Nuclear Waste Management Status and Recent Accomplishments

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EPRI PERSPECTIVE

PROJECT DESCRIPTION

The status and progress of nuclear waste disposal programs is often cited as a major impediment to civilian nuclear power generation. While informed technical opinion has emphasized that wastes from nuclear fission can safely be disposed of in geologic media, regulatory and institutional factors continue to delay a large scale demonstration in the U.S.

Against the background of delay and policy review which is occurring, this document is intended to emphasize the worldwide technical programmatic progress that has occurred in the past two years to effectively implement nuclear waste disposal systems. This work is an extension of a previous EPRI project, Status of Commercial Nuclear High-Level Waste Disposal, EPRI NP-44-SR, September 1976, which described the engineering approach to waste disposal in the context of the more ready-to-operate waste disposal unit processes. The processes described are not necessarily optimum, but represent an adequately safe and effective first generation waste disposal system.

PROJECT OBJECTIVES

The objective of this project is to highlight significant technical and programmatic accomplishments in implementing waste disposal that have occurred over the past two and one half years. The emphasis is placed on high-level waste disposal and spent fuel disposal, with only brief coverage of other wastes. Similarly, the emphasis is upon progress on process equipment and systems that are more practical or advanced in terms of readiness for demonstration.

There is much less emphasis on second generation systems, which may have some eventual merit, because of the belief that the U.S. program objective should

SUMMARY AND CONCLUSIONS

be expeditious implementation of an adequately safe first generation technology, and not selection of an optimum process technology or optimum geologic media which might result in an additional period of programmatic delay.

Significant technical progress has been made both in the U.S. and throughout the world. The most significant accomplishments are perhaps the successful operation of the waste vitrification plant at Marcoule, France, with production of 4.5 tons of waste glass as of August 1978, and the publication of an integrated system design study together with an integrated system assessment in Sweden, under the Nuclear Fuel Safety (KBS) project. In the U.S., significant (but perhaps less glamorous) progress has occurred in demonstrating full-scale prototypes and in performing studies to justify waste disposal technology and regulatory and siting criteria.

It appears there is an adequate and growing technical base for the necessary decisions and action to implement a timely waste disposal program in the U.S.

R. F. Williams
Project Manager

ABSTRACT

The status and technical progress of nuclear waste disposal is reviewed with emphasis on technical and programmatic progress in High Level Nuclear Waste disposal technology during the 1976 to 1978 time period. Process steps in the waste solidification and geologic disposal system are described emphasizing processes and systems that are more advanced in terms of readiness for full-scale U.S. demonstration. Worldwide technical accomplishments in support of the reference U.S. waste vitrification and geologic disposal approach are highlighted.

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ILLUSTRATIONS

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ACRONYMS/ABBREVIATIONS

AEC	Atomic Energy Commission
AGNS	Allied-General Nuclear Fuels Services
ANL	Argonne National Laboratory
AVM	A French continuous vitrification facility at Marcoule, France
BWR	Boiling Water Reactor
CIVEX	Civilian Extraction, a system for processing spent nuclear fuel
CWMS	Commercial Waste Management Statement
CM	Continuous joule-heated ceramic melter (U.S.)
DOE	Department of Energy
DOT	Department of Transportation
EMAD	Engine Maintenance and Disassembly
EPA	Environmental Protection Agency
FINGAL	British process for vitrifying LWR waste
FIPPS	A German vitrification process for HTGR wastes at Julich
ERDA	Energy and Research Development Administration
GCHWR	Gas Cooled Heavy Water Reactor
GCR	Gas Cooled Reactor
GEIS	Generic Environmental Impact Statement
GW_e	10^9 watts of electricity = 1000 MW
GW_e-yr	1 yr production of 1000 MW
HLW	High-Level Wastes
HTGR	High Temperature Gas Reactor
HWLWR	Heavy Water Moderated - Light Water Cooled Reactor
IAEA	International Atomic Energy Agency
ICM	In-Can Melter (U.S.)
ICPP	Idaho Chemical Processing Plant
INEL	Idaho National Engineering Laboratories

INFCE	International Nuclear Fuel Cycle Evaluation
KBS	Kaern-Braensle-Saekerhet (Swedish Nuclear Fuel Project)
LGR	Light Water-Cooled Graphite Moderated Reactor
LLW	Low-Level Wastes
LMFBR	Liquid Metal Fast Breeder Reactor
LWCHW	Light Water-Cooled Heavy Water Moderated
LWR	Light Water Reactor
MTHM	Metric Ton Heavy Metal
NFS	Nuclear Fuel Services
NRC	Nuclear Regulatory Commission
NSTF	Near-Surface Test Facility
ONWI	Office of Nuclear Waste Isolation
ORNL	Oak Ridge National Laboratory
PAMELA	A German vitrification process developed at Julich, Germany
PHWR	Pressurized Heavy Water Reactor
PIVER	A French vitrification process used at Marcoule, France
PNL	Pacific Northwest Laboratory
PWR	Pressurized Water Reactor
R & D	Research and Development
SC	Spray Calciner (U.S.)
SRL	Savannah River Laboratory
SRP	Savannah River Plant
TAD	Technical Alternatives Document
THTR	Thorium Heavy Water Temperature Reactor
TRU	Transuranic Wastes
USGS	United States Geologic Survey
VERA	A German vitrification pilot plant at Karlsruke, Germany
WCF	Waste Calcination Facility
WIPP	Waste Isolation Pilot Plant
WSEP	Waste Solidification Engineering Prototype

SUMMARY

In the last several years, the event which had the greatest impact on the nuclear waste management program in the U.S. was the government decision to indefinitely defer commercial reprocessing of spent nuclear fuels. This measure became effective in April 1977.

IN 1977 SPENT FUEL DISPOSAL TECHNOLOGY ADDED

The decision to defer commercial fuel reprocessing created a need for more storage space for spent nuclear fuels until a decision on their long-term disposition can be made. It also created a need to consider how and where these fuels can be isolated from the biosphere, because they represent a risk to the environment that is at least comparable to that of the high-level waste (HLW) that results from reprocessing. (Categories of waste are defined in Appendix A.)

WASTE DISPOSAL AND HIGH-LEVEL WASTE SOLIDIFICATION CONTINUE

Deferring the reprocessing of spent fuels did not decrease the scope of the nuclear waste management program in the U.S. since the program must still consider the possibility that in the future a decision will be made to reprocess these fuels. In addition, much of the technology is required for existing wastes. Thus, the U.S. R&D program on nuclear waste management remains much the same as it was two years ago, except for the added requirement for interim storage and ultimate isolation of unreprocessed spent fuels. Emphasis on solidifying liquid HLW has shifted from the wastes expected from commercial fuel reprocessing plants to defense wastes at Savannah River, Hanford, and Idaho Falls. The technology developed could also be applicable to wastes already produced and stored at the now inactive Nuclear Fuel Services (NFS) plant at West Valley, New York. Vitrification technology previously developed for future commercial waste is directly applicable to these wastes.

SUBSTANTIAL FUEL VALUE WILL BE LOST IF FUELS ARE NEVER REPROCESSED

The uranium and plutonium present in irradiated nuclear fuels have substantial fuel value that will be lost if these fuels are never reprocessed. Also, recovery and reuse of plutonium is necessary in a breeder reactor program. Reprocessing schemes have been proposed that may resolve the weapons-proliferation issue. For example, CIVEX processes would recover uranium and plutonium for reuse; however, the plutonium would not be in a weapons-usable form.

The planned basic, or reference, nuclear waste management system in the U.S. involves geologic isolation of all commercial HLW and all low-level wastes (LLW) that contain significant amounts of long-lived transuranium elements (TRU wastes). The present limit for non-TRU wastes is 10 nCi of α radiation/g of waste from TRU elements. This limit is currently being reviewed to see if it is unnecessarily low. Other LLW will be buried in near-surface facilities. Because they will decay to innocuous levels in a few tens or hundreds of years, they are considered safe if the near-surface sites are properly monitored and maintained.

GEOLOGIC ISOLATION UTILIZES MULTIBARRIER TECHNOLOGY

Guaranteeing the integrity of a geologic formation for thousands or millions of years is recognized as very difficult. For this reason, isolation of wastes in geologic formations will probably rely on multiple barriers. To alleviate concern that some future event may expose the wastes to fluids (principally water) that could carry the wastes to the biosphere, several barriers can be imposed to prevent or greatly retard dispersal of the radionuclides. Possible barriers include relatively insoluble waste forms, corrosion-resistant containers and over-packs, addition of materials which adsorb and retain radioactive elements, and long paths that the fluid must traverse to reach the biosphere. Some studies indicate that a long path through the natural geologic barrier is adequate by itself. The repository system can also be designed to operate at lower temperatures so that water is of much less concern. Temperatures can be lowered by further aging the waste or spent fuel, using lower

REPOSITORIES CAN BE DESIGNED TO OPERATE AT LOW WASTE TEMPERATURES IF REQUIRED

concentrations of waste, or improving the repository design to increase heat transfer in the region adjacent to the waste canister.

Defense activities produce all types of nuclear wastes. Detailed studies are in progress on how to best fit these defense wastes into the overall U.S. waste management system.

RECENT PROGRESS

Progress toward a waste management system that will ultimately require no surveillance can be divided into two categories: 1) evaluation of specific technological advancements, and 2) evaluation of the costs, risks, and benefits of the many options for the nuclear waste management system.

Category 1 - Recent Technological Advances

FRANCE BEGAN OPERATION OF VITRIFICATION FACILITY IN 1978

In France a production HLW vitrification facility began "hot" operation in June 1978. This facility, known as the AVM, will routinely convert the high-level wastes stored at Marcoule, France, to borosilicate glasses. The AVM process consists of a rotary calciner and a continuous metallic melter. The stainless steel canisters of glass are stored in air-cooled pits located adjacent to the processing plant. A similar waste processing plant (AVH) is being designed for operation at the La Hague fuel reprocessing plant in France.

U.S. WILL PERFORM FIRST LARGE-SCALE VITRIFICATION OF LWR HIGH-LEVEL WASTE

In March 1979, the U.S. will perform the first large-scale vitrification of commercial high-level wastes. The HLW from high burnup Light Water Reactor (LWR) spent fuel will be converted to borosilicate glass using a spray calciner/in-can melting process. The canisters of glass will be 20 cm dia by 2.4 m (8 in. x 8 ft) long. This technology is highly developed and was operated radioactively at Hanford prior to 1970 using defense HLW materials as a source of waste. The 33 cansiters of solidified HLW from the Waste Solidification Engineering Prototype (WSEP) are still stored at Hanford. The borosilicate glass canisters are still being evaluated; however, after eight years storage, there has been no change observed in the glass properties.

BOROSILICATE GLASS HAS PROVEN DESIRABLE WASTE-FORM FEATURES

Borosilicate glass has been chosen internationally as the reference solidified waste form for HLW, and vitrification processes have been developed for this waste form. The glass has a low dissolution rate in water and other environmental solutions that may occur in a properly designed repository. It exhibits good physical integrity. It can accommodate a wide variety of waste constituents, as well as frequent changes in composition. It is thereby fully adaptable to the large numbers of waste compositions requiring treatment. The system is designed so that only a small fraction of the glass will be susceptible to thermal devitrification. Even if devitrification occurs, the only significant result is less than a tenfold increase in the glass dissolution rate. All tests to date show that the glass is extremely stable to all types and levels of radioactivity.

WASTE VITRIFICATION TECHNOLOGY BEING APPLIED IN DEFENSE WASTE PROGRAM

In the U.S. the vitrification technology previously developed for commercial HLW is being evaluated for use in defense waste programs. The most active developmental program is aimed toward the possible vitrification of HLW stored at the Savannah River Plant (SRP). If a decision is made to remove the wastes from the storage tanks, a vitrification facility could be in operation by 1988. The vitrification process would be spray calcination coupled to either an in-can melter or a continuous ceramic melter.

CONTINUOUS JOULE-HEATED CERAMIC MELTER DEVELOPED FOR VITRIFICATION

The U.S. waste vitrification studies at the Pacific Northwest Laboratory (PNL) have emphasized in-can melting of waste glass with joule-heated ceramic melting as a possible backup. Recent engineering-scale studies (nonradioactive) with continuous ceramic melters indicate they may offer advantages over in-can melters. Major advantages are: 1) capacity can be very high, 2) waste canisters are not subjected to high temperatures, and 3) a variety of glass forms (e.g., marbles, monoliths, etc.) can be produced in the same equipment.

