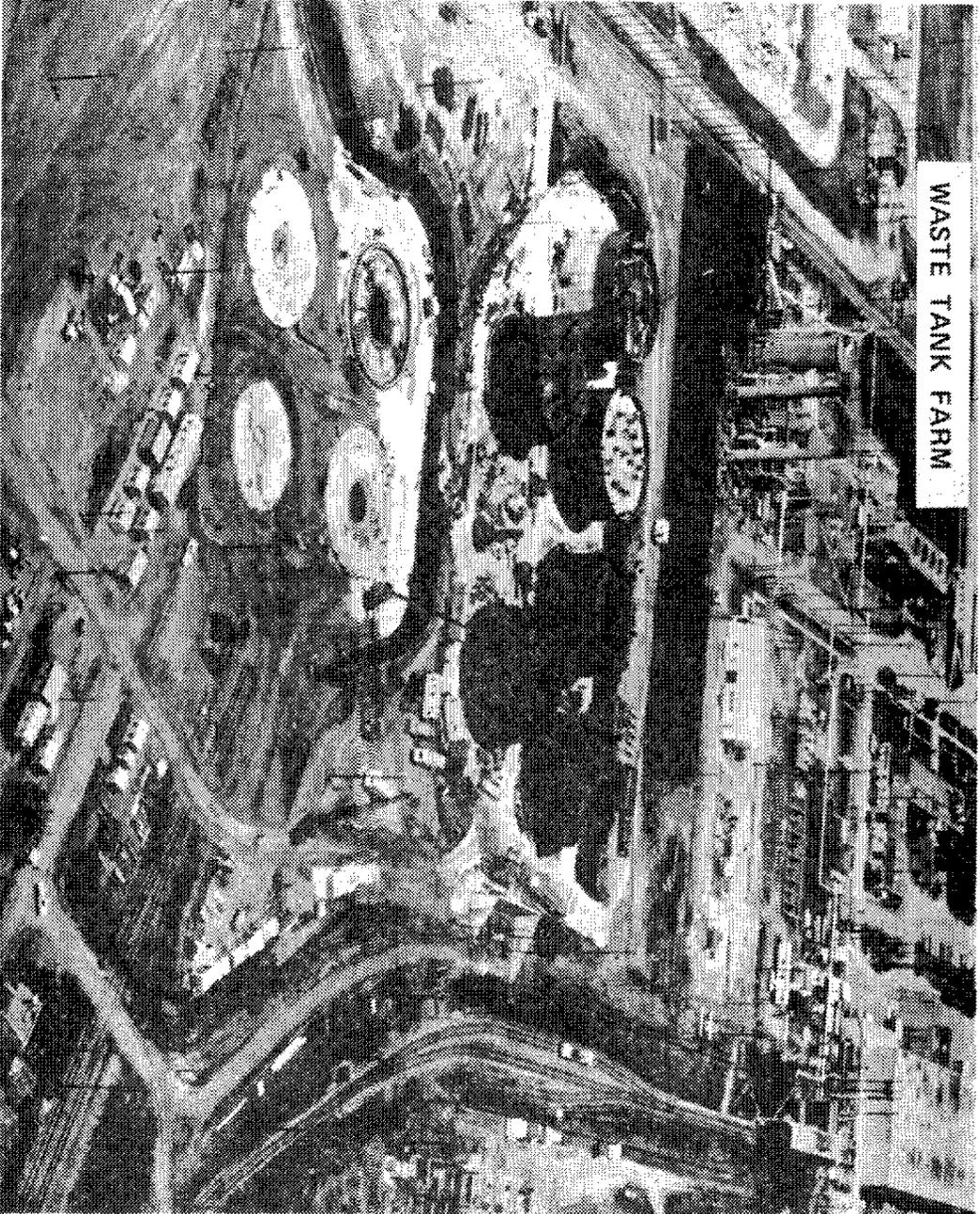


FIGURE 5.



WASTE TANK FARM

FIGURE 6.

DEFENSE WASTE PROCESSING FACILITY

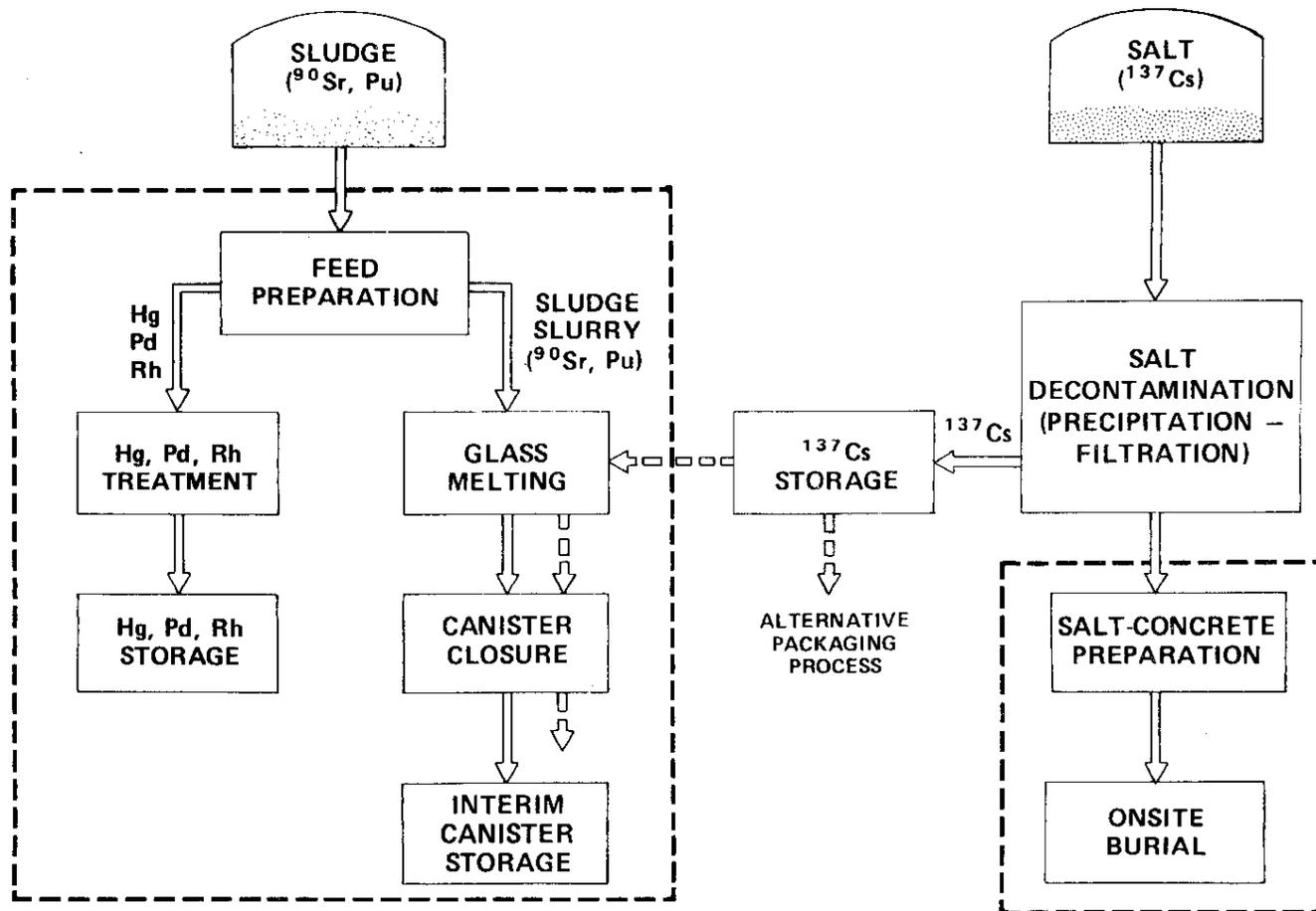


FIGURE 7.

