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DP-MS-80-39

HIGH LEVEL NUCLEAR WASTE

by

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For publication and presentation at the 17th Annual Symposium of the Idaho Section of the American Society of Mechanical Engineers,
Idaho Falls, ID
on May 29, 1980

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ABSTRACT

The DOE Division of Waste Products through a lead office at Savannah River is developing a program to immobilize all U.S. high-level nuclear waste for terminal disposal. DOE high-level wastes include those at the Hanford Plant, the Idaho Chemical Processing Plant, and the Savannah River Plant. Commercial high-level wastes, for which DOE is also developing immobilization technology, include those at the Nuclear Fuel Services Plant and any future commercial fuels reprocessing plants. The first immobilization plant is to be the Defense Waste Processing Facility at Savannah River, scheduled for 1983 project submission to Congress and 1989 operation. Waste forms are still being selected for this plant. Borosilicate glass is currently the reference form, but alternate candidates include concretes, calcines, other glasses, ceramics, and matrix forms.

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*The information contained in this article was developed during the course of work under Contract No. DE-AC09-765R00001 with the U.S. Department of Energy.

INTRODUCTION

High-level nuclear waste is the chemical waste resulting from reprocessing irradiated reactor fuels and targets to recover residual fissionable materials and other values. Although not necessarily the most voluminous, it is certainly the most radioactive nuclear waste. It contains better than 99 percent of the active fission products plus any unrecovered uranium, plutonium, and higher actinides together with varying amounts of inert fuel materials and reprocessing chemicals. My talk will discuss the management of this waste from cradle to the grave, that is, from its generation in the reprocessing plant to its disposal in a repository. I will, however, be placing special emphasis on the preparation of disposal forms. The problems of interment, both for the high-level reprocessing waste and for the alternative of direct disposal of unprocessed spent fuel, were covered in the earlier talk by Mr. Hall from the Office of Nuclear Waste Isolation.

HIGH-LEVEL WASTE DESCRIPTION

The first set of questions I'd like to address is: What are the hazards of these wastes? How long do the hazards last? What do we have to do about them? The hazards are almost exclusively radiological. Although there are some chemical poisons present, they are not too serious. Nitrate is in fact the worst, in quantities rather less than the farmers around Idaho Falls use to fertilize their fields.

My first slide shows a time curve of the radiological activity. It falls into three main periods:

- A high activity period of five to ten years dominated by the radionuclides such as ^{106}Ru, ^{144}Ce, and ^{147}Pm which have half-lives of a year or less.
- A medium activity period of five hundred to a thousand years dominated by ^{90}Sr and ^{137}Cs, both of which have half-lives of about 30 years.
- A low activity period extending out many thousands of years dominated by the higher actinides, the uranium decay product, and a few very long lived fission products such as ^{99}Tc and ^{129}I.

The overlay to this slide then compares these activities with some other hazardous materials. The conclusions are that the high-level wastes are indeed very hazardous, but not appreciably more so than a number of other materials in common use. Also, in something less than a thousand years their hazard has decayed to about the same level as the uranium ores which were mined to produce the reactor fuels from which the wastes were derived.

This analysis would seem to require very careful but not extraordinary protection of the wastes for about a thousand years, and such is indeed the recommendation of a draft ASME standard on these wastes. Not surprisingly, however, the regulatory agencies are asking for considerably more, as shown on my second slide. Draft criteria from the Environmental Protection Agency ask that the waste be protected so as to

produce a risk of only 200 health effects in the next ten thousand years - out of 10^{12} natural deaths in the same time period - and that these risks be mainly exotic ones such as meteorite strikes rather than routine operating risks. Draft regulations from the Nuclear Regulatory Agency in turn ask that the repository and waste package (i.e., the form plus its canister and any further engineered barriers) be completely redundant to each other and that the package alone guarantee zero release for 1,000 years and 10^{-5} to 10^{-7} release per year for 10,000 years.

HIGH-LEVEL WASTE INVENTORIES

The second set of questions I'd like to address concerns what are the U.S. high-level waste quantities and where are they located? Those questions are covered in my third slide. Not surprisingly, since high-level wastes are reprocessing wastes, they're located at the reprocessing plants. These plants comprise the three defense plants, here at Idaho, at Hanford, and at Savannah River, plus the only U.S. commercial reprocessing plant, the now defunct Nuclear Fuel Services (NFS) Plant at West Valley, NY. By 1990, the three defense plants are each expected to have 500 to 750 megacuries of HLW in storage. The NFS waste quantities are much smaller, about 48 megacuries, but, if commercial fuel reprocessing is ever resumed, the quantities of spent fuel now in storage would produce a high-level waste activity about equivalent to that in the existing defense wastes, and by 1990 they would produce about 10 times the activity of the

defense wastes. As I'll be discussing in a few moments, the Hanford, Savannah River, and NFS wastes are all relatively dilute alkaline wastes, while the Idaho, and presumably any future commercial wastes, are much more concentrated acid calcines.

THE DEFENSE WASTE PROCESSING PLANT

The set of questions to which I'd now like to devote most of my talk is: What are we going to do about these wastes? Even though my responsibilities are for the national HLW program, I'm going to address these questions mainly in terms of my own site, Savannah River. The reasons are that Savannah River is scheduled to be the first U.S. site to dispose of its high-level wastes and its alkaline wastes offer rather more complex disposal problems than the calcines here at Idaho.

The quantities of high-level waste at Savannah River are listed in Slide 4, 68 million gallons as originally generated, now evaporated to 23 million gallons with a continued generation of 1.5 million gallons per year (which will eventually also be evaporated to about 0.5 million gallons per year net addition). As I mentioned earlier, this is alkaline waste, meaning that the nitric acid waste solutions coming out of the Purex reprocessing plant have been neutralized with excess sodium hydroxide with results as shown on Slide 5. The greatest bulk of the waste is then the sodium nitrate and sodium hydroxide neutralization products, which are very water soluble, but almost all the fission products and other active materials - except ^{137}Cs - have been

precipitated out as a highly insoluble sludge of oxides, hydroxides, and carbonates. The decision to go to alkaline waste was made some thirty years ago for the purpose of tank storage and still remains a very sensible decision for that purpose. It produces a dilute low-heat waste, which immobilizes the radionuclides well in insoluble sludge or in soluble salt, which can be contained in carbon steel tanks, and which, if it leaks from the tanks, will peptize the Savannah River soils to produce a nonmigrating plug. However, as we will see, alkaline wastes have proved to be a very expensive decision if it is desired to make high-integrity disposal forms from them.

Slide 6 shows a current Savannah River storage tank. It is essentially triple walled, with primary and secondary carbon steel tanks and an outer concrete tank. Although some of the earlier tanks had problems with stress corrosion cracking, the current tanks are stress relieved and are expected to have lifetimes of 50 to 100 years or longer. Slide 7 shows a picture of the soluble waste fraction, which evaporates to a nice crystalline salt, while Slide 8 shows a picture of the sludge (actually from Hanford, but identical to SRP's). As one of our engineers remarked when he saw this slide, he knew the sludge looked like peanut butter but he hadn't been told it was the chunky kind.

Slide 9 gives the overall plan for disposing of the SRP high-level-waste. This plan involves removing the waste from the tanks and then taking advantage of the natural sludge-solution

separation to divide the waste into about 16 million gallons of low-level salt and about 250,000 gallons of high-level sludge to which has been added the ^{137}Cs and other activities from the salt after removal by ion exchange. The reference SRP design is to put the salt into concrete monoliths and the sludge and separated radionuclides into about 1 million gallons of borosilicate glass for terminal disposal. However, as I will be discussing at the end of my talk, a final decision for a glass waste form has not been made, and some twenty alternate forms are under active development.

Waste removal from the tanks is well demonstrated at Savannah River and is not expected to offer problems there. It may be a major stumbling block at Hanford, where the waste is in some 156 tanks, some of which are up to forty years old and may not have the integrity required for the hydraulic mining approach used at Savannah River. This hydraulic mining method is illustrated schematically in Slide 10. Enough water is added to dissolve the salt, which is then pumped out in solution. More water is added, and special slurrying pumps are inserted through risers in the top of the tank to mechanically disperse the sludge into this water, which is then also pumped out. Slide 11 shows the interior of one of the SRP waste tanks after the salt removal step, while Slide 12 shows the same tank after sludge removal. As you can see, the tank looks quite clean after these two treatments, with 95 to 98 percent waste removal. Final cleaning to 99++ percent waste

removal is then effected by chemical dissolution with hot oxalic acid solutions and possibly with H_2O_2 solutions.