Alternative waste forms and processes are being developed for the immobilization of radioactive waste. These forms may provide improved containment

of the waste either by inherent improved properties, such as stable low-leaching crystals, or by providing multiple barriers of containment. Alternatives being investigated include supercalcines by Pennsylvania State University as part of the PNL multibarrier waste form, metal matrices at Argonne National Laboratory (ANL), and cermet at Oak Ridge National Laboratory (ORNL). Considerable development is required before it can be determined whether these alternatives offer a substantial improvement over glass.

ALTERNATIVE WASTE FORMS MAY OFFER SECOND GENERATION IMPROVEMENTS OVER GLASS

Recently the Sandia Laboratories conducted full-scale crash tests on spent fuel shipping casks. Truck- and railcar-mounted casks were crashed into virtually immovable concrete targets at speeds up to 136 km/h (84 mi/h) with only superficial damage to the cask and no rupture of contained fuel elements. Similar casks would be used for transporting canisters of HLW.

SAFE TRANSPORTATION OF NUCLEAR WASTES SEEMS ASSURED

The general concept that nuclear wastes can be safely isolated in geologic formations is receiving widespread support among the nations of the world. Eighteen countries have programs to evaluate various formations for potential disposal sites. The Federal Republic of Germany is already operating the Asse salt mine for the disposal of nonhigh-level wastes. Sweden is conducting field tests in the Stripa mine to support design of a disposal site. Belgium, France, Italy, Spain, Sweden, and the United States plan pilot repositories in the 1980s. Commercial repositories by the year 2000 are planned by Canada, the United Kingdom, the Federal Republic of Germany and the United States.

SEVERAL NATIONS HAVE PILOT GEOLOGIC REPOSITORIES SCHEDULED FOR OPERATION IN LATE 1980s

A Waste Isolation Pilot Plant (WIPP), currently expected to be operational in 1985, is in the planning stages. It will be used primarily as a geologic disposal location for TRU wastes from the defense program and as a facility in which to perform R&D with other waste materials in salt. A site has been proposed near Carlsbad, New Mexico. It is now undergoing detailed geologic investigation and a conceptual facility design has been prepared. The DOE task force on waste management recommends that,

U.S. WASTE ISOLATION PILOT PLANT (WIPP) PLANNED FOR OPERATION IN 1985

in addition to being used for R&D studies on isolating defense HLW, the facility could be used for a demonstration of disposal of a limited number of commercial fuel assemblies.

SEVERAL U.S. GEOLOGIC SITES BEING EVALUATED FOR WASTE DISPOSAL

The Office of Nuclear Waste Isolation, operated by Battelle Memorial Institute for the Department of Energy (DOE), has an extensive program to identify and develop candidate geologic formations and locations for nuclear waste disposal. Salt formations have received major emphasis to date, but other formations (granite, shale) are also being evaluated. Rockwell Hanford Operations has a DOE program to evaluate disposal of wastes in the basalt underlying the Hanford area. Similarly, a program to evaluate the potential for waste disposal at the DOE Nevada Test Site is being conducted by Sandia Laboratories for DOE. A major objective of the study is to determine if waste disposal is compatible with the predicted effects of continued weapons testing.

GERMANY TENDING TOWARD NUCLEAR PARK CONCEPT

The Federal Republic of Germany plans to concentrate reprocessing, recycling, and waste disposal at Gorleben in Lower Saxony. A 1400-t/y reprocessing plant is planned for operation in the late 1980s. The area lies over salt beds. Solidification of high-level waste and intermediate storage in a retrievable mode is planned to allow time to develop and demonstrate final disposal in geologic formations.

FAVORABLE RESULTS FOR SAFE 20- TO 100-y WATER BASIN STORAGE OF SPENT FUELS

Deferral of commercial spent fuel reprocessing in the U.S. requires that spent fuels be held in interim storage longer than in a closed fuel cycle (probably >20 y). Water basin storage is the only proven and extensively used spent-fuel-storage technique available. Dry storage of spent fuel is being considered at this time. An assessment of past water basin storage experience concludes that no significant problems have been encountered during storage of both Zircaloy and stainless-steel-clad fuels for periods ranging up to 18 y, and that prospects are favorable for safe water basin storage of spent fuels for longer periods. Long-term storage

will require more storage capacity in the reactor storage basins, as well as in away-from-reactor storage facilities.

SPENT FUEL PACKAGING PROGRAM UNDERWAY

Temporary storage or permanent isolation of spent fuels in geologic formations will require package designs appropriate to the waste form and geologic formation. This is a new need since prior packaging studies have involved only wastes. A DOE program to develop spent fuel packaging has been formulated and is already in progress. In addition, several alternative high-integrity canister concepts have been proposed in Sweden and are being studied.

Category 2 - Costs, Risks and Benefits

The second category toward implementing the overall nuclear waste management program is related to costs, risks and benefits. Key events in this category are highlighted below.

TECHNICAL ALTERNATIVES DOCUMENT ISSUED

A study has been completed that characterizes and classifies the various technologies for managing nuclear wastes produced in reprocessing fuels from LWRs. This report, ERDA-76-43, is the Technical Alternatives Document (TAD) and is serving as a reference document for the preparation of environmental impact statements and other documents required for licensing the construction and operation of waste management facilities.

DOE REVIEW OF NUCLEAR WASTE MANAGEMENT COMPLETED

In March 1978, a DOE task force under the direction of J. M. Deutch completed a review of all nuclear waste management programs in the DOE. That report (DOE/ER-004/D) endorses geologic disposal of both high-level wastes and spent fuels and recommends government ownership and operation of all nuclear waste disposal sites.

INTERAGENCY REVIEW GROUP ESTABLISHED TO PROVIDE RECOMMENDATIONS TO PRESIDENT

The DOE Review of Nuclear Waste Management was studied by an Administration-established interagency review group. A report on the review (TID 28818) is due to the President in December, 1978, with recommendations to be used in decision-making and implementation. The report will be reviewed by the public and government agencies in early 1979.

A Commercial Waste Management Statement (DOE-1559) based on guidelines from the report by

COMMERCIAL WASTE MANAGEMENT STATEMENT PREPARED

the DOE Task Force for Review of Nuclear Waste Management was completed in September 1978. This is a modification of the generic environmental impact statement on the management of commercially-generated nuclear wastes. Its release to the public is anticipated early in 1979. It will be subject to the normal review and comment process.

TECHNICAL ALTERNATIVES DOCUMENT PREPARED FOR DOE SITES AND NFS PLANT

Technical alternative documents have been prepared defining potential paths for future management of the HLW stored at SRP, Hanford, Idaho National Engineering Laboratories (INEL), and at the NFS plant. Environmental impact statements based on these technologies are in preparation for the government-owned sites and will serve to guide programmatic decisions on the long-term management of these wastes.

INTERNATIONAL FUEL CYCLE EVALUATION PROGRAM UNDERWAY

A worldwide International Fuel Cycle Evaluation Program (INFCE) was established. The purpose of this program is to thoroughly evaluate the major alternative fuel cycles to identify how best to minimize the risks of weapons proliferation and, at the same time, to assure that civilian nuclear power remains a viable energy source. While the results of the INFCE will be advisory only, the outcome could well influence the nature of the future fuel cycle and, consequently, the composition of the high-level wastes which could be produced.

SWEDISH (KBS) STUDY INDICATES GEOLOGIC DISPOSAL SAFE

In Sweden, a total system review and analysis was completed on the handling of spent fuel and the final disposal of vitrified HLW. An extensive safety analysis was made of a system involving 10-y storage of spent fuel before reprocessing, vitrification of the HLW to borosilicate glass, near-surface storage of the glass for 30 y, containment of the glass in a multilayer waste canister, and isolation of the canister in a granite repository with ion exchange material surrounding the canister.

All these barriers prevent the escape of the wastes to the biosphere via water. The maximum potential dose to individuals living near the repository was calculated to be a factor of nearly 40 less than the International Committee for Radiation Protection (ICRP) limit for continuous exposure of individuals.

THE EPA AND NRC TO ISSUE STANDARDS AND REGULATIONS SOON

The Environmental Protection Agency (EPA) and the Nuclear Regulatory Commission (NRC) are currently preparing regulations related to HLW. The EPA published radioactive waste criteria in late 1978. These standards or criteria are currently undergoing public review and comment. The NRC is preparing a new regulation, 10 CFR 60, that deals with disposal of high-level wastes in geologic repositories. A draft of this regulation may be available for public comment early in 1979.

RISK ASSESSMENT STUDIES IN PROGRESS

Several risk-assessment studies are in progress to define the performance of and provide the bases for criteria for geologic repositories. These studies treat the waste form/containment/repository/surrounding geology/ecosystem as a system of barriers. Studies are underway at Lawrence Livermore and Sandia Laboratories (NRC-sponsored), A. D. Little and the University of New Mexico (EPA), and the Pacific Northwest Laboratory (DOE).

TECHNOLOGY EXCHANGE MEETINGS ON SOLIDIFIED WASTE FORMS UNDERWAY

Several important technical meetings on solidified waste forms were planned in late 1978 and early 1979. The results of these meetings will be documented in proceedings to be published in 1979. These meetings include a symposium on Science Underlying Radioactive Waste Management, held in conjunction with the 1978 meeting of the Materials Research Society (November 28-December 1, 1978, Boston, MA); the NRC Conference on High-Level Radioactive Solid Waste Forms (Denver, CO, December 18-21, 1978); and the upcoming DOE/American Ceramic Society sponsored International Symposium on Ceramics in Nuclear Waste Management (to be held April 30-May 2, 1979, Cincinnati, OH).

CONCLUSIONS

TECHNICAL PROGRESS TOWARD IMPLEMENTING A WASTE MANAGEMENT STRATEGY APPEARS ADEQUATE

1. The strategy for safe management of nuclear wastes in the U.S. appears well defined. The U.S. strategy is similar to that being proposed and adopted by other countries, i.e., geologic isolation of spent fuel or immobilized HLW, geologic isolation of TRU wastes and engineered surface storage, or burial of low-level non-TRU wastes.

SYSTEMS ARE AVAILABLE FOR IMMOBILIZING RADIOACTIVE WASTES

2. The current successful operation of the French AVM vitrification facility, the prior operation of U.S. solidification processes using fully radioactive waste (WSEP), and the current program to use U.S.-developed processes for potential vitrification of defense HLW clearly show that systems are available for immobilizing radioactive wastes.

MANY COUNTRIES STUDYING GEOLOGIC FORMATIONS TO ISOLATE RADIOACTIVE WASTE

3. Many countries are actively studying geologic formations to isolate radioactive waste, and several countries plan pilot-plant repositories in the 1980s (Belgium, Canada, France, West Germany, Italy, Spain, Sweden, Great Britain and the United States). West Germany already operates the Asse salt mine as a repository for nonhigh-level wastes. The United States is studying bedded salt, domed salt and basalt, granite, and shale locations, where these media can provide safe isolation of wastes. Sweden conducted perhaps the most thorough systematic study of geologic isolation of HLW and unreprocessed spent fuels available to date. This study clearly concluded that geologic storage of HLW or spent fuel could be carried out safely.

TECHNICAL CONCENSUS THAT WASTES CAN BE SAFELY ISOLATED IN GEOLOGIC FORMATIONS

4. There appears to be a consensus among technical experts that HLW and TRU wastes can be safely isolated in geologic formations, but that extensive data on each proposed site is needed as a basis for choosing the optimum waste form, waste containment, and emplacement mode. Progress appears adequate for acquiring the needed data and for determining the risks of geologic isolation.

DEGREE OF CONSERVATISM IN REPOSITORY DESIGN IS REMAINING TECHNICAL ISSUE

5. The degree of conservatism to be used in geologic disposal appears to be one remaining techical issue on which there is not yet consensus. At one extreme, some European countries plan 40 to 50 y surface cooling before emplacement of HLW. At the other extreme, some U.S. designers contemplate waste emplacement at high heat load after only 10 y. Sweden selected a maximum 70°C canister wall temperature after emplacement; INFCE is considering 110°C. Early U.S. designs for dry repositories have been as high as 375°C. The degree to which

multiple barriers are required as part of the overall waste management system is part of this technical issue. The DOE task force stated that implementation of geologic disposal should emphasize technical conservatism.