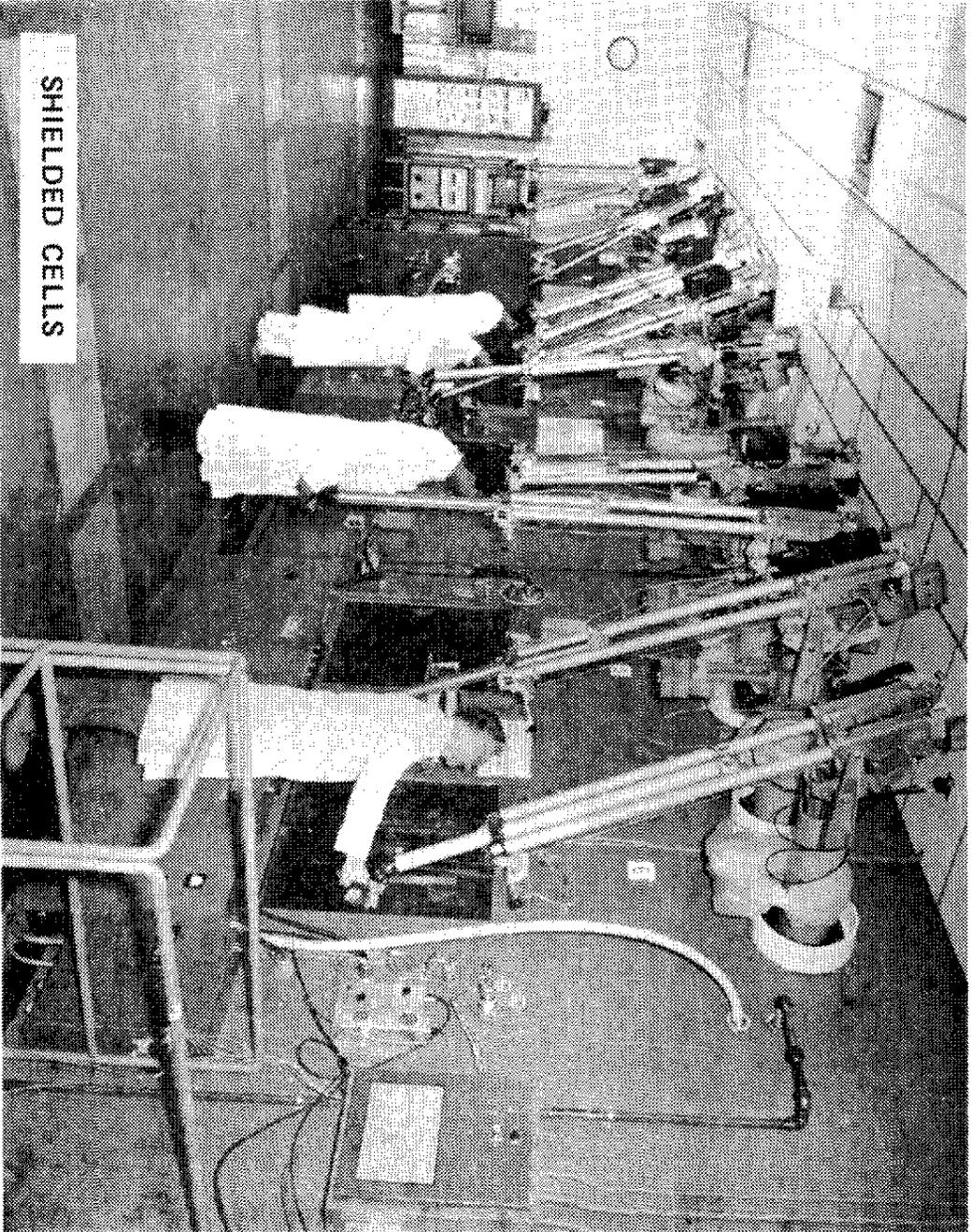
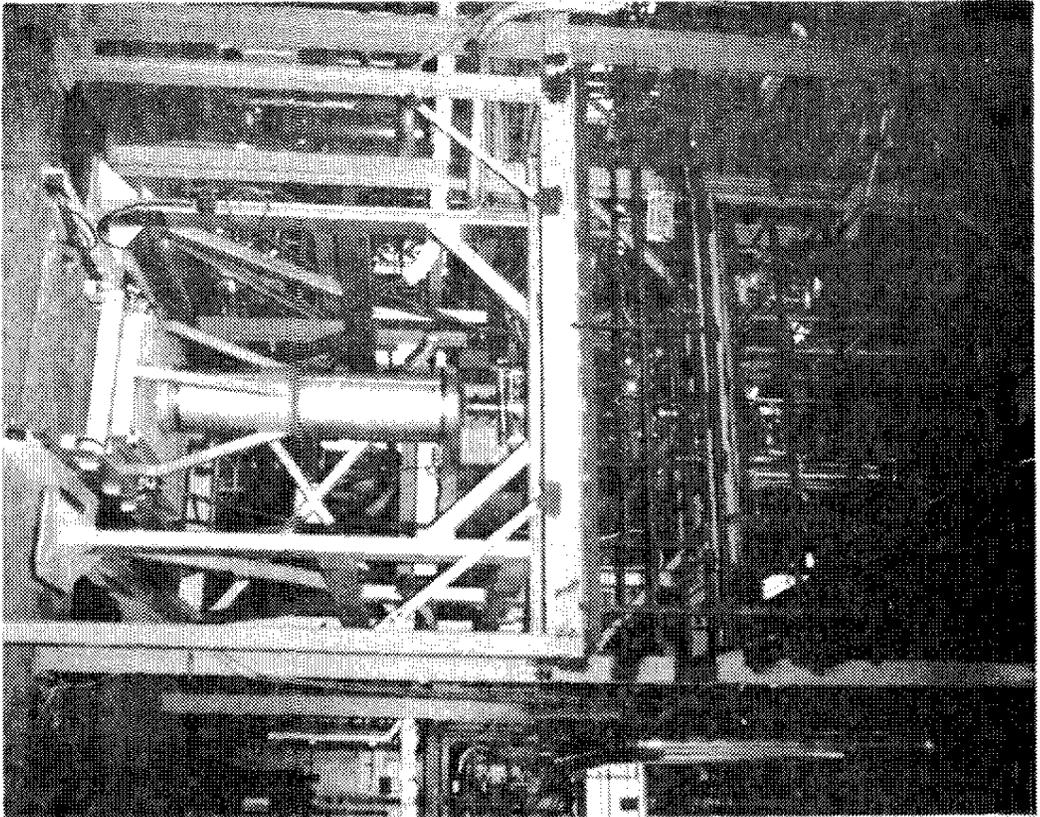


FIGURE 8.