The separate salt and sludge streams are pumped to a proposed new facility, the Defense Waste Processing Facility or DWPF, illustrated schematically in Slide 13. This facility will complete the sludge-salt separation, remove the high-level salt activities by ion exchange, and prepare the HLW waste forms. The salt will go directly to onsite disposal while the HLW waste forms will be accumulated in an onsite storage facility (good for up to 100 years storage) until our friends at the Office of Nuclear Waste Isolation have a repository for us.

Slide 14 is an artist's rendition of the current DWPF design. The main processing building is a canyon-type facility. That is, all equipment is operated and maintained remotely behind heavy concrete shielding walls in an environment which is never entered by humans during its operating life. Although this approach is initially more expensive than a contact-maintained facility, our experience in operating the reprocessing canyons at Savannah River for nearly thirty years without a major incident and with about 90 percent process innage, has convinced us it is the preferred way to go.

Slide 15 is a cross section of the proposed processing building showing the double-canyon arrangement and the most unique piece of

equipment, the calciner-melter used to produce the borosilicate high-level waste forms. This calciner-melter is drawn in more detail in Slide 16. The spray calciner operates at 300 to 400°C primarily to dewater the sludge - ^{137}Cs zeolite slurry feed. The calcine then falls into an electrode-heated glass melter, joule-heated ceramic melter, or JCM in the jargon. In the melter, the calcine is mixed with glass frit and heated to about 1150°C to form a borosilicate glass containing 20 to 30 percent waste oxides, 30 to 40 percent silica, 5 to 10 percent B_2O_3 and 10 to 15 percent alkali oxides plus other additives. This glass is then tilt-poured into a stainless steel canister about 2 feet in diameter and 10 feet long, typically containing about 165 gallons of glass. This glass-waste system has been extensively developed at both Pacific Northwest Laboratories (PNL) and Savannah River. A large number of nonradioactive full-scale canisters, as illustrated in Slide 17, have been made and tested. Two full-scale radioactive canisters have also been made at PNL, and large numbers of smaller-scale radioactive specimens have been prepared at a number of sites. Further, France has a commercial plant, in operation at Marcoule, to produce borosilicate glass high-level waste.

The other waste stream from the DWPF is the low-level salt waste. It is proposed to fix this salt in large concrete monoliths which would be buried at depths of about 30 feet in the SRP burial ground as depicted in Slide 18. According to draft criteria from the Nuclear Regulatory Commission the radioactivity

levels in this salt are such - about 300 nanocuries per gram - that it can be placed in this intermediate-depth burial with no further surveillance.

There appears to be essentially no doubt that the DWPF, as I have described it, can be built and would make a technically acceptable disposal system for the Savannah River high-level waste. The Du Pont Company, through DOE, is in fact preparing to recommend it to Congress for a 1983 project with a projected operating date of 1989. There are, however, two major possible pitfalls to be resolved. The first concerns the facility cost, which, as shown by Slide 19, is currently estimated at $1.5 billion in 1979 dollars or $2.4 billion in escalated project dollars. An aggressive research program has reduced this cost nearly half a billion in the last year and a half and is continuing to look at items such as preliminary processing in the waste form, slurry-fed melters and more compact canyon designs to reduce these costs still further. Staged construction of the facilities might be another way to reduce at least the initial cost impact. Still, the mere size of the project will make the approval process arduous.

HIGH-LEVEL WASTE FORMS DEVELOPMENT

The other possible pitfall is the waste form. We do regard borosilicate glass as an acceptable form. Slide 20 for example, which I borrowed from PNL, shows that its leachability compares very favorably with most common rocks, and it has been demonstrated to have outstanding radiation resistance. Still, it may not be

the best form, and both NRC and a number of review groups have indicated they would like to apply the "as low as reasonably achievable" (ALARA) principle to demand the best form. Accordingly, DOE is pursuing a major program to develop alternate high-level waste forms.

Since, as shown by Slide 21, most of the DWPF is concerned with salt sludge treatment rather than waste form preparation - the reason why alkaline waste is such an expensive choice for disposal - the decision on the waste form for the DWPF can be left until the project approval date in 1983. The DOE alternate waste form schedule is therefore geared to that date. Slide 22 shows a three-phase program. The first phase, in progress until the end of 1981, examines essentially all available forms. The second phase, in progress until the end of 1982, then examines the three or four "best" forms, i.e., those that the initial study has shown to have the greatest potential of demonstrating better properties than borosilicate class, in further detail. A choice between them is made at the end of 1982. The final phase is then to perform the detailed engineering design on the winning form, either borosilicate glass or one of the alternates, so as to be able to get the DWPF in operation by 1989. The winning form will obviously be chosen by regulatory, environmental, public, and finally Congressional action as well as by DOE and Savannah River. A continuing program will still proceed on second generation forms even after the DWPF form choice is made.

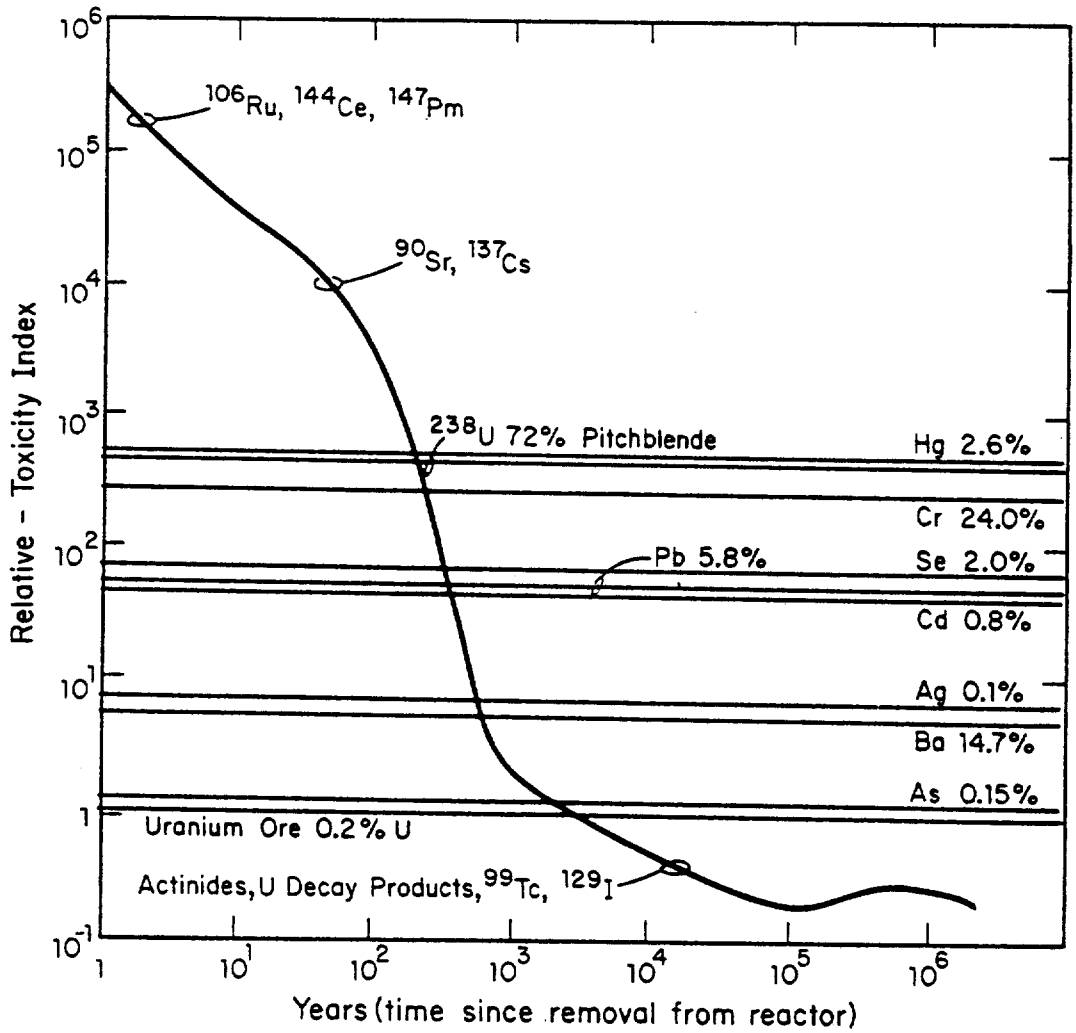
The criteria to be used in the waste form choice are listed in Slide 23. I won't dwell on them except to indicate that they are many and varied and must extend over the whole waste management system rather than focusing exclusively on leachability in the repository.