DEVELOPMENT OF COMMERCIAL WASTE MANAGEMENT TECHNOLOGIES SHOULD BE CONTINUED

6. Progress toward developing technologies for commercial waste management should be continued, even though fuel storage and disposal is planned at this time:

- These technologies are required for wastes that already exist.
- The amount of commercial waste is growing rapidly (e.g., annual ^{90}Sr production in 1985 will exceed the total inventory in defense HLW).
- Reprocessing of spent fuel may be reinstated.

Section 1

INTRODUCTION

In 1976, EPRI published a status report on the management of commercial nuclear fuel cycle waste with emphasis on high-level waste (HLW) from spent fuel reprocessing.(1) That report described the technology that had been developed, the significance of the technology to waste management, and plans for future work. Since that time, there has been a significant change in the policy of the United States on fuel reprocessing. In April 1977, the government announced that reprocessing of spent nuclear fuels was to be indefinitely delayed. Obviously this decision has affected the plans for the management of commercial waste, even though the disposal requirements for unreprocessed spent fuel will be similar to those for HLW. In light of this policy change, this report will review recent progress made toward implementing an effective waste management program.

One of the major effects of the new policy has been the introduction of spent fuel as a longer term waste-management problem. Therefore, although the emphasis of this report will be on HLW, considerable attention will be given to the storage and potential isolation of spent fuel. The technology described by Dau and Williams (1) is briefly reviewed, but emphasis is placed on accomplishments since 1976. These accomplishments include those made internationally and those related to defense HLW from production-type reactors.

The proposed U.S. waste-management reference system for commercial wastes is shown in Figure 1-1.(2) It provides isolation of HLW and transuranic (TRU) wastes in a location away from the biosphere. This reference system assures that uncontrolled reentry of those wastes into the biosphere is very unlikely. (The origin of the

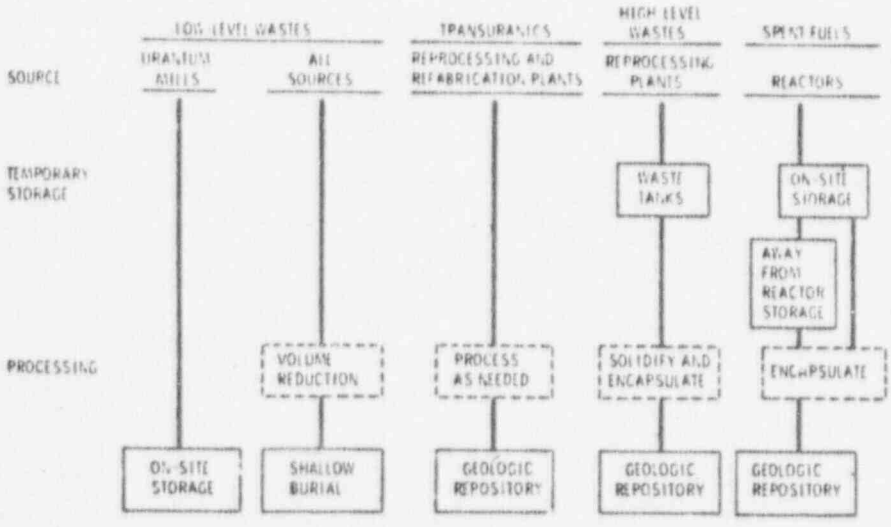

FIGURE 1-1. Proposed Reference U.S. Nuclear Waste Management System (2)

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wastes is described in Appendix A.) Because both wastes contain significant amounts of long-lived (tens of thousands of years) radioactive elements, complete isolation from the biosphere is considered necessary. The need for spent fuel isolation will be at least equivalent to that for these wastes if spent fuel is never reprocessed. Non-TRU low-level wastes, because they will decay to innocuous levels in a relatively short time (a few hundred years), are considered safe if properly stored and monitored in near-surface sites. Mining and milling wastes will be managed onsite even though they contain long-lived TRU constituents.

Previous and present defense activities have produced and will continue to produce all these waste types. Detailed studies are in progress on how to best fit these defense wastes into the overall U.S. nuclear waste management program.

Isolation of HLW and TRU waste in geologic formations depends on a multiple-barrier system to prevent reentry of radionuclides into the biosphere.[3] Figure 1-2 shows some of the barriers which may exist or be built into this multibarrier system. The inner-most barrier is a relatively insoluble waste form, such as glass, which will resist leaching of radioactive constituents if water enters the repository and contacts the waste form. Entering water can be delayed from contacting the waste form and allowing radioactive constituents to migrate into the nearby geologic formation by any of several types of engineered barriers. These barriers include the waste canister wall, secondary canisters, and materials impervious to water or materials which absorb and retain radioactive elements. Remoteness of the repository from the biosphere and the slow movement of radioactive constituents through rock and soil paths to the biosphere provide natural isolation barriers.

In this report we ask: Where do we stand and where are we going toward implementing this waste management system? It must be recognized that safe management of nuclear wastes is a complicated problem involving many technical disciplines. It is also a problem about which the general public is highly concerned.

The authors believe that the conclusions drawn in this report represent a fair assessment of the attitude of the technical community. A general concensus has not yet been reached, however, on what would constitute the most satisfactory waste management system. Similarly, divergent views on the adequacy of our progress are evident from comments in the public and technical media. The authors hope that the material presented here will provide a basis from which the reader can draw his own conclusions.

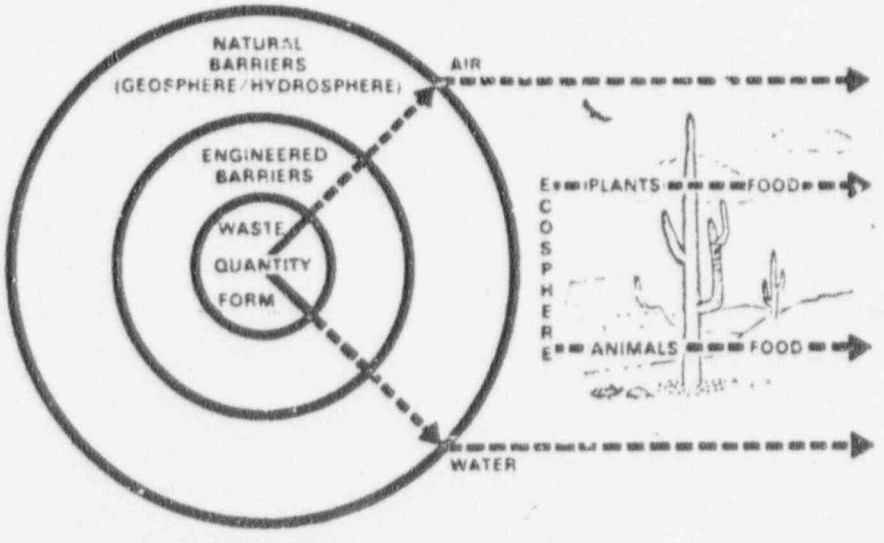

FIGURE 1-2. Multibarrier Nuclear Waste Isolation System

Section 2

QUANTITIES OF WASTE AND SPENT FUEL

This section presents data on existing and predicted quantities of nuclear wastes, both in the U.S. and worldwide, to assess the magnitude of the nuclear waste problem. Mining and milling wastes and wastes which will be generated during future decontamination and decommissioning of nuclear facilities are not included.

COMMERCIAL WASTES, U.S.A.

At present, only a relatively small amount of commercial HLW exists in the U.S., about 2300 m^3 (610,000 gal) of slurry in storage at the Nuclear Fuels Services (NFS) plant at West Valley, NY. (This includes some defense-waste from production reactors.) About 446,000 m^3 of solid commercial LLW and TRU wastes containing about 123 kg of TRU elements are buried at six sites, as shown in Table 2-1.

Until and unless a decision is made to reprocess spent fuel, there will be no more commercial HLW produced in the U.S. Unreprocessed spent fuels will accumulate with the growth of the nuclear power industry. A DOE Task Force (2) for Review of Nuclear Waste Management, under the direction of J. M. Deutch, predicted U.S. industrial growth using a "low" rate and a "high" rate consistent with the National Energy Plan. Figure 2-1 shows these predictions through the year 2000. The high growth rate agrees with a prediction issued by the Oak Ridge National Laboratory (ORNL).(4) Also shown is an ERDA growth rate predicted in 1976. This rate is significantly higher than current growth rate predictions. The DOE Task Force report also gives the spent LWR (⅔ PWR, ⅓ BWR) fuel which will be discharged under each of the two predicted growth curves. These data are shown in Table 2-2.

With the DOE high growth rate, some 8100 metric tons of heavy metal (MTHM) would be discharged in the year 2000 from U.S. reactors and the accumulated discharge from 1975 to 2000 would be about 97,900 MTHM.

The DOE report (2) also estimates the amounts of commercial LLW and TRU wastes that would be produced in each of these cases. These data are summarized in Table 2-3 for both cases because they represent an extreme case range of conditions. The DOE report assumes

TABLE 2-1. Existing Commercial TRU and LLW Wastes, January, 1977 (2)

Site	TRU Waste Buried kg TRU Elements	LLW Buried Thousands of m^3
Barnwell, SC	0	97.7
Beatty, NV	14.3	55.8
Hanford, WA	22.7	14.4
Maxey Flats, KY	69.1	140.2
Sheffield, IL	13.4	68.0
West Valley, NY	3.6	70.0
Total	123.1	446.1

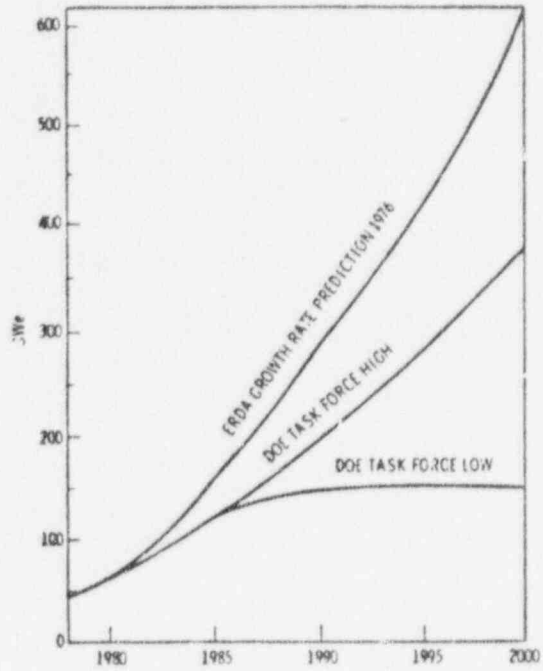

FIGURE 2-1. Predictions of the Growth of Nuclear Power in the United States

TABLE 2-2. Spent Fuel Generated, Annual and Cumulative, in the United States

Year	Metric Tons of Heavy Metal (MTHM)(a)			
	148 GWe in 2000		380 GWe in 2000	
	Annual	Cumulative	Annual	Cumulative
Existing	--	2,300	--	2,300
1977	1,000	3,300	1,000	3,300
1978	1,100	4,400	1,100	4,400
1979	1,300	5,700	1,300	5,700
1980	1,300	7,000	1,300	7,000
1981	1,400	8,400	1,400	8,400
1982	1,600	10,000	1,600	10,000
1983	1,900	11,900	1,900	11,900
1984	2,200	14,100	2,200	14,100
1985	2,700	16,800	2,700	16,800
1986	2,900	19,700	2,900	19,700
1987	3,400	23,100	3,400	23,100
1988	3,600	26,700	3,600	26,700
1989	3,700	30,400	3,900	30,600
1990	3,700	34,100	4,200	34,800
1991	3,800	37,900	4,600	39,400
1992	3,800	41,700	4,900	44,300
1993	3,800	45,500	5,200	49,500
1994	3,800	49,300	5,700	55,200
1995	3,700	53,000	6,000	61,200
1996	3,700	56,700	6,500	67,700
1997	3,700	60,400	6,900	74,600
1998	3,600	64,000	7,300	81,900
1999	3,600	67,600	7,800	89,760
2000	3,500	71,100	8,100	97,800

(a) Unpackaged volume is about 13.1 ft^3/MTHM. Excludes discharges prior to 1975.