LARGE SLURRY-FED MELTER

FIGURE 9.

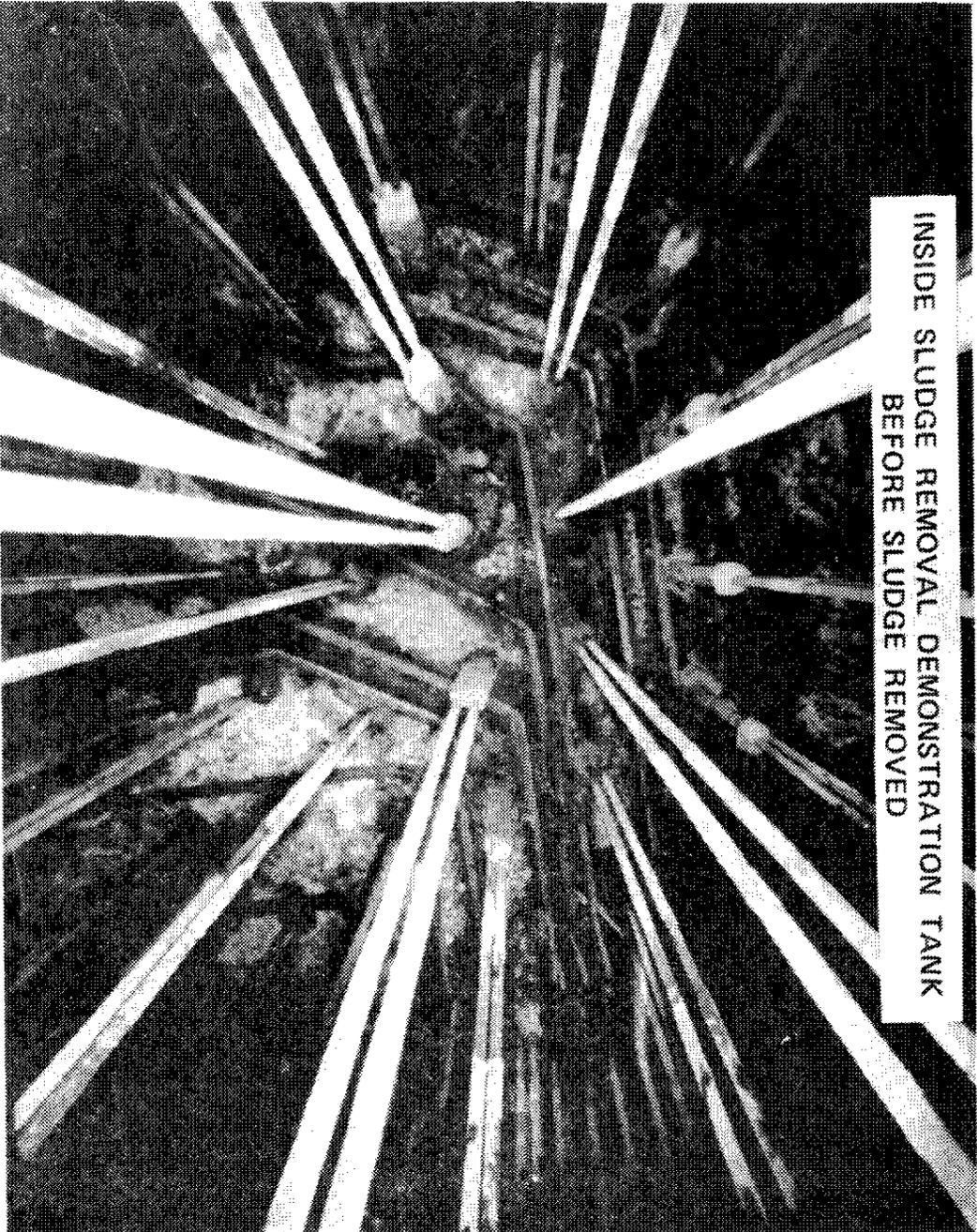
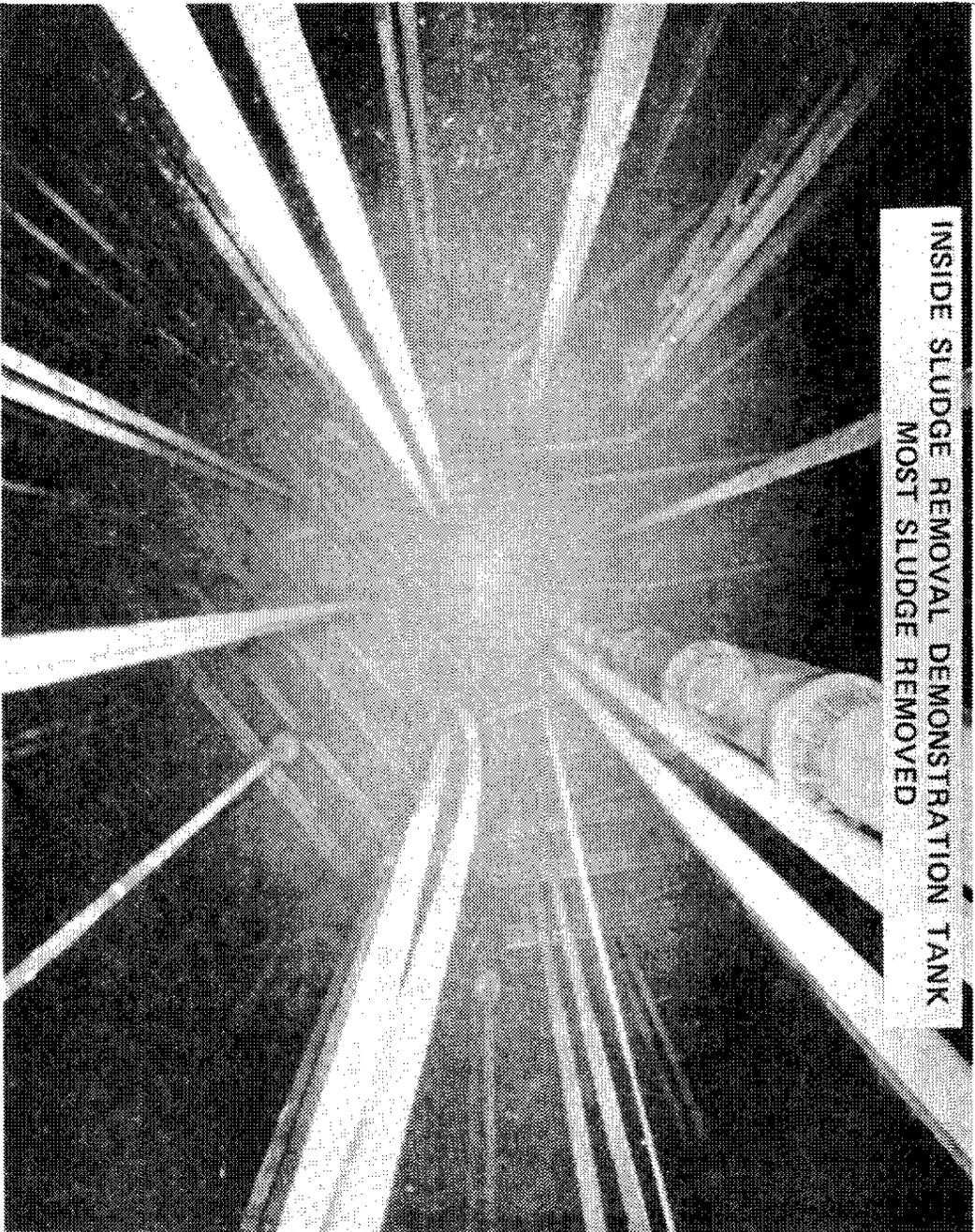


FIGURE 10.