Slide 24 lists the waste forms being considered in the present stage of the program. The in-place solidification forms are known to be inferior to borosilicate glass, but are being pursued against the contingency that removing some of the older defense wastes for immobilization may offer greater hazards than hardening them in place. Normal concretes are also known to be inferior to borosilicate glass, but special concretes which have been hot pressed to eliminate excess water and reduce void volumes and leaching areas are still being actively pursued as well as concretes which essentially act as matrices for highly insoluble particles such as supercalcines. Calcines are again known to be inferior to borosilicate glass, but are useful intermediates and might be pelletized to make acceptable forms. Among the glasses, both high silica and high alumina glasses are known to be superior to borosilicate glass, the problem being to prepare them at temperatures low enough so the ^{137}Cs and other radionuclides are not volatized out in the glass formation. Ceramic waste analogues of minerals which have contained radioactive elements for long periods in nature are viewed as particularly attractive form candidates.

Two main mineral series are being investigated, the silicates as exemplified by the supercalcines originally developed at Penn State and the titanates as examplified by SYNROC originally developed at the Australian National University. The primary question on these materials is their relative complexity. The most complex, but perhaps the most advanced forms of all, however, are the matrix forms in which any of the earlier waste forms are dispersed in metallic or other lattices, with or without protective coatings on the waste particles before they are put in the matrix.

The forms developers are tabulated in Slide 25. The attempt has been to disperse them widely through the DOE laboratories, industry and universities. The intent is not only to secure the broadest possible input, but to let each developer act as a proponent for his particular form.

Finally, and I hope appropriately, Slide 26 is a pretty picture of some of the waste forms which the Idaho Chemical Processing Plant has been considering for its high-level wastes.



SLIDE 1. High-Level Waste Toxicity Vs. Time

DRAFT EPA CRITERIA (40 CFR 191, WORKING DRAFT 11/9/79)

- o DEFENSE IN DEPTH - HIGH-INTEGRITY WASTE FORMS, ENGINEERED BARRIERS, REPOSITORIES
- o ZERO RELEASE FOR 1,000 YEARS
- o RISK OF LESS THAN 200 HEALTH EFFECTS IN 10,000 YEARS
- o RISKS ONLY FOR UNLIKELY EVENTS (LESS THAN 1 IN 100 PROBABILITY)

DRAFT NRC REGULATIONS (10 CFR 60, WORKING DRAFT 12/6/79)

- o FULLY REDUNDANT REPOSITORY AND WASTE PACKAGE
- o ZERO RELEASE FROM WASTE PACKAGE FOR 1,000 YEARS
- o 10^{-5} - 10^{-7} PER YEAR RELEASE FROM WASTE PACKAGE 1,000 - 10,000 YEARS

SLIDE 2. Draft Regulatory Requirements for High-Level Waste Disposal

SOURCE	TYPE	1980 QUANTITIES 10^6 GAL	TANKS	MEGACURIES	1990 Q MEGA
HANFORD PLANT	ALKALINE	50	156	190	2
	Cs/Sr SOURCES	-	-	350	4
IDAHO CHEMICAL PROCESSING PLANT	ACID LIQUID	2.5	9	13	
	CALCINE	0.45	(3)	61	1
SAVANNAH RIVER PLANT	ALKALINE	23	33	570	6
NUCLEAR FUEL SERVICES	ALKALINE	0.6	1	64	
	ACID	0.01	1	2.1	
SPENT LWR FUEL	NOT PROCESSED	-	-	1,900	14,0

SLIDE 3. U.S. High-Level Nuclear Wastes

- 68 MILLION GALLONS GENERATED FROM OPERATION OF TWO RADIOCHEMICAL SEPARATIONS PLANTS
  - CURRENT RATE IS 1.5 MILLION GALLONS PER YEAR
- VOLUME REDUCED TO 23 MILLION GALLONS
  - STORED IN 33 TANKS