TABLE 2-3. Future Range of U.S. Commercial LLW and TRU Waste Generation (2)

	Low Growth[a]					High Growth[b]		
		LLW, 1000 m3		TRU, 1000 m3			LLW, 1000 m3	
Year	GWe	ann.	accum.	ann.	accum.	GWe	ann.	accum.
1977	48	92	92	0.28	0.28	48	48	92
1980	61	110	400	0.28	1.10	61	110	400
1985	127	100	850	0.28	2.50	127	210	1260
1990	148	86	1280	0.28	4.00	195	310	2650
1995	148	100	1780	0.28	5.40	283	450	4650
2000	148	110	2350	0.28	6.80	380	590	7360

(a) Volume reduction employed
(b) No volume reduction
(c) Volume of TRU in DOE Task Force study is same for both low and high cases because it is assumed these wastes will be combined with larger volumes of DOE TRU wastes in a DOE repository.

conditions designed to minimize wastes in the low case (148 GWe in 2000) and maximize wastes in the high case (380 GWe in 2000).

Platt and McElroy summarize data on LWR wastes generated per GWe-y in a once-through (no reprocessing) fuel cycle and for the LWR fuel cycle with full reutilization of plutonium.(5) This data, reproduced in Table 2-4, can be utilized to predict the approximate volumes of future waste. The area under the DOE high growth curve in Figure 2-1 integrates to about 4400 GWe-y for the period 1978 to 2000. Based on 25 m^3 of spent fuel per GWe-y, the volume of accumulated spent fuel in 2000 would be 110,000 m^3.

If reprocessing were reinstituted in the U.S., we could also estimate the volume of future waste from that type fuel cycle. Past experience in reprocessing LWR [illegible] indicates that the volume of HLW b[illegible] solidification is about 20 m^3 per [illegible] of energy production. For 4400 [illegible] there would be 88,000 m^3 of liquid before solidification. Vitrification w[illegible] reduce this to about 12,000 m^3 of bo[illegible] silicate glass. With the reprocessi[illegible] cycle, additional wastes would be gene[illegible]ated. Table 2-4 shows that reprocessing almost doubles the volume of wastes, but the aggregate fissile plutonium content in the waste is reduced about 50-fold. In addition, the volume of HLW compared to that of spent fuel is nearly a factor of 10 less; both require similar handling for disposal.

In summary, based on nuclear power growth consistent with the National Energy Plan and continued deferral of fuel reprocessing, commercial nuclear activities could be expected to generate about 110,000 m^3 of spent fuels, about 7,000 m^3 of TRU wastes, and up to about 7 million m^3 of LLW by the year 2000. If the spent fuels were reprocessed, about 12,000 m^3 of vitrified HLW waste would be produced; there would be increases in the production of LLW and TRU wastes.

COMMERCIAL WASTES, FOREIGN

Future production of nuclear [illegible]stes outside the U.S. will, of course, depend on the growth of nuclear power there. In March 1978, a DOE-sponsored summary was issued of openly-available information on foreign and international waste management programs and technology.(6) Table 2-5, [illegible]aken from that summary, shows power-reactor plans for 40 countries throughout the world. Data in Table 2-5 can be summarized to indicate a nuclear power growth from about 210 GWe (operable, under construction, or committed) in 1978 to about 800 GWe in the year 2000. The DOE Task Force report indicates a growth from about 85 GWe (operating) in 1978 to about 600 GWe in 2000. (The U.S. has offered to store

TABLE 2-4. LWR Nuclear Wastes, 10-y Cooling, 1 GWe-y (Packaged Volumes) (5)

Fuel Type	Units			
	Volume, m3	wt U, kg	wt Pu, kg	Radioactivity, MCi
Unreprocessed Spent Fuel	25	36,400	310	13.0
Packaging Waste	9			0.0002
Reprocessed Wastes				
Vitrified HLW (10 y after discharge from reactor)	2.8		3.4(a)	10.4
Intermediate-Level Waste	52		1.7(a)	0.07
Hulls and Spacers	7.4(b)		0.24(a)	0.03
Mixed Oxide Fuel Waste	18(c)		0.65(a)	

(a) contained Pu
(b) compacted
(c) in concrete

10% of foreign spent fuels if the countries involved forego spent fuel reprocessing. The task force report estimates the amount of spent fuel that would be received for storage under this arrangement.) Earlier predictions (7,8) were for significantly higher growth rates.

Compilations of existing and expected nuclear wastes worldwide are not available. However, since most of the reactors planned are LWRs, experience in the generation of wastes in the LWR cycle can be applied and the amounts of wastes which will be produced in the future can be approximated from data in Table 2-4.

If we assume that the free world (except U.S.) nuclear-power-generating capacity will grow to 600 GWe by the year 2000, then it can be estimated that in the period of 1978 to 2000, about 6600 GWe-y of nuclear energy will be generated. Table 2-6 summarizes the amounts of unreprocessed spent fuel and various nuclear wastes which would be produced without spent fuel reprocessing, and with spent fuel reprocessing.

TABLE 2-5. Nuclear Power Projects, Foreign

Country	Committed		Projected	Year
	GWe		GWe	
Argentina	0.9	(PHWR)	9	2000
Austria	0.7	(LWR)		
Belgium	5.5	(LWR)		
Brazil	3.1	(LWR)	10	1990
			70	2000
Bulgaria	1.8	(LWR)	7	
Canada	11.9	(PHWR)	75	2000
Chile			0.6	1988
Cuba	1.7	(LWR)	Several reactors	1990
Czechoslovakia	1.8	(LWR)	12	1990
	0.1	(GCHWR)		
Egypt	0.6	(LWR)	7	2000
Finland	2.2	(LWR)	6	1985
France	37.2	(LWR)	40	1985
	2.2	(GCR)		
	0.07	(GCHWR)		
	1.4	(LMFBR)		
Germany (East)	2.7	(LWR)		
Germany (West)	26.3	(LWR)	30	1985
	0.3	(LMFBR)		
	0.3	(THTR)		
	0.05	(PHWR)		
Hungary	1.8	(LWR)	5.7	1990
			12	2000
India	0.4	(LWR)	6	1990
	1.3	(PHWR)	20	2000
Iran	4.2	(LWR)	22	1992
			34	2000
Israel			0.6(b)	
Italy	5.1	(LWR)	9-13	1985
	0.2	(GCR)		
	0.04	(LWCHR)		
Japan	19	(LWR)	40	1986
	0.3	(LMFBR)	90	1990
	0.2	(GCR)		
	0.2	(LWCHR)		
Korea (South)	1.2	(LWR)	4	1986
	0.6	(PHWR)	40-46	2000
Kuwait			6(b)	
Luxembourg	1.3	(LWR)		
Mexico	1.3	(LWR)	10	1990
Netherlands	0.5	(LWR)	3.5	?
New Zealand			1.2	?
Pakistan	0.1	(PHWR)	4.8	1990
			16	2000
Phillipines	1.2	(LWR)	3.9	2000
Poland	0.4	(LWR)	8	1990
Rumania	0.4	(LWR)	+ 16 PHWR	2000
South Africa	1.8	(LWR)	12	2000
Spain	15.6	(LWR)		
	0.5	(GCR)		
Sweden	9.4	(LWR)		
Switzerland	5.9	(LWR)		
Taiwan	4.9	(LWR)		
Turkey			4.4	1990
United Kingdom	11.4	(GCR)	100	2000
	0.1	(HWLWR)		
	0.3	(LMFBR)		
U.S.S.R.	5.0	(LWR)	26	?
	14.9	(LGR)		
	1.0	(LMFBR)		
Yugoslavia	0.6	(LWR)	0.6	1987

(a) Operable, under construction, or on order.
(b) With desalination capacity.

TABLE 2-6. Future Foreign Wastes from 6600 GWe-y LWR Production

	Volume, m^3 (a)
Once-through cycle	
Spent fuel	165,000
Conditioning waste	59,000
Reprocessing with full plutonium utilization	
Vitrified high-level waste	18,000
Intermediate-level waste (contained in concrete)	340,000
Cladding and hardware (compacted)	49,000
Mixed oxide fuel waste (contained in concrete)	119,000

(a) Based on quantities of waste produced per GWe-y.(5)

All the wastes shown in Table 2-6 will require special treatment, e.g., disposal as HLW or TRU wastes. The worst-case total volumes for each option are between 200,000 and 500,000 m^3. These volumes are equivalent to cubes about 60 to 80 m on a side. While treatment, transportation, and disposal of these wastes will be costly because of the special precautions required, the volumes are small compared to those of conventional mining.

DEFENSE WASTES, U.S.

The origin and status of defense high-level wastes already in storage at the Savannah River Plant (SRP), the Hanford Reservation, and the Idaho National Engineering Laboratory (INEL) are given in Section 4. In addition to these wastes, large amounts of LLW and TRU wastes are buried or in retrievable storage at these and at other government-owned sites. Table 2-7 shows the amounts of DOE wastes presently in existence and which are expected to be in existence in 1985.(9) The intermediate-level wastes at ORNL are unique and may be considered disposed of when injected into shale by hydrofracturing.

Currently, DOE is conducting studies on various options for permanent disposal of these wastes, particularly the HLW and TRU wastes. Details on these studies are presented in Sections 4 and 5.

Volumes of the DOE HLW are large. However, because the integrated exposure level of defense-production fuels is relatively low and because the defense reprocessing and waste storage operations created relatively large volumes of HLW per ton of heavy metal processed, the wastes are quite low in radioactivity compared to commercial HLW. Von Hippel and Krugmann made a comparison between defense HLW and commercial HLW (currently largely unreprocessed spent fuel) based on ^{90}Sr inventory.(10) This isotope was chosen because of its relatively long half-life (28 y) and because it has a high potential risk for a few hundred years. Their survey indicated that: 1) currently, the inventory of ^{90}Sr from U.S. commercial power generation is about equal to that in all defense HLW; and 2) based on their predicted commercial power growth rate, in 1985 the annual commercial production of ^{90}Sr will equal the total inventory in all defense HLW. Such surveys emphasize that, although the current deferred spent fuel reprocessing situation may allow more emphasis on defense waste disposal, the commercial waste problem is growing rapidly and adequate solutions must be vigorously sought.

TABLE 2-7. Defense Waste Inventories

	January 1, 1977	December 1985 Projections
High-level waste (millions of gallons)		
Liquid in tanks	34.9	15.3
Salt cake and sludge in tanks (Savannah River and Hanford)	38.6	53.7
Calcine in bins (Idaho)	0.4	1.1
Total	73.9 (270,000 m^3)	70.1 (260,000 m^3)
ORNL intermediate-level liquid waste (millions of gallons)		
Liquid in tanks	0.2	0.1
Sludge in tanks	0.4	0.0
Liquid mixed with cement and disposed of as grout in shale formation underlying site	1.1	2.5
Total	1.7 (6,300 m^3)	2.6 9,600 m^3)
Low-level and TRU solid waste (millions of m^3)		
Non-TRU in land burial sites (17 sites)	0.83	1.15
TRU in land burial sites (6 sites)	0.37	0.37
TRU retrievably stored (6 sites)	0.04	0.11
Total	1.24	1.63

Section 3

STATUS OF REPROCESSING AND REFABRICATION

No commercial spent nuclear fuel is being reprocessed in the U.S. at the present time. Defense fuel reprocessing continues at the U.S. DOE sites, and commercial fuel reprocessing is in progress in several foreign facilities. These operations are discussed in more detail in Appendix B.

Solvent extraction is the basic separations technique used in all current spent-fuel reprocessing facilities in the world. The technology for solvent extraction is well established. Many plant years of experience with the technique have been accumulated without any incident of significant risk to the environment. Most of the highly active radioactive wastes have been stored as liquids; they are discussed in Section 4.

Nuclear fuel reprocessing as currently practiced separates high purity uranium and plutonium from other spent fuel constituents to minimize problems of refabricating the uranium and plutonium into new nuclear fuels. Because the level of penetrating radiation of these highly purified products is relatively low, both can be handled without expensive and sophisticated remotely controlled equipment. Extensive reprocessing of spent nuclear fuels may result in large quantities of purified plutonium throughout the world, which might be fabricated into nuclear explosives if it were produced without adequate safeguards.