INSIDE SLUDGE REMOVAL DEMONSTRATION TANK
MOST SLUDGE REMOVED

FIGURE 11.

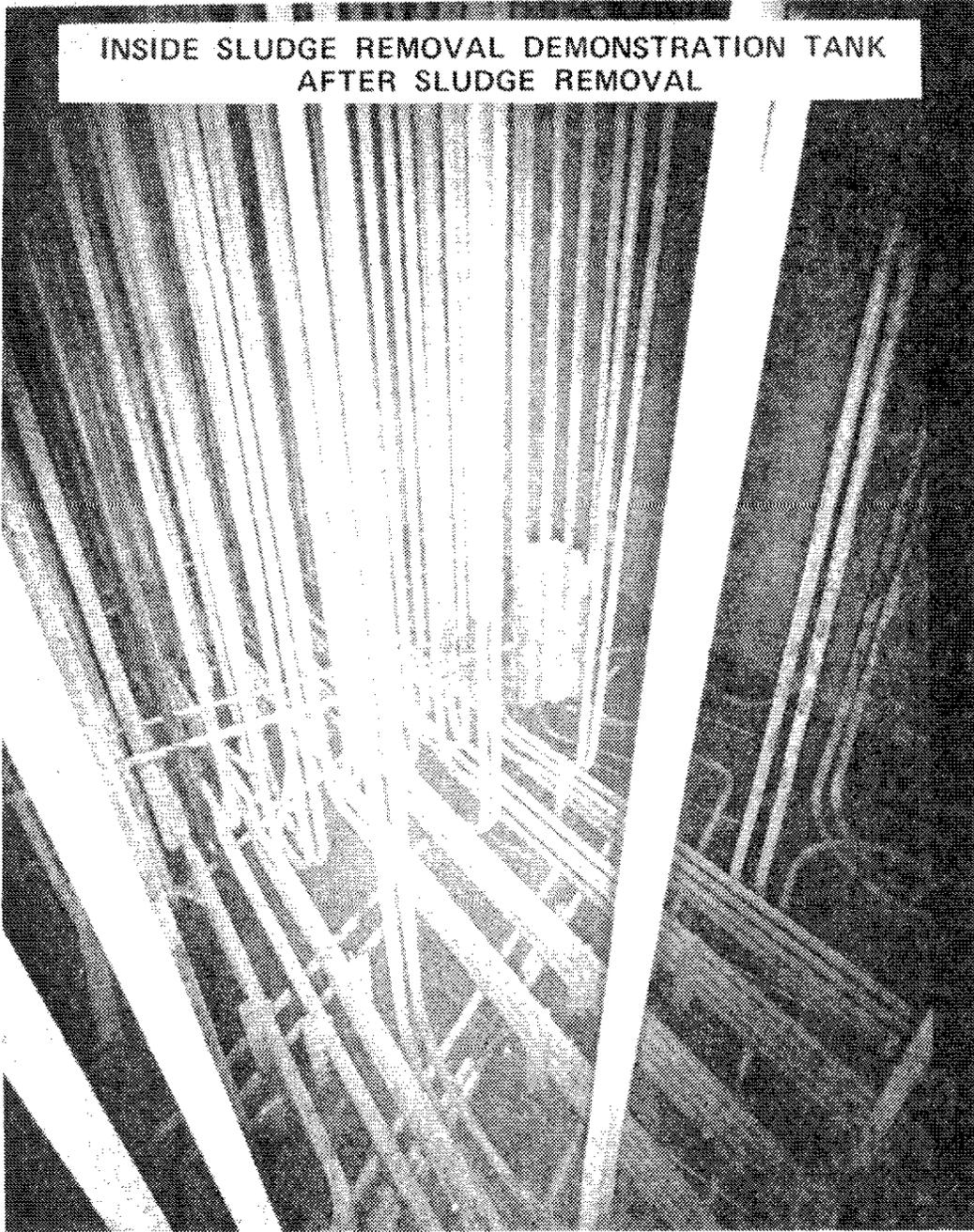


FIGURE 12.