SLIDE 4. Volume of Waste at Savannah River

PRINCIPAL RADIONUCLIDES ARE STRONTIUM-90, CESIUM-137 AND PLUTONIUM-239

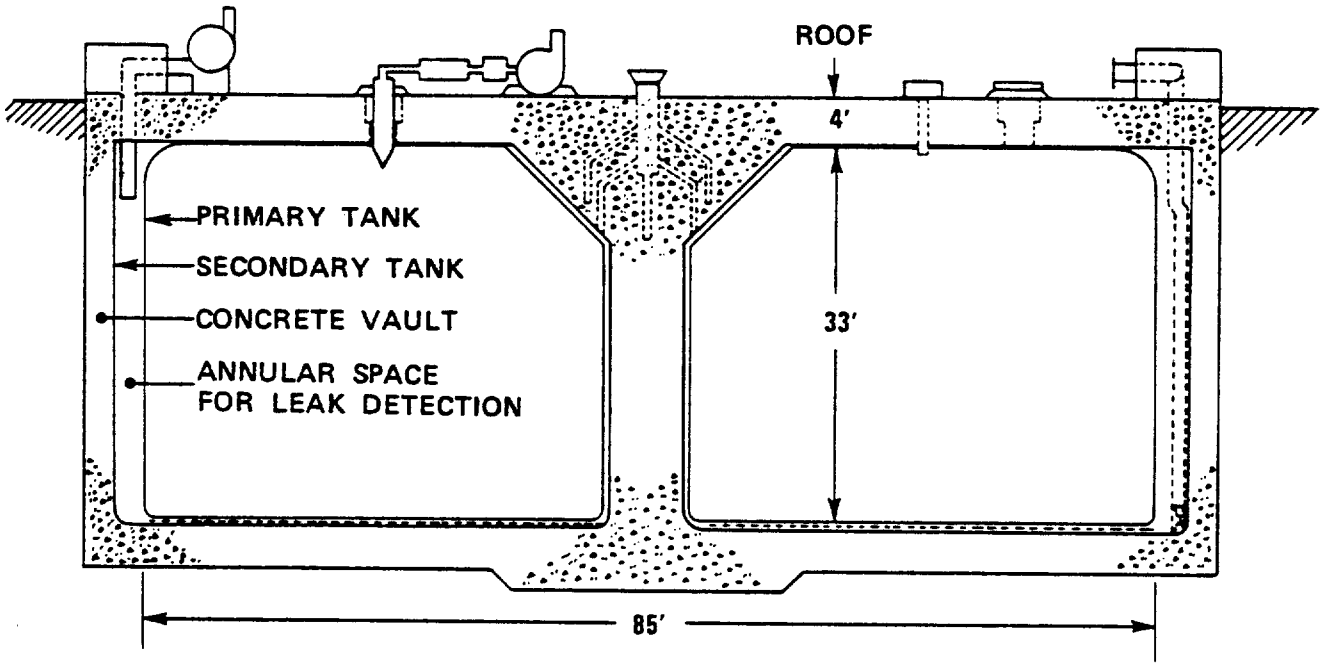
- **10% AS SLUDGE (^{90}Sr, ^{239}Pu)**

  IRON
  MANGANESE
  ALUMINUM

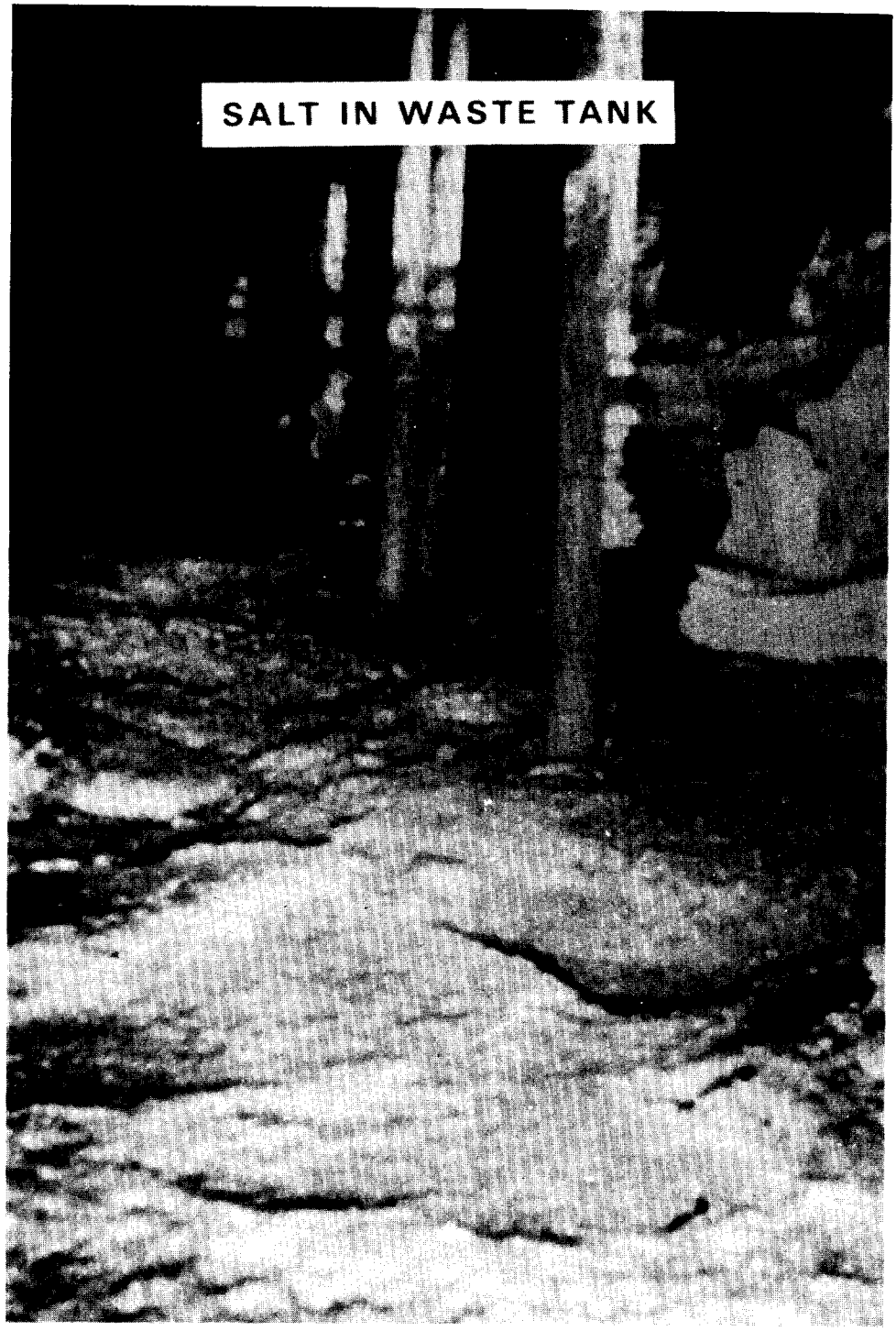
- **REMAINDER AS LIQUID AND SALT CAKE (^{137}Cs)**

  SODIUM NITRATE
  SODIUM ALUMINATE
  SODIUM HYDROXIDE

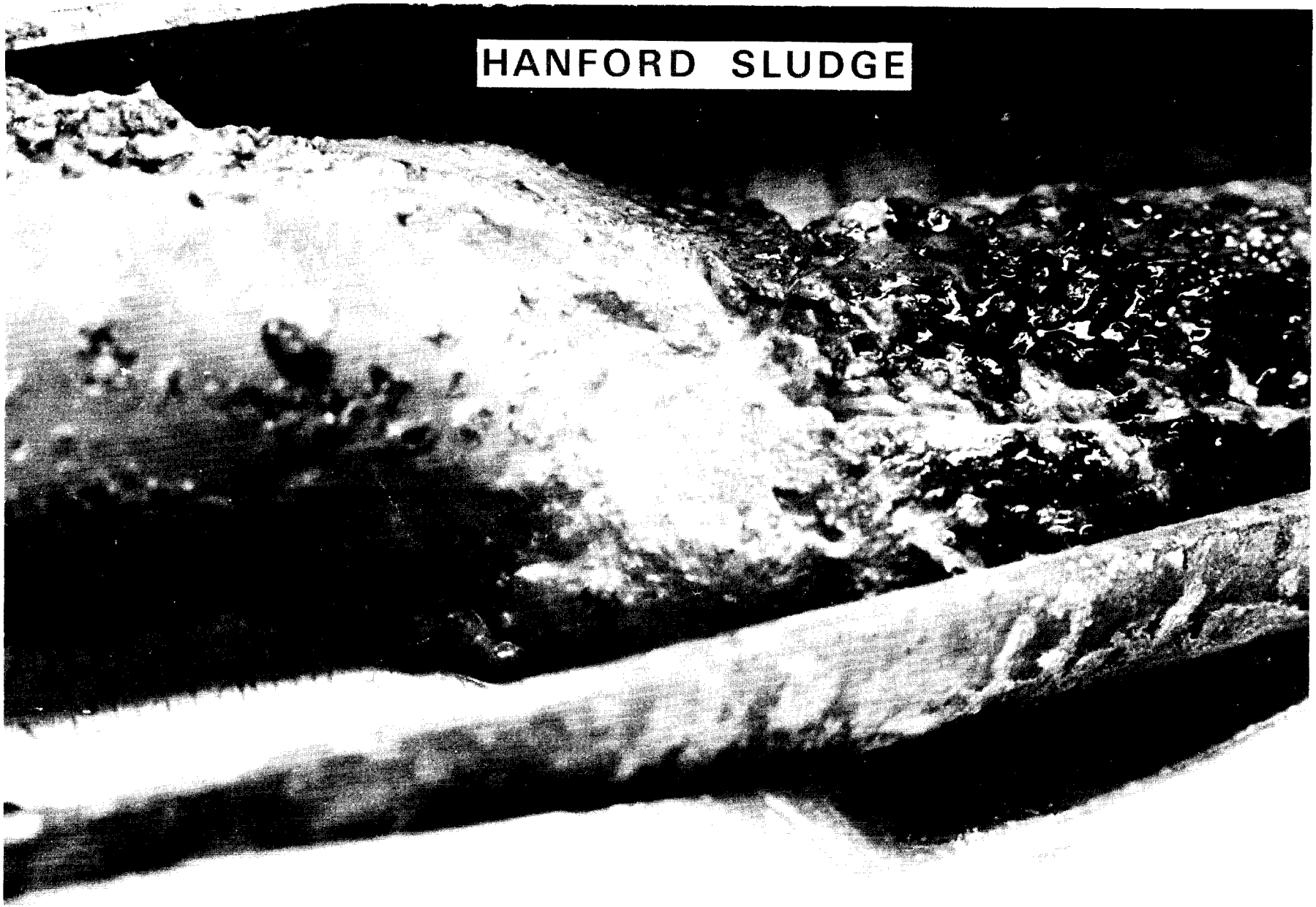
SLIDE 5. Characteristics of Liquid Wastes at Savannah River



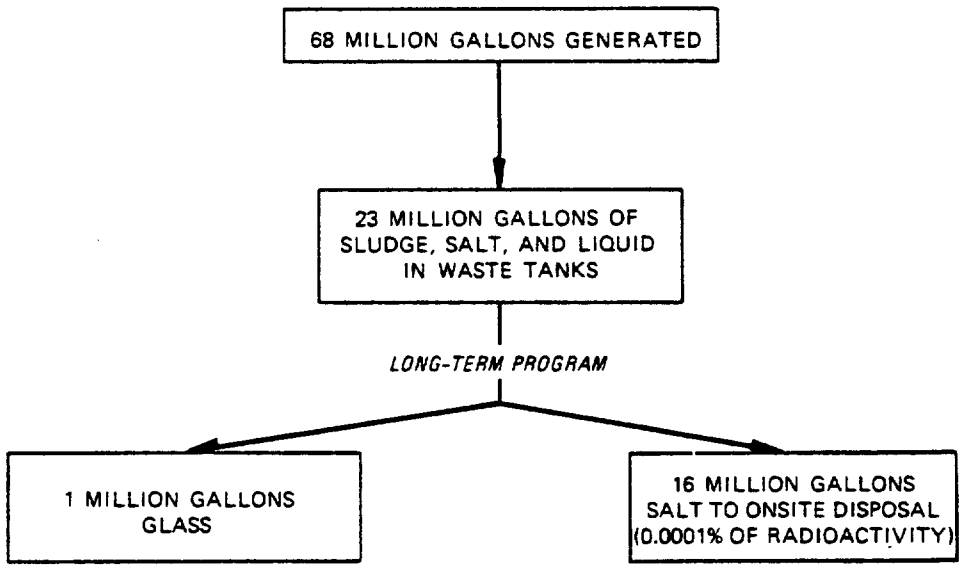
SLIDE 6. New Waste Storage Tank
(1,300,000-Gallon Stress-Relieved Tank)