These considerations led to the U.S. decision to suspend reprocessing of spent commercial nuclear fuels and to attempt to persuade other nations to do so until the potential for nuclear weapons proliferation can be minimized. One step in this direction is the International Nuclear Fuel Cycle Evaluation (INFCE) program.(11) The INFCE was established under U.S. sponsorship to study world energy needs and how nuclear energy can be employed to safely satisfy these needs with minimum potential for nuclear weapons proliferation. One of the activities of the INFCE is to evaluate fuel cycle processes which have improved safeguards.

Other efforts in the U.S. include the study of alternative nuclear fuel cycles and reprocessing/refabrication schemes which would not result in weapons-usable materials. These efforts are discussed in Appendix B, but are beyond the scope of this report.

Section 4

INTERIM STORAGE OF SPENT FUEL AND HIGH-LEVEL WASTES

SPENT FUEL

In a closed nuclear fuel cycle, spent fuels are stored at the reactor site for a short period (usually < 1 y) and then are transferred to storage at a reprocessing plant until they are processed. Deferral of fuel reprocessing in the U.S. has emphasized the need for facilities for longer interim storage of spent fuels and presents the possibility that the fuels will finally be isolated without processing.

Spent fuels are stored at reactor sites and at reprocessing plants in specially-designed water basins, such as the General Electric Morris plant pool shown in Figure 4-1. The fuels hang in racks designed to maintain proper spacing of the fuels so that a critical nuclear assembly cannot be formed. They are generally not in canisters but are in direct contact with the basin water, which is circulated and cooled. Spent-fuel storage basins are sized and operated so that there is room for one reactor charge in case an unscheduled reactor discharge is required.

Some radioactive materials may enter the basin water from surface contamination on the fuel assemblies and through the leaching of fuel material from defective elements. Several techniques, including ion exchange, filtration, and skimming, are used to remove these materials to keep the radioactivity level of the basin low and to minimize exposure of operators. Pool chemistry, (e.g., addition of hydrazine, boric acid, and lithium hydroxide) and pH control are tailored to the particular fuel being stored.(12)

In the once-through fuel cycle imposed by deferred spent fuel reprocessing, longer interim storage of spent fuels will be required, perhaps in the 20-to-100-y range. This poses the question of the long-term reliability of water basin storage of spent fuels. A. B. Johnson has made a survey of the experience to date with water basin storage of spent fuels.(13) The survey covers 30 U.S. and Canadian storage pools. Spent fuel storage experience is also summarized for the British pools at Winfrith and Windscale and for a German pool at Karlsruhe.

At the end of 1976 there were approximately 8700 power reactor fuel bundles in storage in U.S. pools; approximately 90% of these have Zircaloy cladding. The remainder have stainless steel cladding. Approximately 70,000 smaller Zircaloy-clad bundles were in storage in Canadian pools at the end of 1976.

Maximum pool residence to date for Canadian fuels is 14 y. Zircaloy-clad U.S. fuel has been in storage up to 18 y. Experimental and commercial stainless-steel-clad fuel has been stored up to 7 y. Unirradiated stainless-steel fixtures have been stored up to approximately 20 y. Maximum burnups for stored commercial fuel are approximately 33,000 MWd/tU for both Zircaloy- and stainless-clad fuel.

Experience in water basin storage of irradiated nuclear fuel has been very good. To date, based on visual observation of the fuel and monitoring of the basin water, there has been no evidence of degradation of either Zircaloy or stainless steel cladding. Release of fuel material from defected fuels to the basin water has been minimal even though most defected fuel is stored without enclosure in a canister. Mechanical damage to fuels or the water basin during handling and storage of fuels has been minimal.

Following the survey of past experience in water basin storage of spent fuels, an

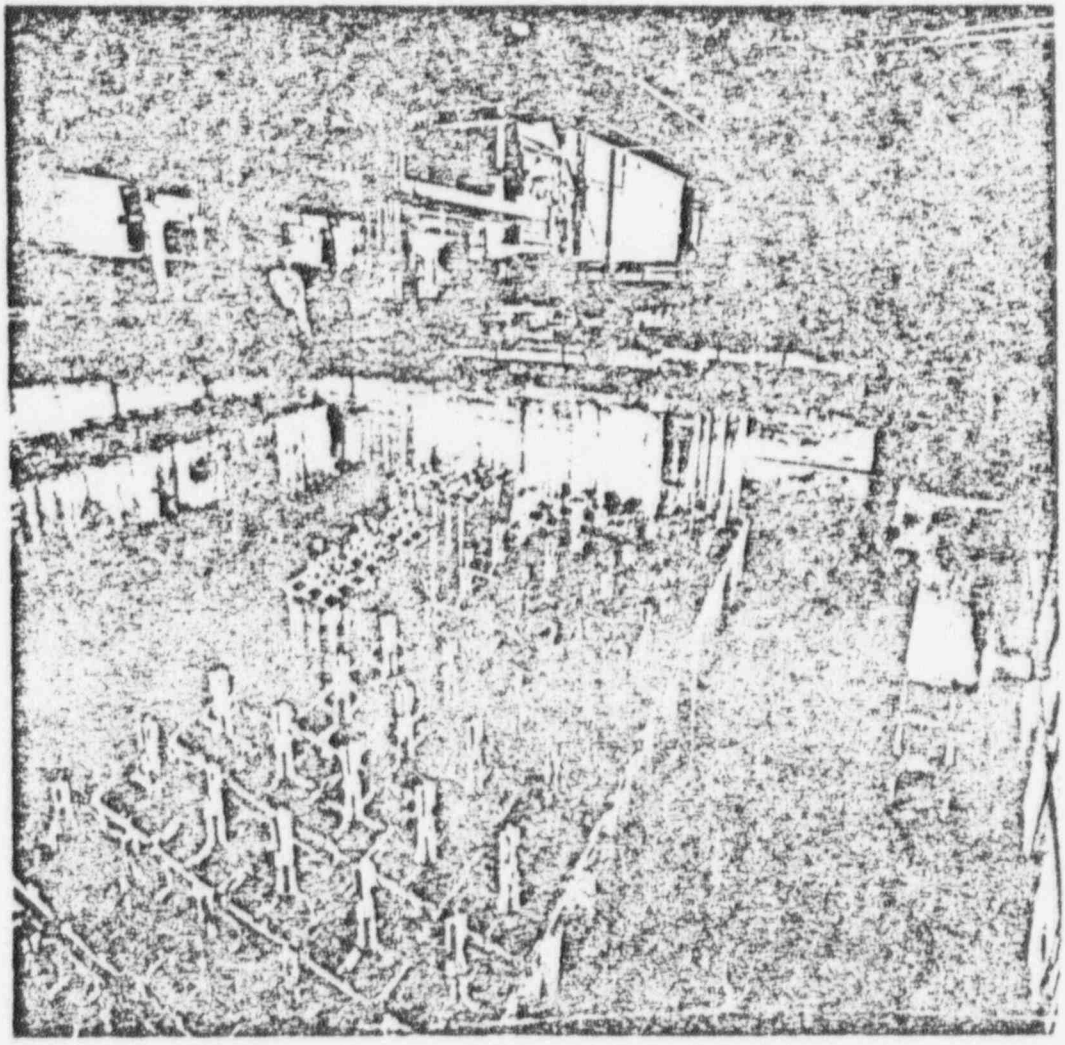

FIGURE 4-1. G. E. Morris Operation - Fuel Pool

assessment of potential degradation mechanisms for materials in pool storage was made.(13) This assessment included studies of hydriding effects in Zircaloy and stainless steel, fission product (e.g., I, Cs) attack, and helium embrittlement. Corrosion studies included aqueous, biological, galvanic, crevice pitting and stress-corrosion cracking in cladding. The assessment also included localized attack at fuel defects, crud layers, and pool equipment corrosion. These studies were summarized by the statement, "Review of the corrosion literature has not developed major concerns regarding obvious mechanisms which have high probability to degrade fuel bundle materials."(13)

The assessment concluded that the prospects are favorable to extend storage of spent nuclear fuel in water pools. It was recommended that periodic examination of selected pool-stored fuel be made to

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determine if slow degradation of fuel bundle materials is operative to an extent that could cause problems in the 20- to 100-y time frame.

Dry spent fuel interim storage options are also being considered.(14-16) However, a fully developed dry storage option is still several years away.(13)

Under the present U.S. deferral of spent fuel reprocessing (and the offer to store 10% of foreign spent fuels), more storage space will be needed than is available at reactor sites. The DOE Task Force Review of Nuclear Waste Management (2) concludes that the first away-from-reactor (AFR) storage will be needed by 1983. How this AFR storage is to be achieved--by government or private interests--is currently under discussion. One suggestion for the initial need is to utilize the fuel storage basin at the AGNS Plant, Barnwell SC.(17) Interim retrievable storage of spent fuels in a geologic formation may also be done on a test basis to determine the feasibility of permanent isolation of the fuels in the site.

HIGH-LEVEL WASTES

Present nuclear fuel reprocessing schemes separate and purify uranium and plutonium and leave virtually all of the nongaseous fission products and the nonplutonium actinide elements in a waste stream. This waste stream is commonly called the high-level waste (HLW), as shown in Figure 4-2.

High-level waste has been stored as a liquid or slurry at the reprocessing site, pending further treatment to convert it to a form for long-term storage or isolation. For commercial fuel reprocessing plants, this interim storage period for liquid waste is limited by existing regulations to 5 y.(18) The NFS plant was granted an exemption from this regulation since it was in operation at the time this became law.

Most of the U.S. experience with interim storage of HLW has been with defense wastes. Because several different processing schemes have been used for defense fuels (Bi_2PO_4, Redox, Purex, and ICPP processes) and because the wastes from different processes have been mixed in the storage tanks, the stored wastes are not uniform in composition. Also, since the defense wastes have generally been made alkaline (by adding NaOH) before storage, they have a much higher salt content than commercial high-level waste, which is acidic. Acidic high-level defense wastes have been stored only at the Idaho Chemical Processing Plant (ICPP).(19)

Alkaline HLW has been stored in carbon-steel-lined, reinforced concrete tanks buried underground. Early tanks used at Hanford had a single steel liner. Stress cracks have developed in several of these tanks, and they have developed leaks. The wastes have been removed from such tanks and transferred to nonleaking tanks. The tanks used at Savannah River and newer tanks being built at Hanford have a double-walled steel liner with an air space between the walls and provision for pumping any liquid waste that may leak from the inner liner back into the tank. A section of a double-walled tank is shown in Figure 4-3.