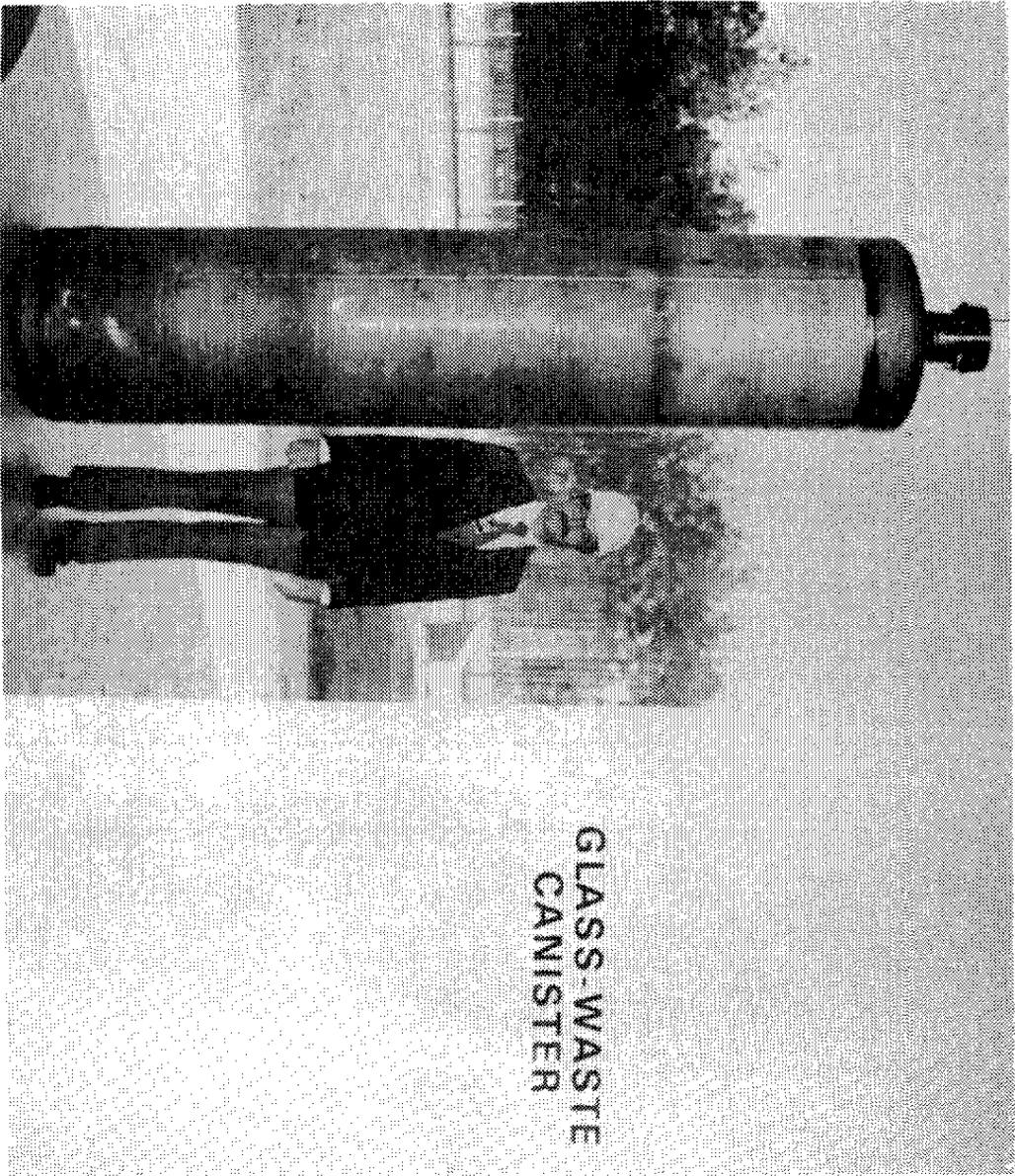


FIGURE 13.

DEFENSE WASTE PROCESSING FACILITY

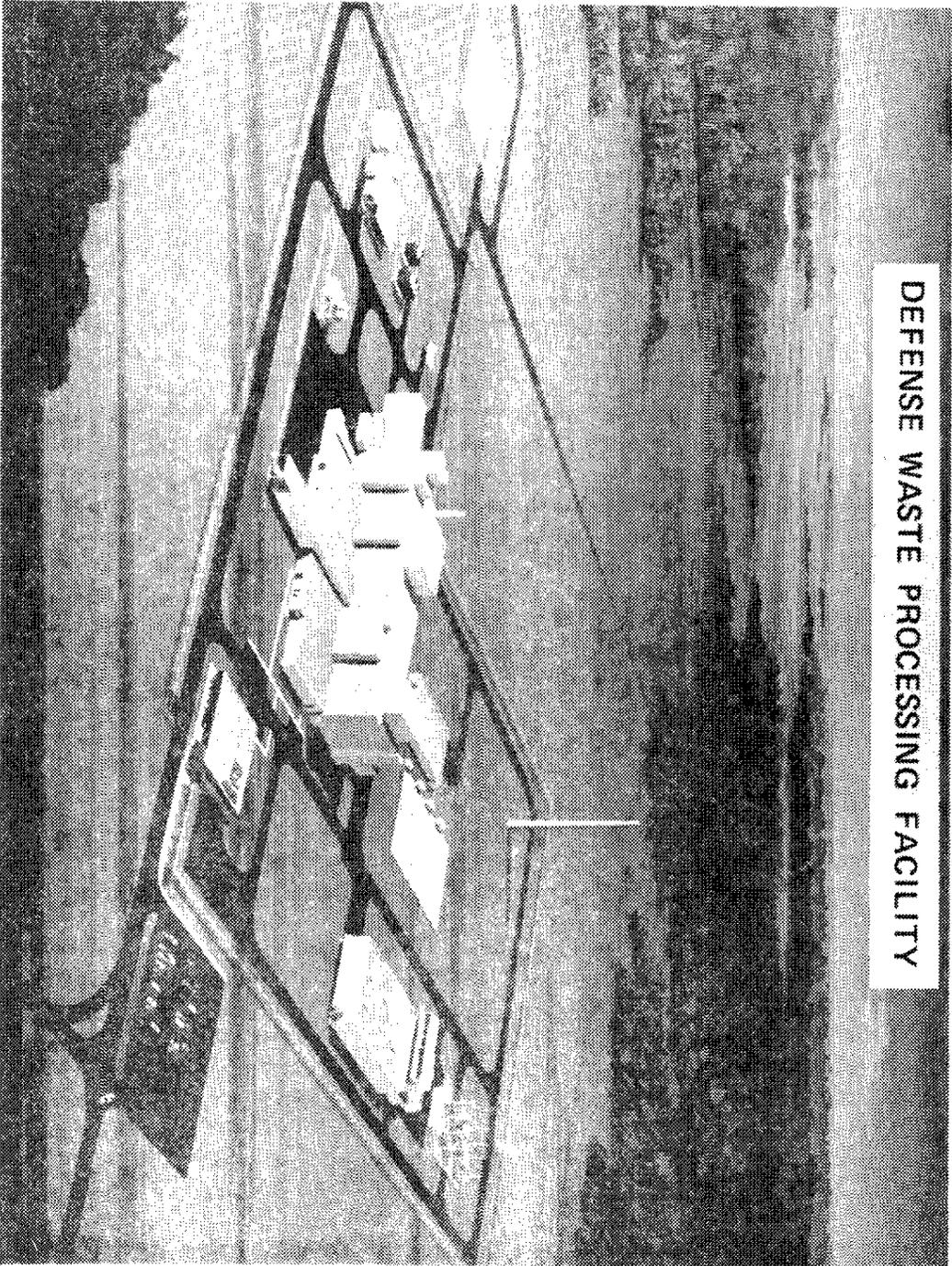


FIGURE 14.