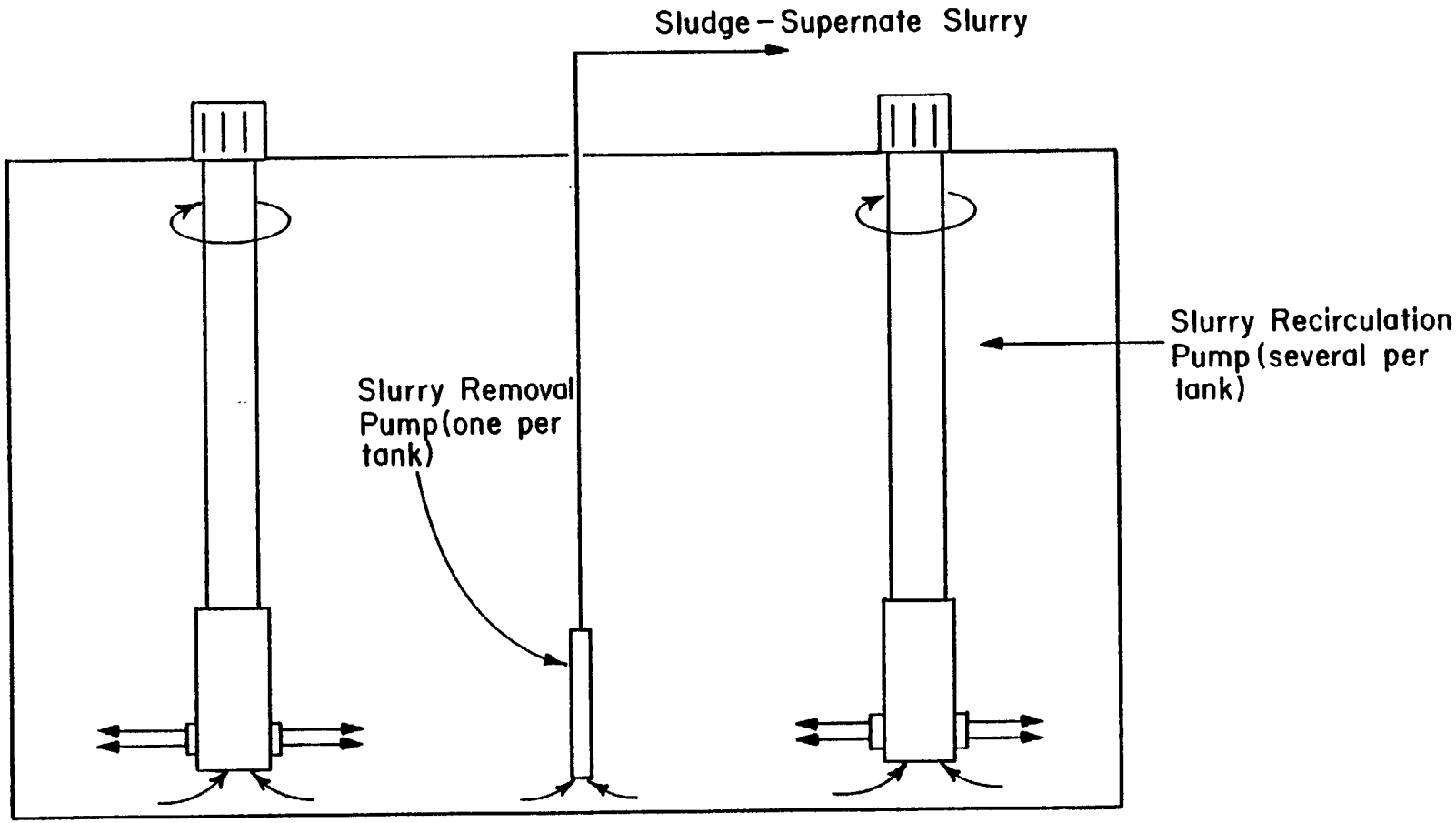
SLIDE 7. Salt in Waste Tank



SLIDE 8. Hanford Sludge

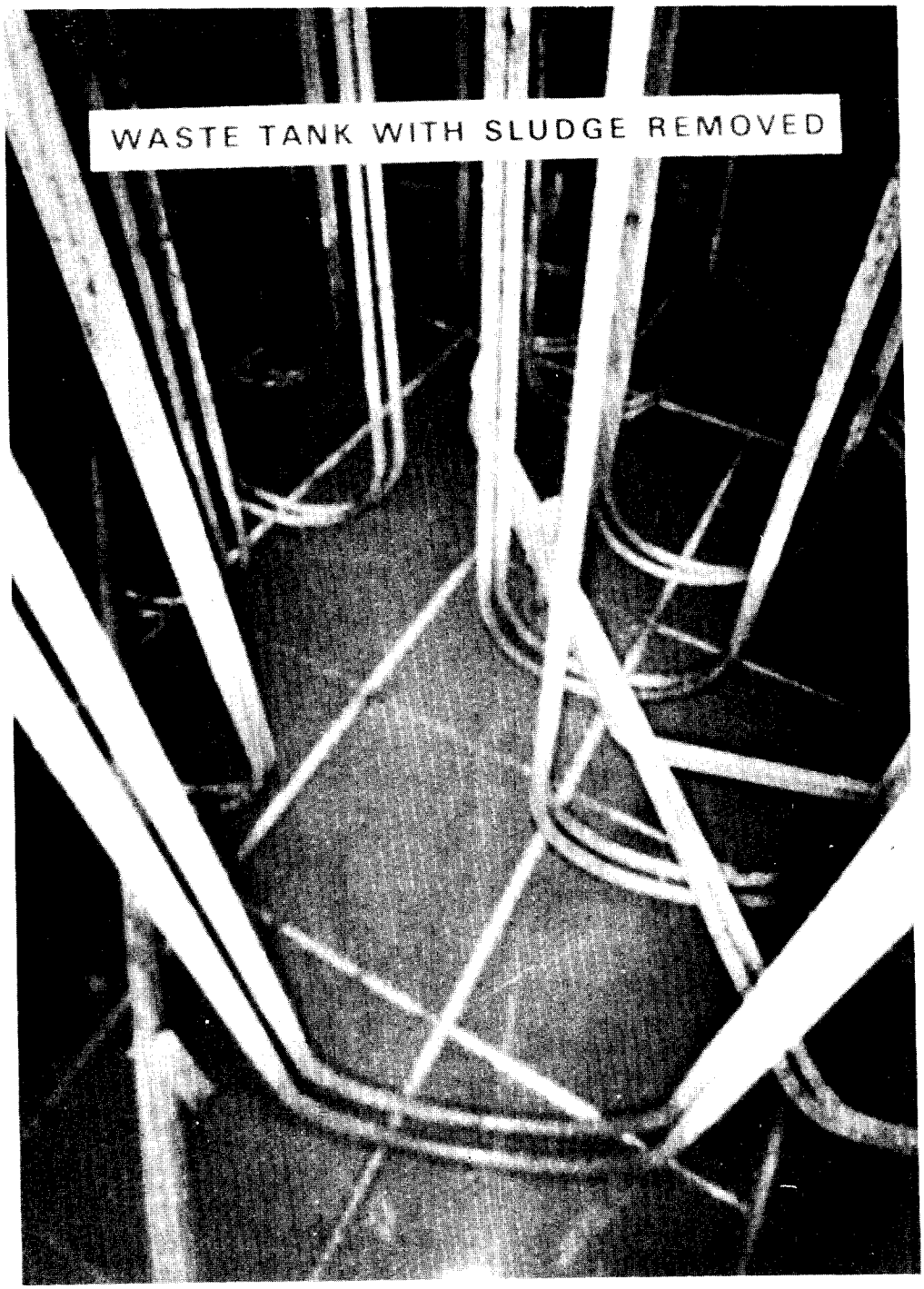


SLIDE 9. Disposition of Current Waste Inventory

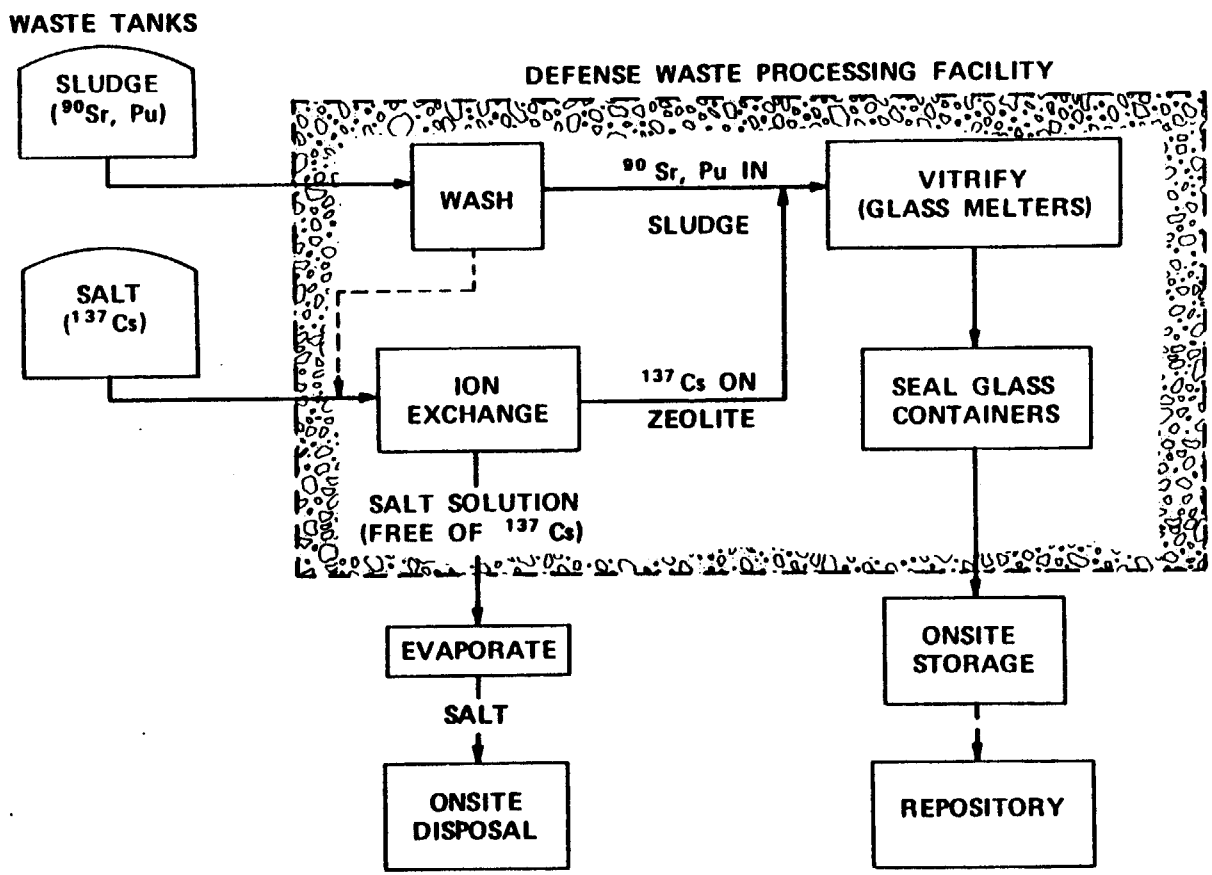


SLIDE 10. Sludge Removal

SLIDE 11. Sludge in Waste Tank

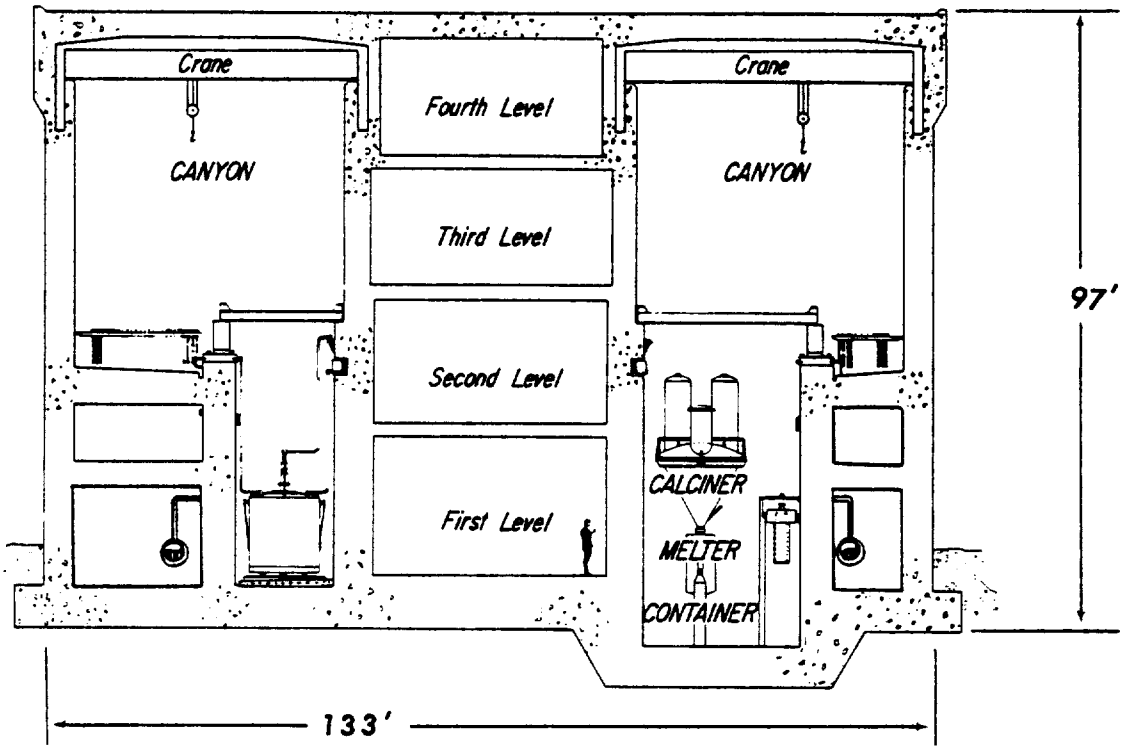