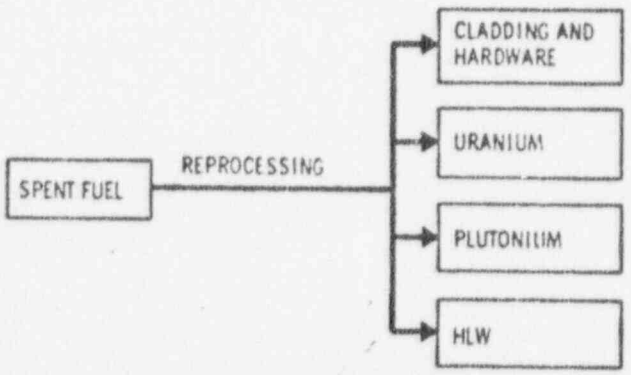

FIGURE 4-2. Origin of High-Level Waste

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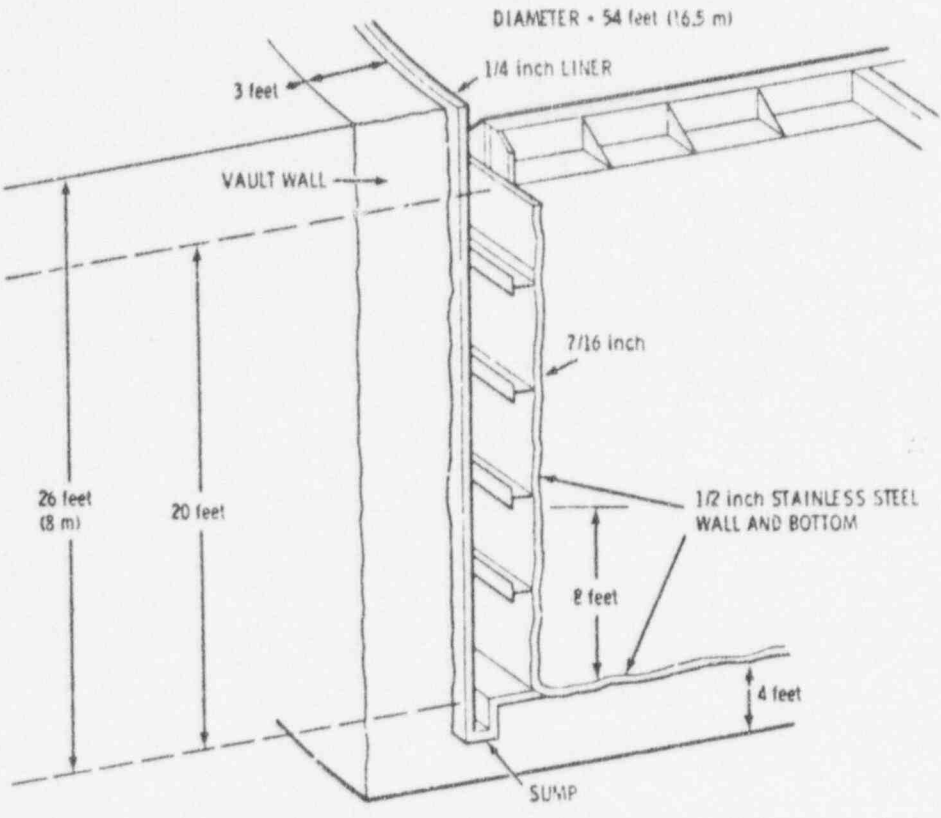


FIGURE 4-3. Cross Section of Tank for Storage of High-Level Liquid Waste

Stress cracks have developed in the inner liner of some Savannah River tanks, but no waste is known to have escaped from the outer liner. The double-liner tanks appear to provide safe interim storage for high-level wastes. While the leaks which have occurred in the single-lined Hanford tanks have received considerable attention, none of the leaked material has entered the water table or biosphere, and no significant risk to the surrounding biosphere has been identified.

Acidic high-level waste is stored at the ICPP in stainless-steel tanks contained in stainless-steel-lined concrete vaults. The vaults can be monitored for leaks from the tanks. No leaks have ever been found.

Experience with interim storage of waste from commercial nuclear fuel reprocessing plants is very limited in the U.S. The only commercial reprocessing plant which has operated in the U.S. is the NFS plant at West Valley, NY; however, this plant has not been operational since 1972. Most of the waste produced while the plant was operating (ca. 600,000 gal) was made alkaline and stored in a double-lined carbon steel tank similar to those at Savannah River. The only acidic waste stored at the NFS plant was produced while thoria fuels were reprocessed. About 12,000 gal of this waste is stored in a 15,000-gal, stainless-steel tank contained in a stainless-steel-lined concrete vault.(20)

Activities and Plans for Defense HLW

While interim storage of liquid high-level wastes in properly designed tanks appears adequately safe, operations are in progress at all three DOE sites to reduce the volume of the HLW and to make these wastes less mobile. At ICPP, the wastes are being calcined to a solid in a fluidized-bed waste calcination facility (WCF).

The solids are stored in underground storage bins contained in concrete vaults. The WCF has been operated as a plant-scale facility since 1963 without any significant releases of radioactivity to the environment. A new calcination facility (NWCF) is being built for operation in 1980.(19)

At Hanford, (21) cesium and strontium are being removed from the wastes, and the residual wastes are concentrated to damp salt cakes, damp sludges, and residual (not evaporable) liquor. Cesium and strontium are removed because in the aged wastes (>5 y), ^{137}Cs and ^{90}Sr are the major heat-producing radioactivities left. With the removal of these wastes, the remaining wastes can be stored as damp solids or liquids in the large tanks without concern about overheating. The Sr and Cs are converted to solids, canned in double-walled containers, and stored in special water-cooled vaults.

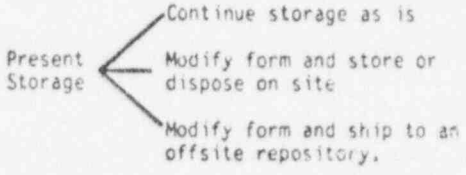
When the program is completed on currently-stored wastes, the high-level waste inventory at Hanford will consist of approximately:

- 25,000,000 gal (bulk) of damp salt cake
- 11,000,000 gal (bulk) of damp sludge
- 3,000,000 gal of liquid wastes, which will remain in active processing
- 2900 capsules containing compounds of strontium or cesium.

Process improvements may reduce the quantity of residual liquor.

At the Savannah River Plant, (22) the alkaline wastes are allowed to age to reduce the radioactivity through decay. During the aging, a sludge settles to the bottom of the tanks, carrying most of the radioactive constituents with it. The primary radioactive constituent of the supernatant liquid is cesium. After it has aged, the supernatant liquid is processed through a continuous evaporator for dewatering, and the concentrate is transferred to a cooled tank, where suspended salts settle out. The supernate is returned to the evaporator and the process is repeated until the waste is converted to a damp salt cake. In 1985 it is expected that the inventory of HLW at the SRP will occupy 31 waste tanks and will consist of 13,300,000 gal of sludge and 5,600,000 gal of liquid waste. The small amount of commercial HLW produced and stored at the NFS plant has not been further processed.

Although the HLW interim storage at the DOE sites and at the NFS plant appear adequately safe for many years, procedures for putting them into more immobile forms and for isolating them from the biosphere are being studied. Documents have been prepared for each site in which a number of alternative routes are described and for which preliminary cost estimates have been prepared.(19,21,22) The basic options are the same for three sites:



After suboptions were considered, such as waste form and possible removal of TRU elements numerous potential options were derived for each site. Before any decisions on further treatment of these wastes are made, environmental impact statements defining the risks and cost-benefits of each route will be completed. These are in preparation.

Section 5

CONVERSION AND PACKAGING OF WASTES FOR ISOLATION

HIGH-LEVEL WASTE TREATMENT

All nations reprocessing nuclear fuels now plan to solidify the liquid HLW to a less mobile form for interim storage and long-term disposal. Borosilicate glass has been developed by several countries for this purpose.(23-25) The relatively insoluble glass is the inner-most barrier in a geologic disposal scheme comprised of waste, engineered barriers, and geologic barriers. Other waste forms, such as crystalline materials, are also under consideration.

Due to the decision to defer spent fuel reprocessing in the U.S., emphasis has shifted from solidifying commercial HLW to solidifying defense HLW. Results of studies in this area should apply to potential alternative fuel cycle wastes, as well as to the commercial HLW stored at the NFS plant. A preliminary assessment of the potential effects of alternative fuel cycles on HLW vitrification was published in 1978.(26)

U.S. HLW Conversion System

In the U.S. basic HLW conversion system, aqueous wastes are calcined, mixed with glass-forming reagents, then melted and allowed to cool. As the mixture cools, it solidifies to a glassy product, as shown in Figure 5-1.

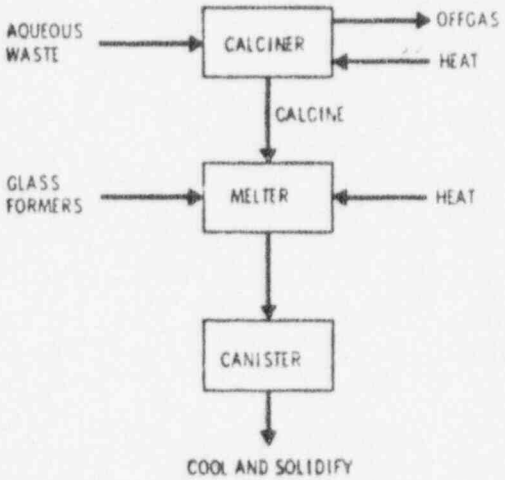


FIGURE 5-1. Reference HLW Conversion System

The wastes are calcined in a spray calciner, shown schematically in Figure 5-2. High-level aqueous waste is sprayed into a heated (~700°C) chamber where the droplets of spray are dried and calcined (primarily to oxides) as they fall through the chamber. Particulate matter in the off-gas stream is caught on filters, which are periodically cleaned by a back pulse of air. Glass-forming frit is added to the calcine as it falls from the calciner to the melter.(27)

Two melters have been highly developed in the U.S.: an in-can (batch) melter, and a ceramic (continuous) melter. The in-can melter, shown in Figure 5-2, consists of the waste canister contained in a six-zone furnace; forced air cools the glass that is heated by the decay of the contained fission products. Calcine and glass-forming frit fall into the heated canister, where they are melted at a temperature of ~1050°C. When the canister is filled, calcine and frit are diverted to an empty canister; the filled canister is then cooled and removed from the furnace.

Figure 5-3 illustrates the joule-heated ceramic melter. Calcine and frit are fed into a ceramic-lined chamber, where they are melted by alternating current passing between electrodes immersed in the melt.

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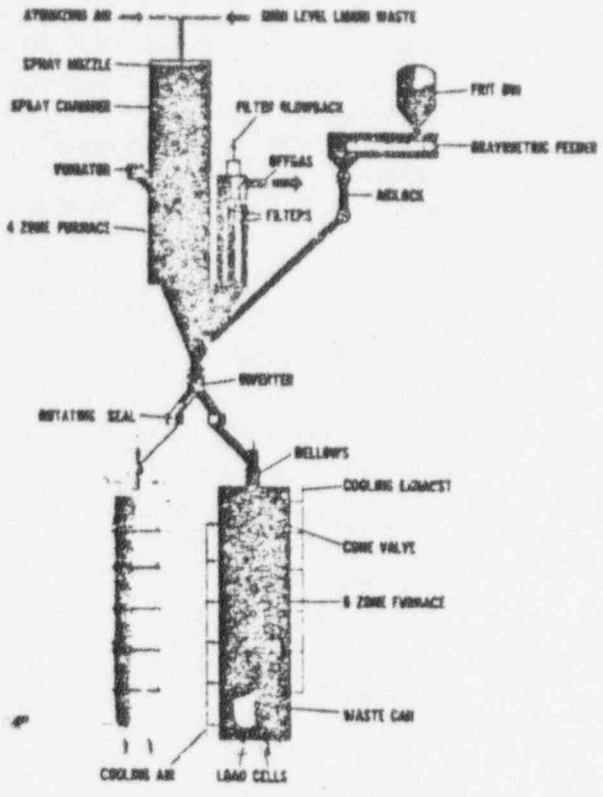

FIGURE 5-2. Schematic of Spray Calciner/In-Can Melter Process

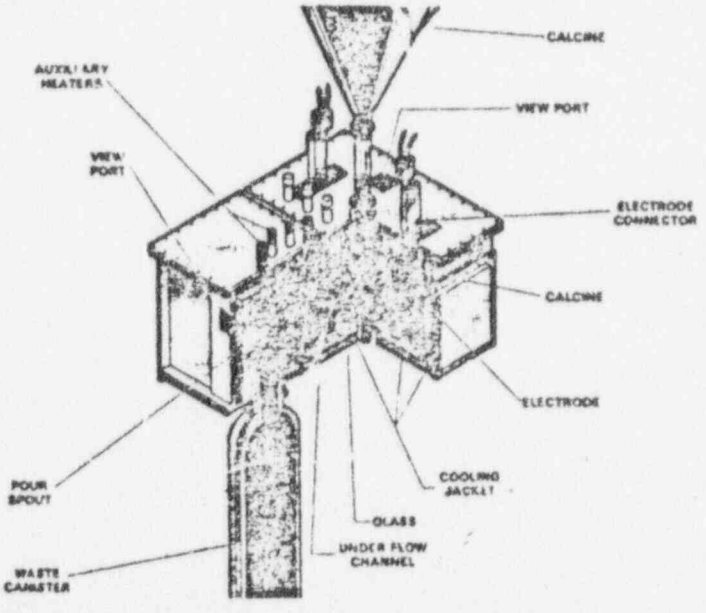

FIGURE 5-3. Schematic of Spray Calciner Ceramic Melter System

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The melt overflows continuously and may go into a canister or into molds for producing glass shapes, such as marbles. Both types of melters and the spray calciner are being developed for the DOE by PNL.