DWPF BUILDING 221-S - LEVEL 1

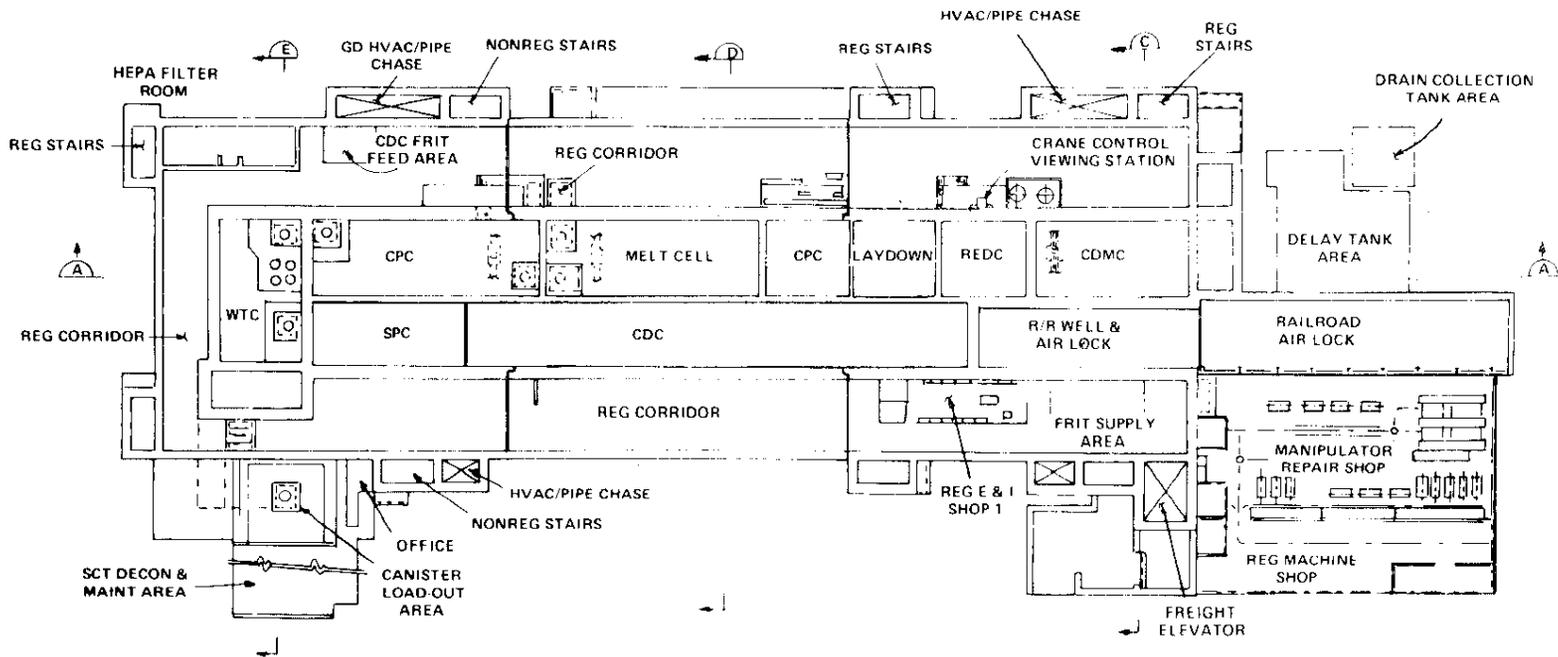


FIGURE 15.

DWPf MELTER OFF-GAS SYSTEM

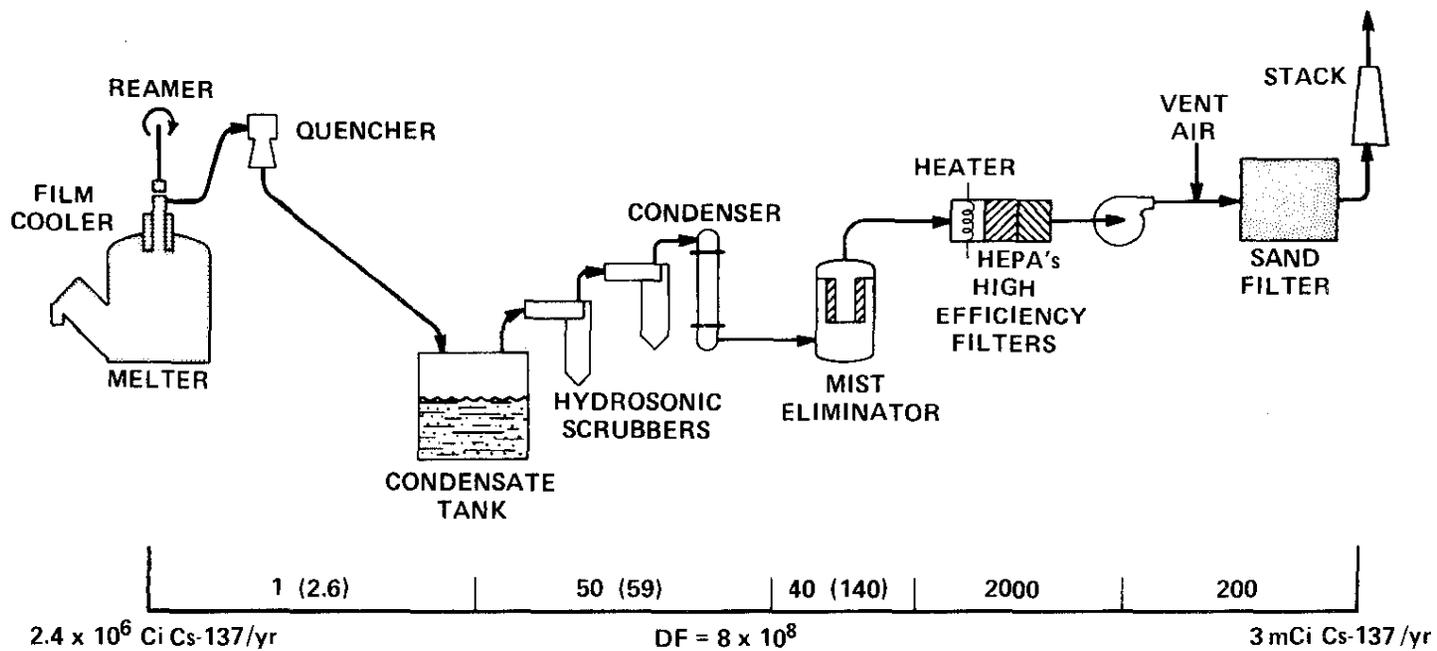


FIGURE 16.

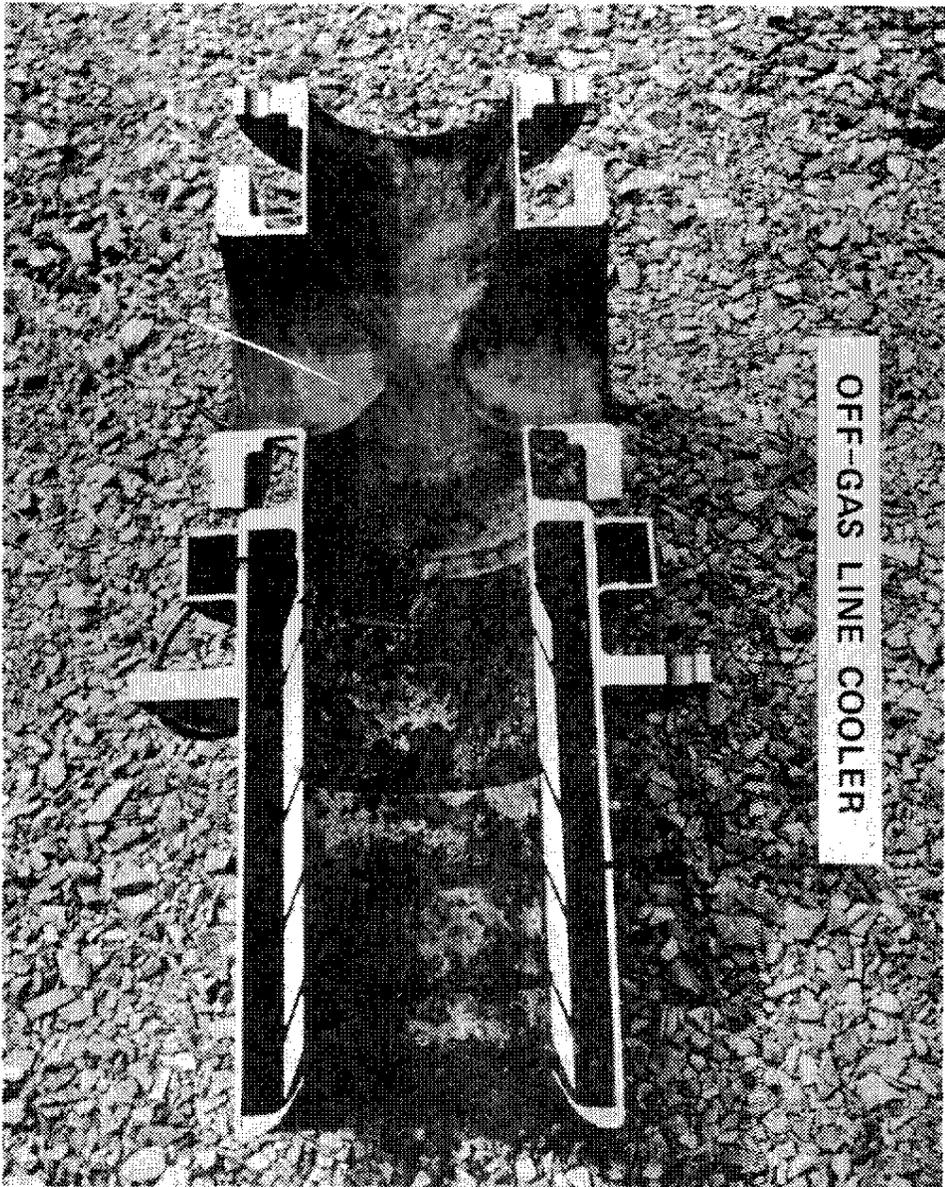


FIGURE 17.