SLIDE 12. Waste Tank with Sludge Removed

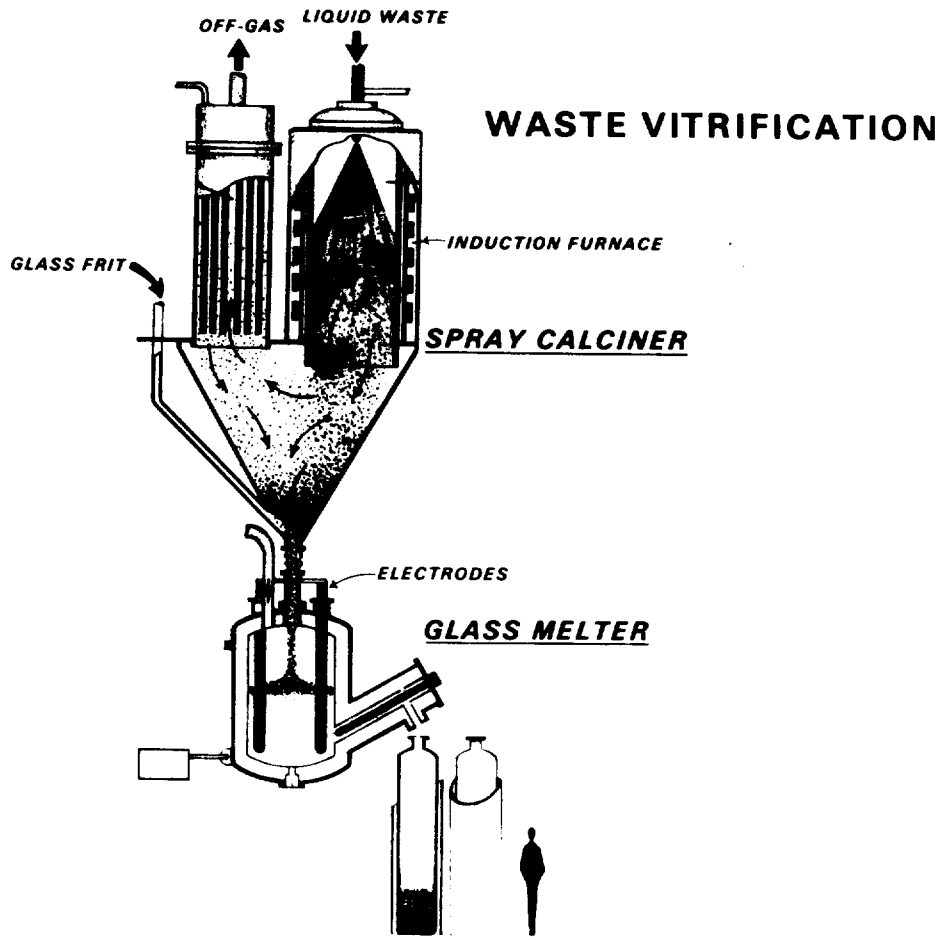


SLIDE 13. Reference Process for SRP Waste

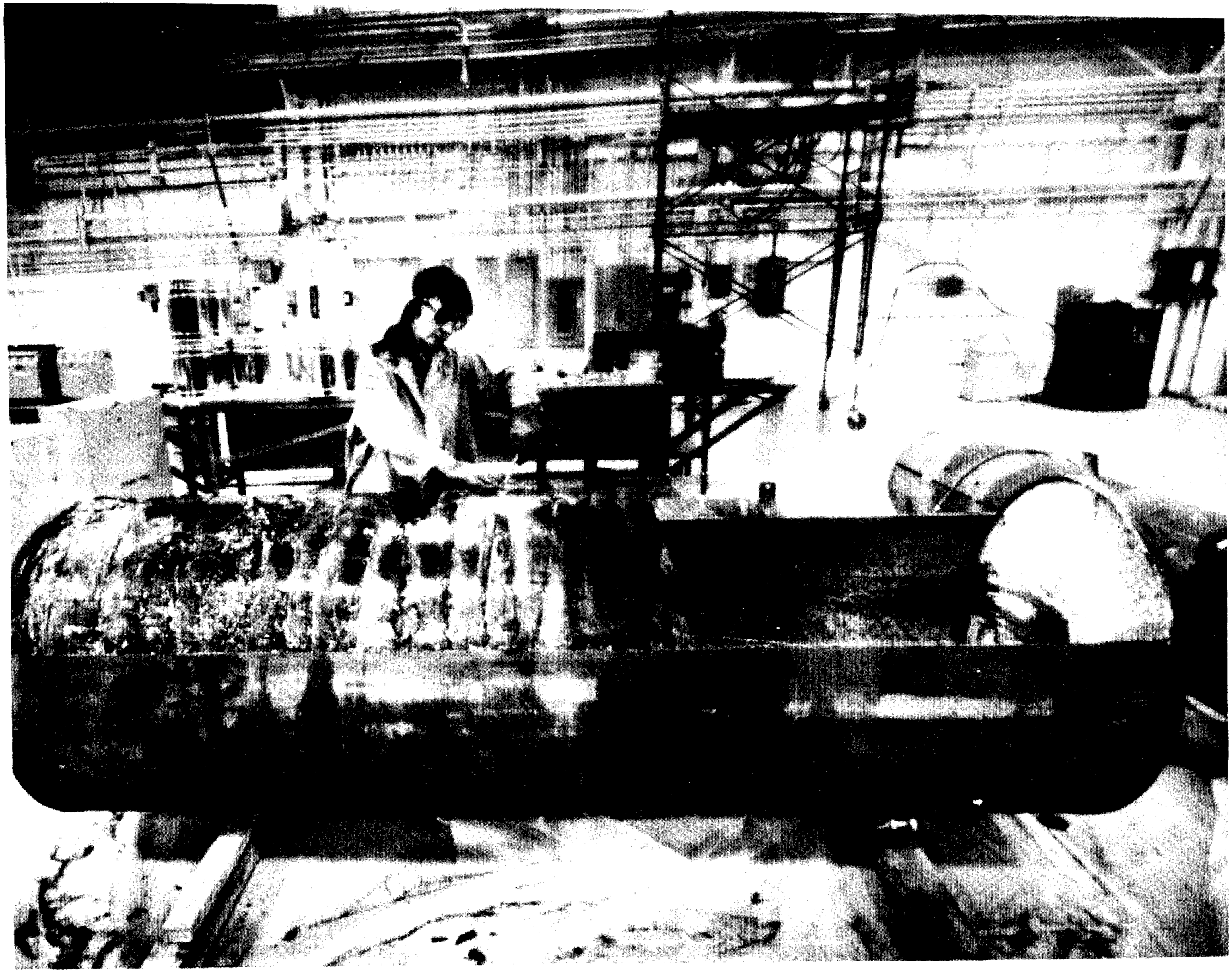
SLIDE 14. Defense Waste Processing Facility



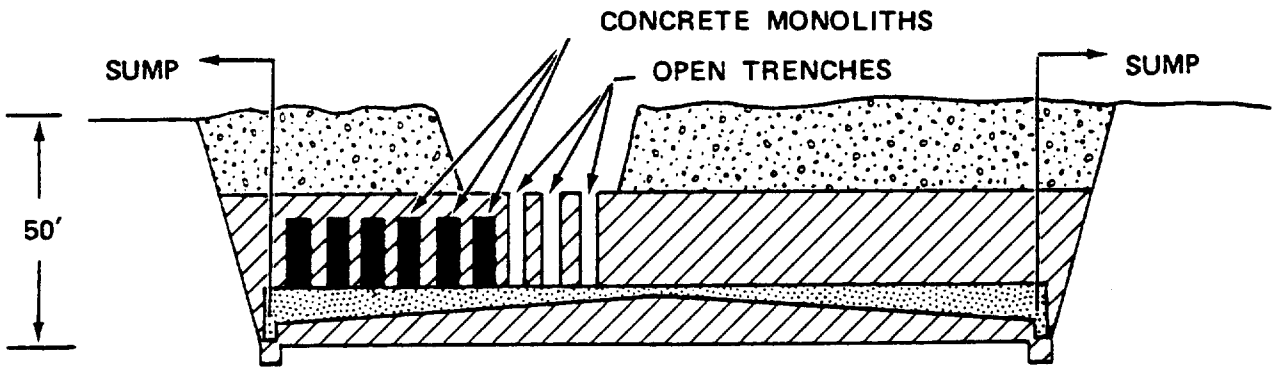