Development of the spray calciner on an engineering scale with both nonradioactive and radioactive wastes has been extensive. During the Waste Solidification Engineering Prototype Progr (1966 to 1970), 13 canisters of solidified waste were produced in the spray calciner with actual high-level radioactive waste.(28)

Recent efforts with the spray calciner have been directed toward scaling up the equipment so that it can meet the high throughput requirements for defense wastes, such as those at Savannah River. One plant-scale calciner has calcined more than 300 ℓ HLW/h (5), three times the HLW processing rate required by a large commercial fuel reprocessing plant, such as the Barnwell 1500 t/y plant. The main feature that makes the spray calciner attractive for use with both commercial and defense HLW is its ability to calcine a wide variety of waste compositions.(29) In addition, it is a simple system, enabling fast startup and shutdown, with little holdup of radionuclides.

The in-can melter has been demonstrated in lab-, pilot plant-, and plant-scale systems. Over 40 engineering-scale canisters of nonradioactive glass and two of actual radioactive glass have been produced. The in-can melter itself is well developed. During the past two years, the mechanical features have been improved and the equipment has been enlarged to achieve a high throughput. A 12-in. dia in-can melter will process 50 kg/h of melt, typical of what would be required for a large 1500 t/y reprocessing plant. Larger diameter 20- to 24-in. canisters have processed approximately 90 kg/h. Although this approaches the requirements for defense waste needs, it may be desirable to use two melters in parallel.

Some advantages of the in-can melter are: 1) its overall simplicity since no melt transfers are required, 2) a low sensitivity to changes in composition, and 3) a minimum number of process steps, because the melter is also the final storage vessel.(30) The process is limited to a maximum melt temperature of about 1050°C with a stainless steel canister; an Inconel® canister permits temperatures up to 1150°C.

Studies on the joule-heated ceramic-lined melter for producing radioactive glasses were begun at PNL in late 1973.(24) Over 17,000 kg of nonradioactive glass have been produced and melters have been at operating temperature for over three years without serious degradation. In January 1977, a large-scale ceramic melter became operational. It has operated at glass-producing rates up to 130 kg/h when fed calcine.(31) This melter was designed to also accept direct liquid feeding.(32) The ability to feed liquid waste directly to the melter enables potential elimination of the calciner, thereby simplifying the system. In April 1978, testing was begun on a spray calciner coupled to a ceramic melter (SC/CM).(5) The SC/CM is capable of producing nonradioactive glass at over 100 kg/h (equivalent to waste from a ~3000 t/y commercial reprocessing plant). No serious operating problems have been encountered to date with any of these melters.

The ceramic melter offers such attractive features as:(5)

- production of several waste forms, such as monoliths or marbles
- sufficient capacity for nearly any size waste plant
- melter operation at higher temperatures than the in-can melter, possibly allowing some improvement in glass quality
- waste storage canisters which do not have to be heated above about 600°C.

Previous U.S. studies on the production of HLW glasses, including the WSEP program discussed earlier, have concerned wastes expected from commercial fuel reprocessing plants. Wastes for the WSEP program were prepared from fresh defense HLLW spiked with a highly radioactive rare-earth fission product mixture. This provided the complete spectrum of fission product activity. It also produced a vitrified waste with a high heat generation (≤300 W/ℓ) in the glass, simulating short-cooled commercial HLW.(28) The 33 canisters of solidified HLW from WSEP are still stored at Hanford and are still being evaluated.

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After eight years of storage no change has been observed in the glass properties. In 1976, a program was started at PNL to design and construct engineering-scale equipment for preparing commercial HLW and to produce a limited number of canisters of borosilicate glass from it. The reference spray calciner in-can melter system is to be used in this fully remotely operated plant. The target date for completing two canisters of this waste is Spring 1979.

Foreign Activities

Conversion of high-level waste to glassy materials for interim storage or ultimate disposal has been adopted in several countries other than the U.S.(23) Processes in France, Germany, and England have been operated at various levels of size and radioactivity, including actual plant operation in France.

France has developed and tested two processes at Marcoule.(23,24) In one, the PIVER process, liquid HLW and glass formers are added to a pot and are calcined. The temperature is raised to melt the contents and the resulting glass is drained into a canister. The process has been tested on full-level Marcoule waste on a pilot-plant basis; 12 t of glass were produced at a maximum activity of 3000 Ci/ℓ.

The second process at Marcoule uses a rotary calciner coupled to a melting furnace.(33) Product glass drains batchwise into waste canisters. The process has been tested on nonradioactive materials in a pilot plant with a throughput of 20 kg glass/h. Based on this continuous process, a demonstration pilot plant (AVM) with a capacity of 15 to 18 kg glass/h has been constructed at Marcoule. Figure 5-4 shows the AVM process and facility.(33) The plant began operation with actual radioactive waste in June 1978. Eighteen m^3 of radioactive HLLW were vitrified, producing 9000 kg of glass during the first campaign.(34)

The AVM process is shown in Figure 5-5. It consists of a rotary calciner and a continuous Inconel melter (1150°C). The stainless-steel canisters of glass are stored in air-cooled pits located adjacent to both the PIVER and AVM processing facilities. A plant (AVH) similar to the AVM is being designed for operation at the La Hague reprocessing plant in France. So the required reprocessing rates at La Hague may be attained, parallel AVM-type calciners and melters will most likely be required.

For several years, German development centered on a spray calciner-melter process to produce a borosilicate glass.(23,24) This process was tested on nonradioactive material in the VERA pilot plant. Since 1977, work has involved a ceramic melter unit in which liquid HLW is fed directly into a melter already at the melting temperature. It has been tested extensively on a pilot-plant scale (30 ℓ/h) with nonradioactive HLW. A drum dryer rising-level inpot melter process (FIPS) has been developed which produces borosilicate glass for thorium-bearing (HTGR) wastes.(23) A 10 kg glass/h pilot plant is planned. Another German process, the PAMELA process, produces phosphate glass in a ceramic melter.(23) The glass is formed into beads and incorporated in a lead alloy matrix. Cold testing of the process has been completed. A 30 to 40 ℓ/h HLW pilot plant is planned for construction at the Eurochemic plant in Belgium to be operational in 1982-1983.

Waste vitrification work in the United Kingdom has concentrated on the FINGAL/-HARVEST process which produces a borosilicate glass by the rising-level process.(23) Glass formers and HLW are added slowly to a pot maintained at 1050°C. Drying, calcining, and melting occur simultaneously. This process was demonstrated in the FINGAL pilot plant with actual waste from Windscale (24), and in a full-scale pilot plant at Harwell using nonradioactive waste. Several pilot plant and industrial-scale facilities based on the process are planned for operation in the 1979 to 1990 period.

Currently, India, Italy, Japan, and the USSR have waste vitrification studies in various stages of progress.(23)

While there are many approaches to the vitrification of high-level nuclear wastes under study and development, the objective in all cases is the same: The waste is being converted to a form having good physical stability and relatively low solubility (i.e., in ground water, brines, etc., which might come in contact with the waste at its disposal location).

Borosilicate Glass

Borosilicate glass has been chosen as the reference glass form in the U.S. because it has a low dissolution rate in water and good physical integrity, can accommodate a wide variety of waste constituents, is relatively slow to devitrify, and is stable to radiation effects. Glass samples have been spiked with the highly

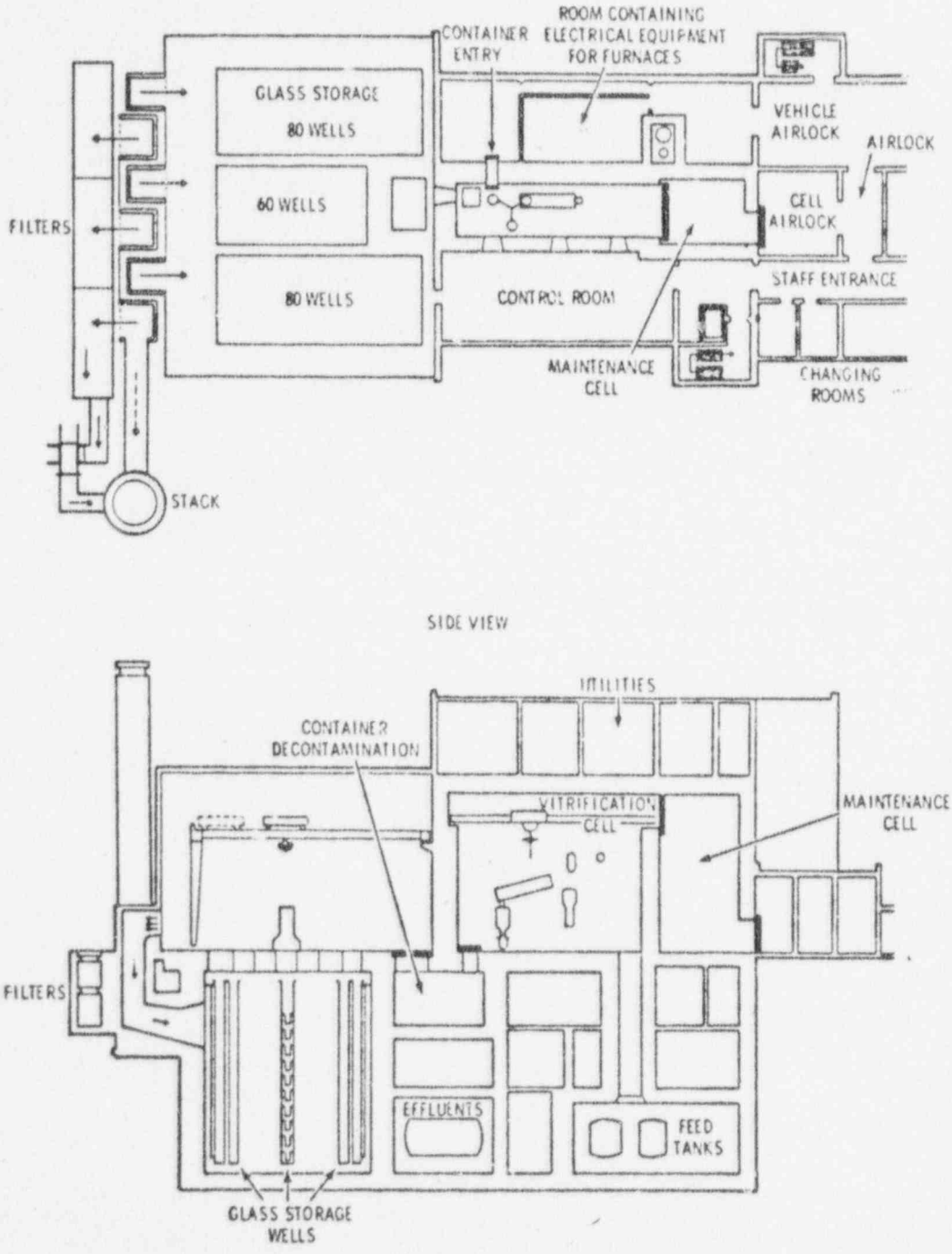


FIGURE 5-4. Plan and Elevation of Vitrification Plant at Marcoule (AVM)

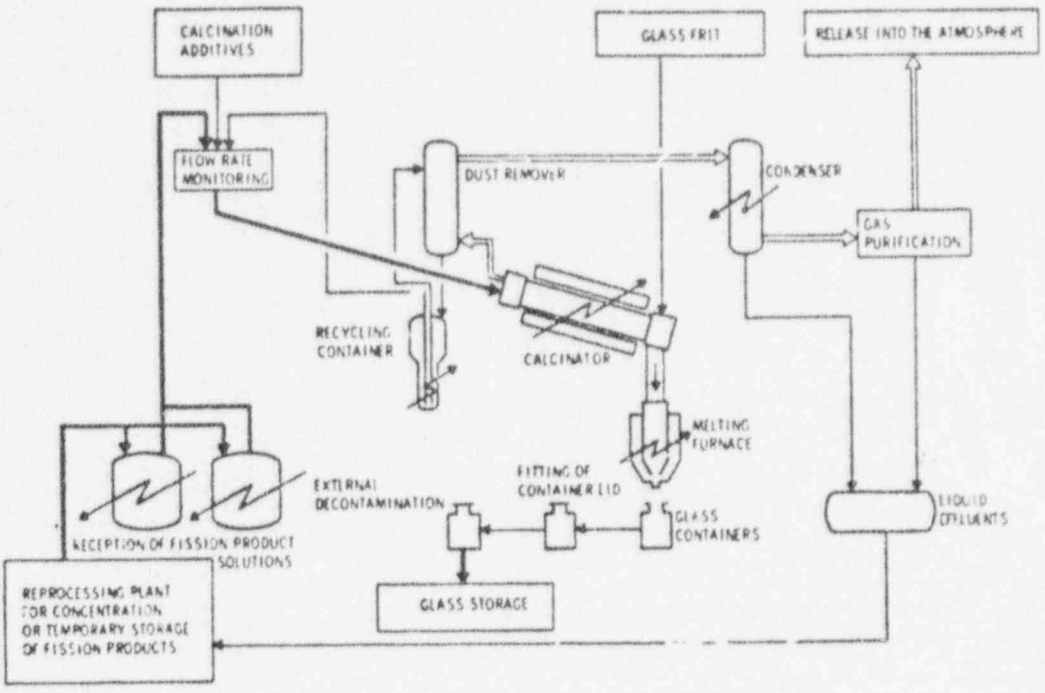


FIGURE 5-5. French AVM Process at Marcoule

active alpha emitter, ^{244}Cm. This accelerates the effects of radiation within the glass.(35) Glass samples exceeding the equivalent of over 500,000 y of radiation dose retain their original physical features, as seen in Figure 5-6. The only effects noted on the glass thus far have been a slight change in density (less than 1%) and a storage of approximately 30 cal/g of energy in lattice defects, a negligible amount. Leach rates and mechanical impact behavior remain essentially unchanged.