DWPF BUILDING 221-S - SECTION C

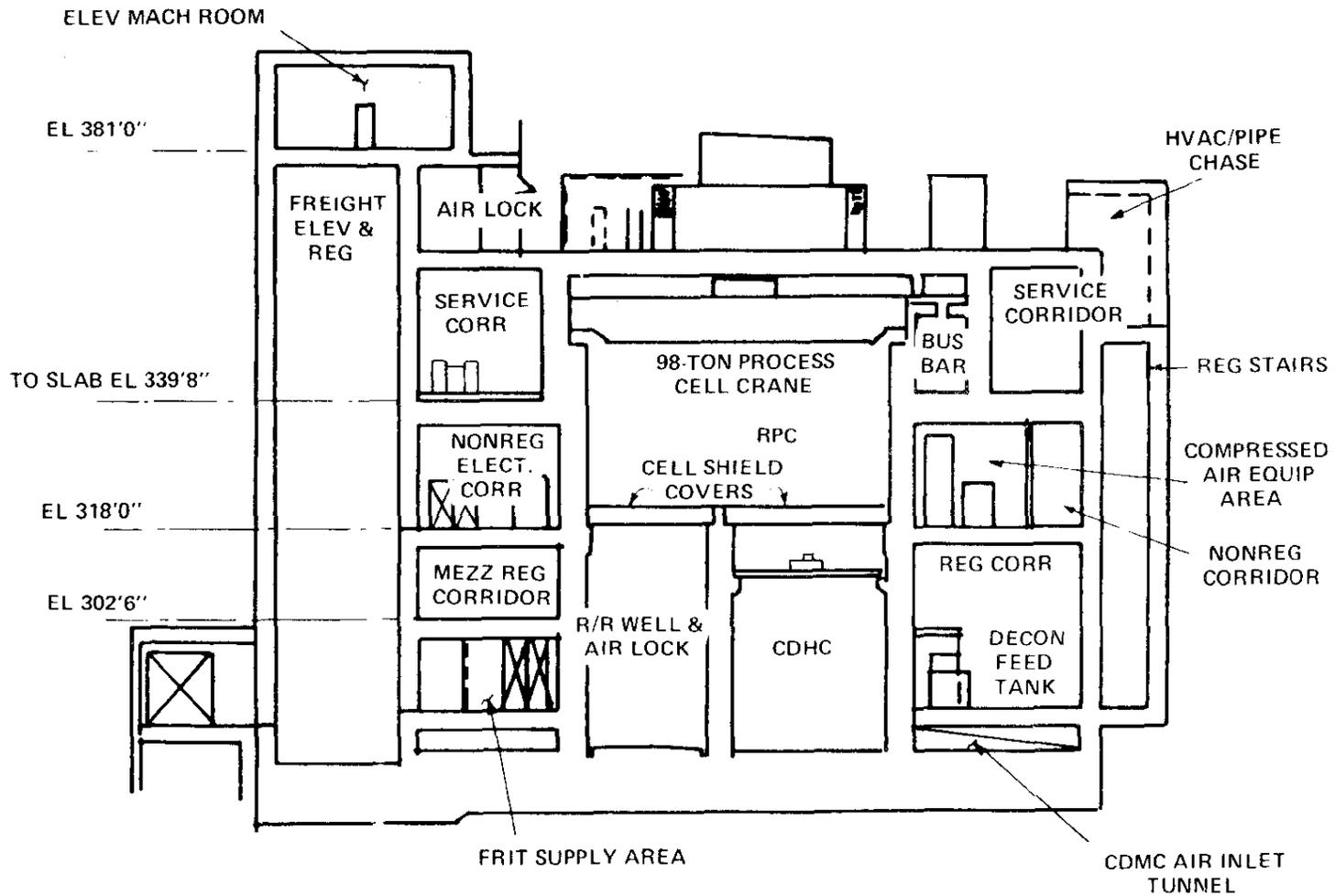


FIGURE 18.