SLIDE 15. Process Building Section - Defense Waste Processing Facility



SLIDE 16. Calciner-Melter



SLIDE 17. Nonradioactive Full-Scale Canister of Glass

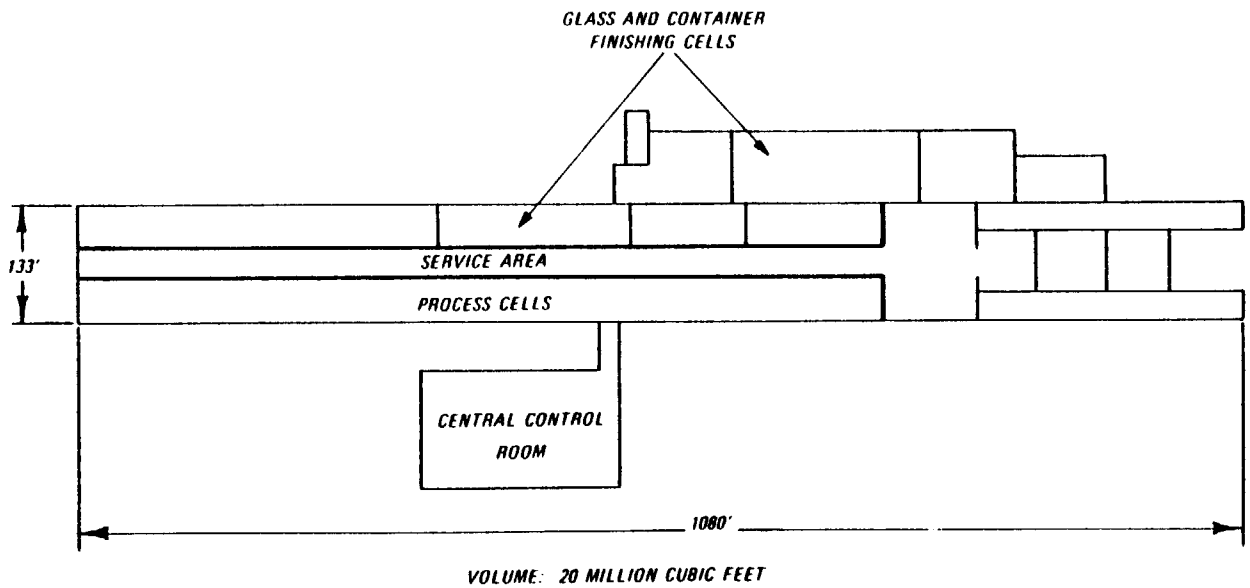


SLIDE 18. Salt Disposal System

	$ BILLIONS		
	JULY 1978	OCTOBER 1979	JULY 1980
LAST QUARTER 1979 DOLLARS	1.9	1.5	?
ESCALATED DOLLARS	2.8	2.3	?

SLIDE 19. Cost Estimates, Defense Waste Processing Facility

SLIDE 20. Comparative Chemical Durabilities



SLIDE 21. Process Building Plan - Defense Waste Processing Facility

DEFINITION AND CHARACTERIZATION OF CANDIDATE FORMS — FY 1979-1981

- EXAMINE ALL LIKELY FORMS
- SELECT 3-4 FORMS FOR LARGE-SCALE DEVELOPMENT

INTENSIVE DEVELOPMENT AND EVALUATION OF 3-4 FORMS — FY 1982-1983

- RADIOACTIVE, LARGE-SCALE TESTING
- SELECT CANDIDATE DWPF FORM(S) AT END OF FY 1983

LARGE SCALE EQUIPMENT AND PROCESS DEVELOPMENT OF DWPF FORM — FY 1984-1986

- TIME TO ALLOW FY 1989 START UP FOR DWPF

SECOND GENERATION FORM DEVELOPMENT — FY 1984-

- FOR RHO, ICPP, NFS, POSSIBLE COMMERCIAL REPROCESSING WASTE
- FOR POSSIBLE SECOND GENERATION DWPF PROCESSING

SLIDE 22. Terminal Forms Development Schedule

FORM	LEACHABILITY - MATCH TO REPOSITORY CONDITIONS
	DISPERSABILITY AND IMPACT RESISTANCE - TRANSPORT AND STORAGE
	VOLATILITY - MATCH TO TRANSPORTATION ACCIDENTS
	STABILITY - AGING RADIATION TRANSMUTATION
	SENSITIVITY TO WASTE LOADING AND COMPOSITION
	WASTE VOLUME LOADING
	DEVELOPMENT STATUS
PROCESS	PROCESS SAFETY - REMOTABILITY
	PROCESS SENSITIVITY - QA REQUIREMENTS
	FEED PREPARATION REQUIRED
	PROCESS WASTE GENERATION - OPERATING AND DECOMMISSIONING
	CAPITAL AND OPERATING COSTS
	APPLICABLE LARGE SCALE EXPERIENCE

SLIDE 23. High-Level Waste Immobilization Forms Selection Factors

IN-PLACE SOLIDIFICATION FORMS

CONCRETE, RICH CLAY, POLYMERS

CONCRETE FORMS

NORMAL CONCRETE, POLYMER CONCRETE, HOT PRESSED CONCRETE

CALCINE FORMS

DIRECT CALCINES, PELLETIZED CALCINES

GLASS FORMS

BOROSILICATE GLASS, HIGH SILICA GLASS, PHOSPHATE GLASS

CERAMIC FORMS

SUPERCALCINES, SYNROC, MINERAL ION EXCHANGERS, OTHERS

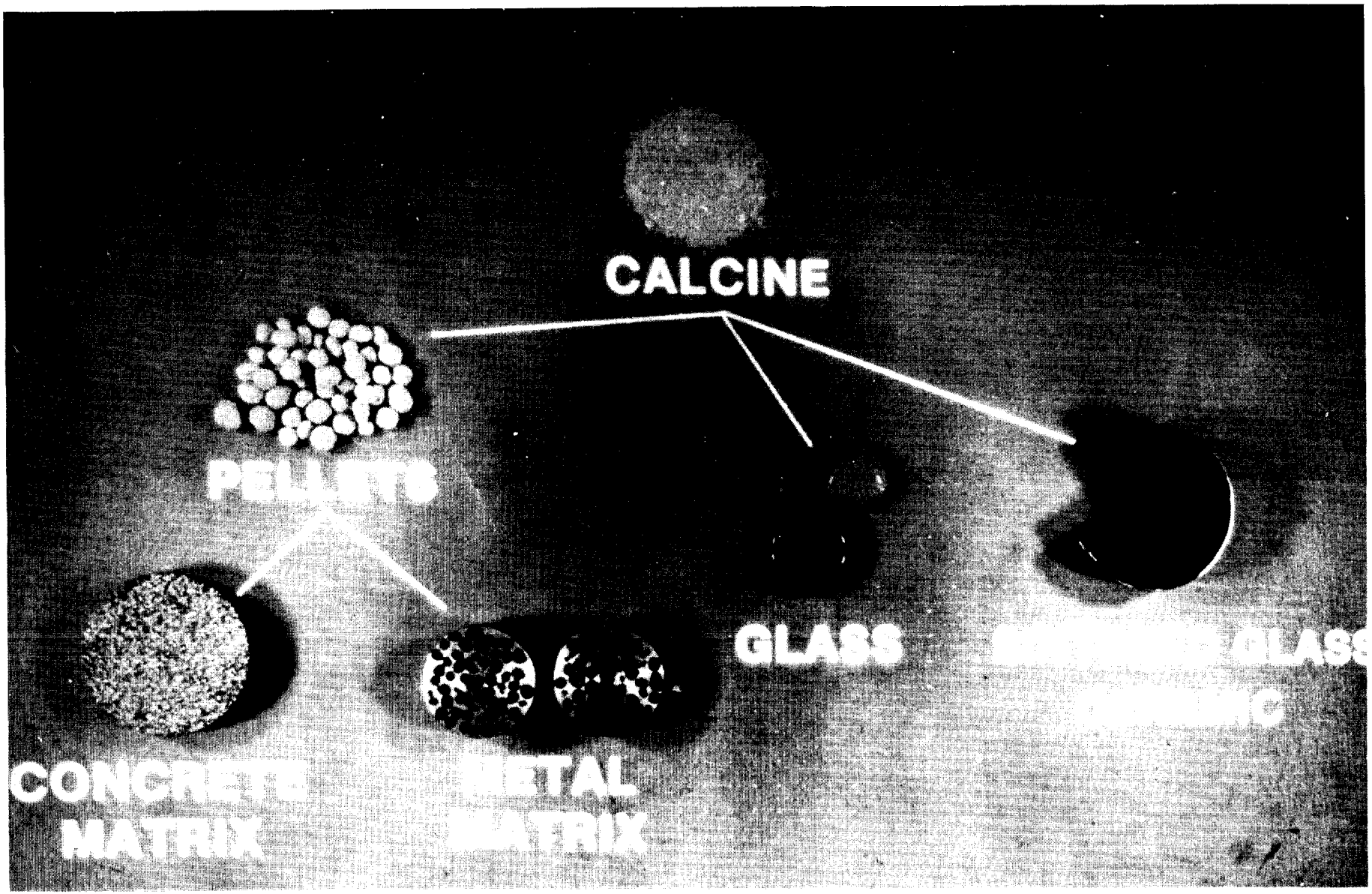
MATRIX FORMS

METAL MATRIX WITH GLASS OR CERAMIC MARBLES
CERMETS
MULTIBARRIER AND COATED PARTICLE FORMS

SLIDE 24. Primary High-Level Waste Forms

FORM	DEVELOPER
IN-PLACE SOLIDIFICATION	ROCKWELL HANFORD LABORATORY
CONCRETE	OAK RIDGE NATIONAL LABORATORY PENN STATE UNIVERSITY - ROCKWELL INTERNATIONAL SAVANNAH RIVER LABORATORY
CALCINE	IDAHO EXXON LABORATORY
GLASS	SAVANNAH RIVER LABORATORY BATTELLE PACIFIC NORTHWEST LABORATORY ROCKWELL HANFORD LABORATORY IDAHO EXXON LABORATORY CATHOLIC UNIVERSITY WESTINGHOUSE CORPORATION UNIVERSITY OF FLORIDA NASA - UNIVERSITY OF DELAWARE
CERAMIC	ROCKWELL HANFORD LABORATORY BATTELLE PACIFIC NORTHWEST LABORATORY PENN STATE UNIVERSITY - ROCKWELL INTERNATIONAL UNIVERSITY OF CALIFORNIA - LIVERMORE LABORATORY ARGONNE NATIONAL LABORATORY NC STATE UNIVERSITY SANDIA LABORATORY OAK RIDGE NATIONAL LABORATORY
MATRIX	ARGONNE NATIONAL LABORATORY BATTELLE PACIFIC NORTHWEST LABORATORY OAK RIDGE NATIONAL LABORATORY

SLIDE 25. Waste Form Developers



SLIDE 26. Final High-Level Waste Alternatives