INTERIM STORAGE BUILDING

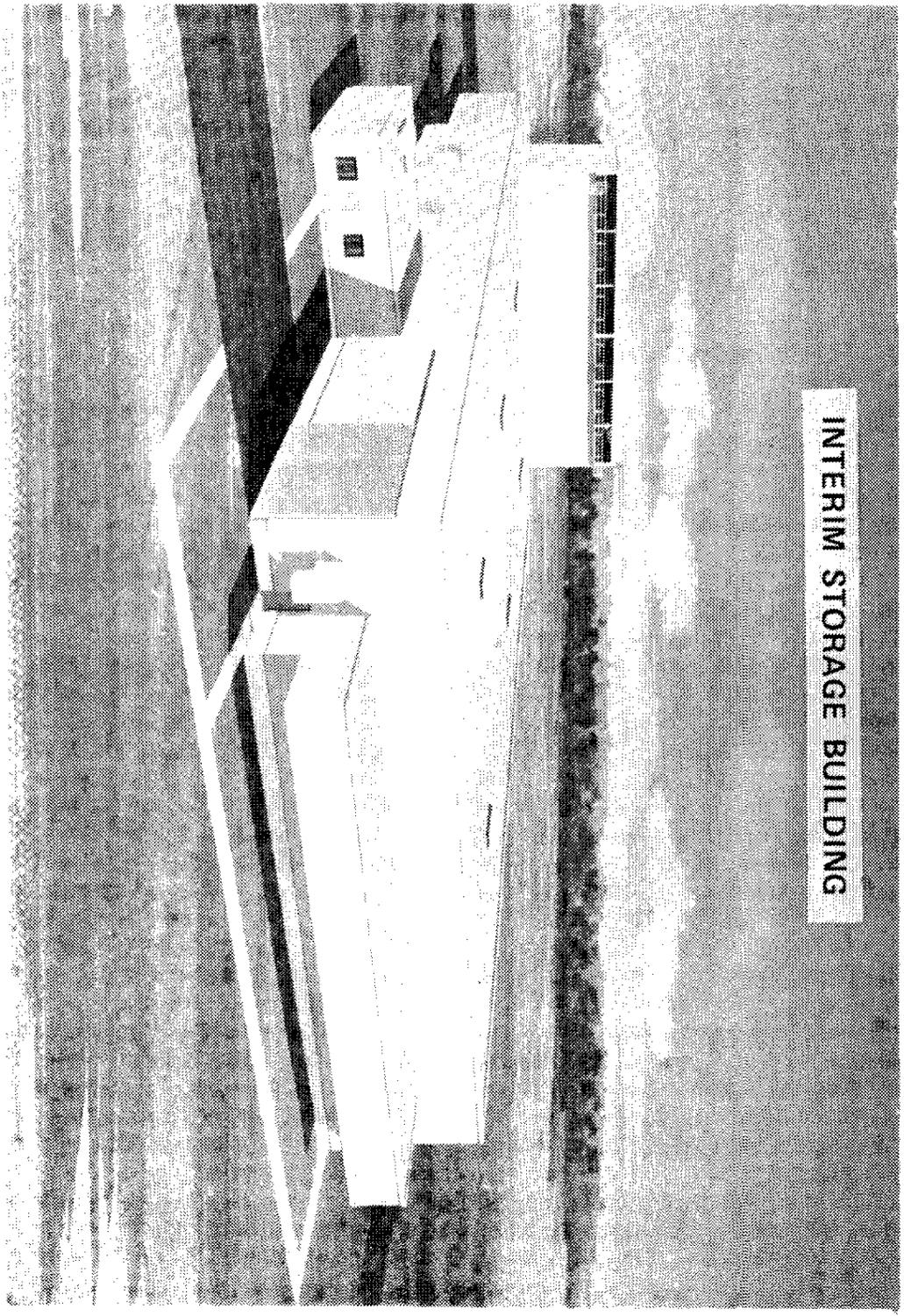


FIGURE 19.

GLASS STORAGE BUILDING
SECTION B - B

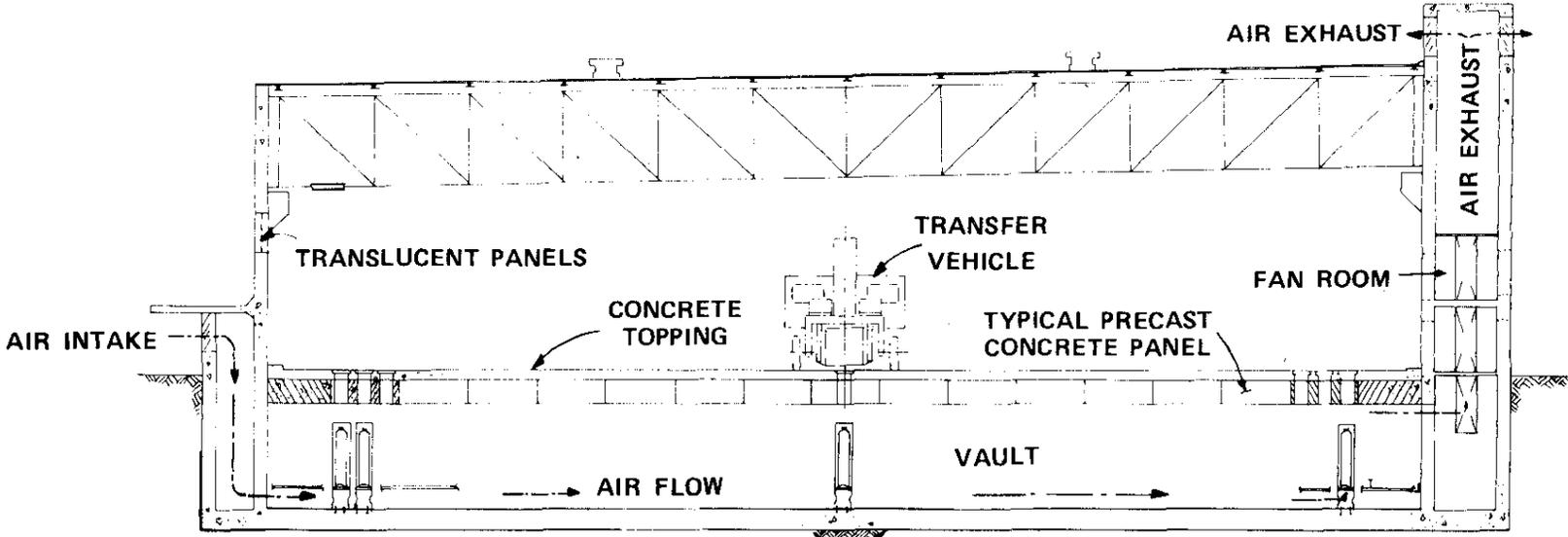


FIGURE 20.

SALTSTONE MONOLITH CROSS SECTION

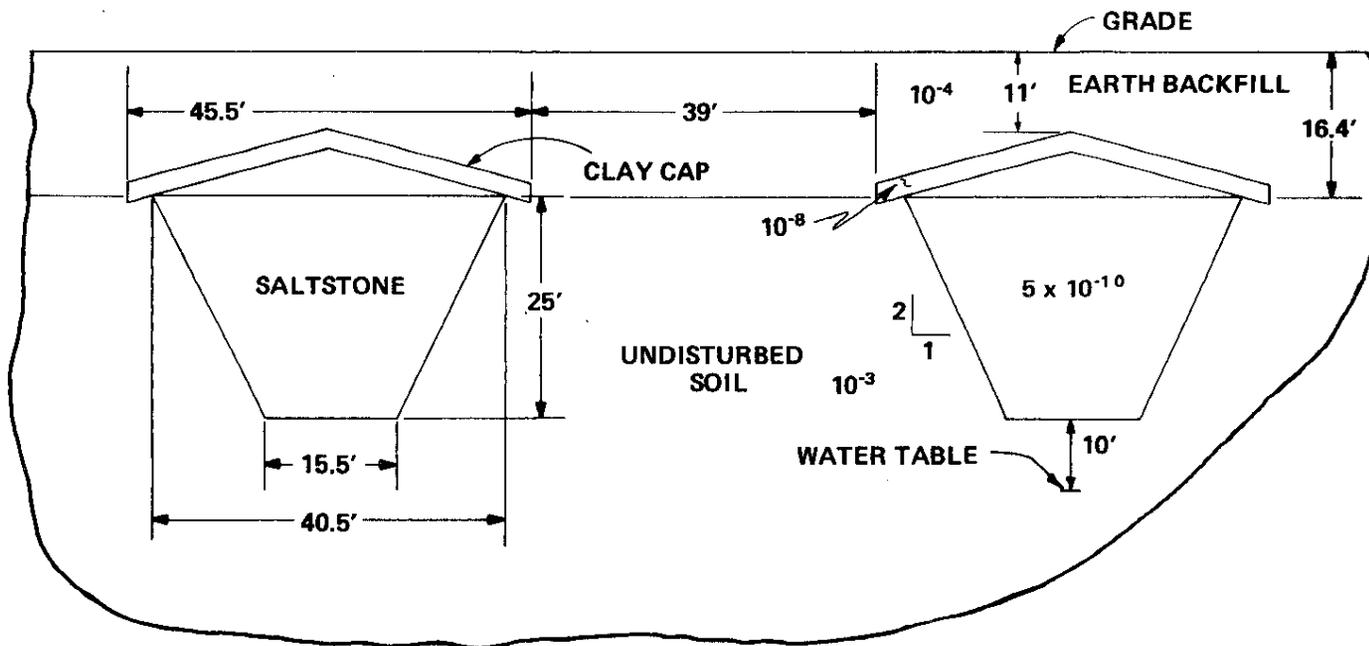


FIGURE 21.