Data are also available on devitrification and its possible effect on glass.(25,35) Figure 5-7 shows the leach rate of glass as a function of storage temperature. Crystal growth (devitrification) does not occur below 500°C or above 1000°C, and the maximum growth occurs at 500° to 700°C over long periods. As can be seen in the figure, the devitrification causes only a modest change in the glass leach rate, less than a factor of 10 for these two glasses. Perhaps more important to leach rate is the effect of glass composition; as seen in Figure 5-7, different glasses have different leach rates.

Figure 5-8 compares the rate of attack by water at 99°C on a zinc-borosilicate waste glass as compared to several common materials. The waste glass shown here is almost as inert to attack by water as Pyrex glass. To put these data more in perspective, we can assume that the waste glass, at the time it is emplaced in a dry geologic formation, is an integral cylinder ~30 cm (1 ft) in diameter encased in one or more metallic canisters. Then, we can assume that through some process, water enters the formation, penetrates through the canisters, and immerses the waste glass. If the surface temperature is about that of atmospheric boiling water (100°C), then in 1000 y the outer 1 cm of the glass cylinder will be corroded. This corresponds to about 7 vol% of the waste cylinder.

If we assume a uniform penetration rate, about 15,000 y would be required to completely alter the glass. However, by 1000 y the repository and glass temperatures have decreased to less than 100°C. This is because the fission products in the waste glass continue to decay and are nearly gone after 500 years. Figure 5-9

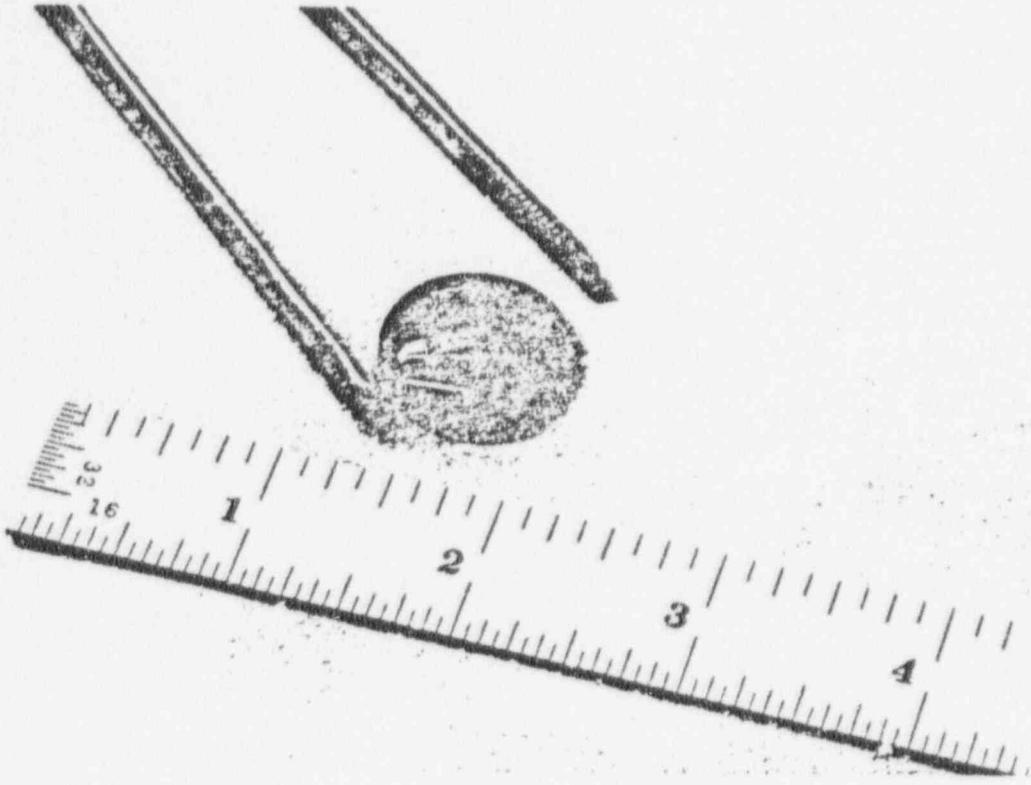


FIGURE 5-6. HLW Glass Aged to the Equivalent of 500,000 y

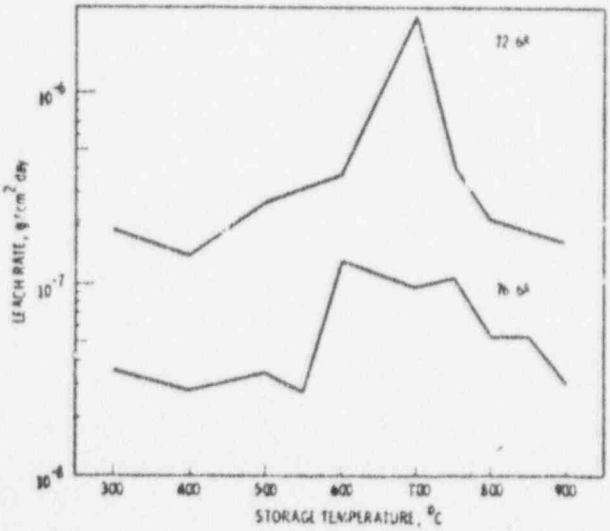


FIGURE 5-7. A Comparison of Devitrification Effects on Leaching of Waste Glass

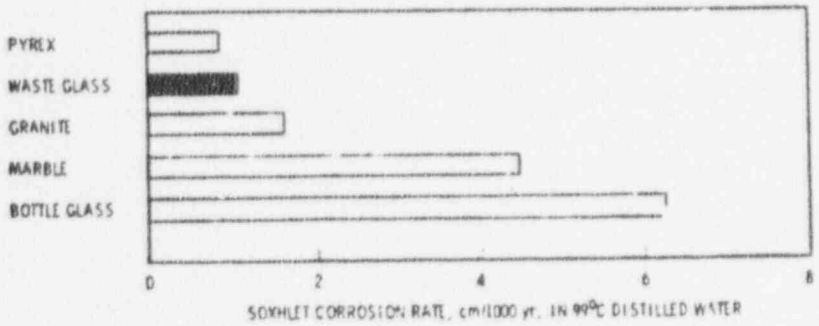
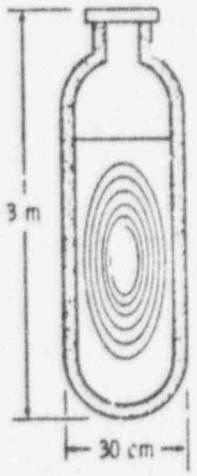


FIGURE 5-8. Comparison of Corrosion Rates of Waste Glass and Other Durable Materials (Based on Weight Loss)



DECAY TIME, yr	HEAT GENERATION RATE, kW	CUMULATIVE α DOSE/GRAM	DOSE-RATE 30 cm FROM SURFACE
1	22	1.0×10^{17}	1.1×10^{8} R/hr
10	3.1	2.5×10^{17}	6.2×10^{4}
100	0.36	7.1×10^{17}	5.8×10^{3}
1000	0.02	1.5×10^{18}	1.6
10,000	0.006	3.0×10^{18}	1.3
100,000	0.003	6.1×10^{18}	0.6

CAN MATERIAL - 304L SS
VOLUME - 0.21 m^3
CONTENTS - 2.5 MTU EQUIVALENT

FIGURE 5-9. Typical High-Level Waste Canister

shows this change in heat content with time. A canister emplaced at ten years' decay time with 3.1 kW will contain only 360 W at 100 y and 20 W at 100 y. The change in waste canister and repository temperature with time is shown in Figure 5-10. These temperatures represent a typical early repository design and are typical of maximum heat loadings in a repository. A more conservative design, with lower heat loadings, would lead to lower temperatures.

As expected from Arrhenius's principle, the rate of attack by water on the glass decreases rapidly with decreasing temperature. If 350°C water could exist in a repository, a 30 cm dia block of waste glass could be completely altered or fully crystallized in a few decades. However, with proper repository design and operation, such temperatures at the glass surface will never exist, particularly if a substantial quantity of water is present to further improve heat transfer. The maximum temperature that can exist at the canister wall in the repository is 375°C (36) under dry conditions, as shown in Figure 5-11. At the same time, the temperature in the rock away from the can

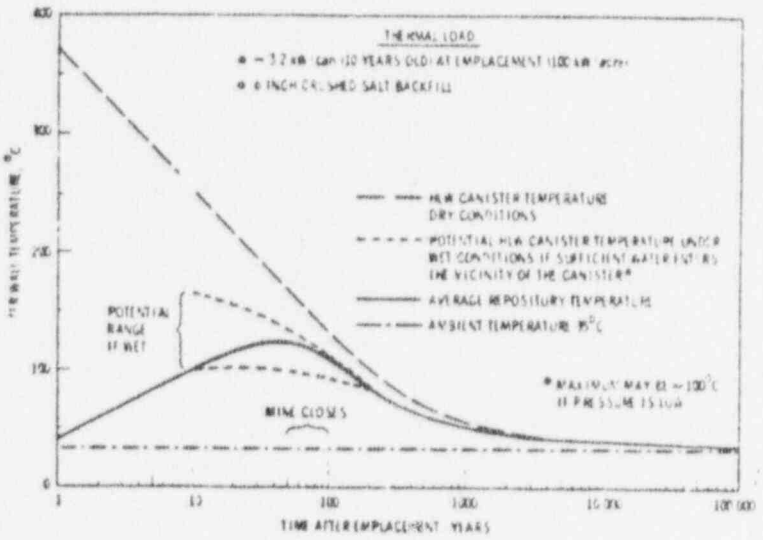


FIGURE 5-10. HLW Canister Temperatures in a Salt Repository (Typical Early Design with High Heat Loading)

is at 140°C to 200°C (Figure 5-11), depending on the host rock. The large temperature rise occurs across the dry backfill material and air gap near the canister. If the salt or rock near the canister becomes wet, the heat transfer near the canister improves; if sufficient water comes in contact with the canister, the temperature may be near that of the 140° to 200°C field temperature (Figure 5-10).

The higher temperatures at the canister wall only exist for a short time (~100 y) in the repository life. If higher temperatures could exist with water present, then the host rock (e.g., granite in Figure 5-8) could also be undergoing attack.(35) Westsik provides data indicating that the relative durability of glass is as good as other materials, such as the geologic host rock, even under extreme conditions.(37) These data do emphasize the need for thorough evaluation of any given disposal location and proper design/operation of the repository.

Many repository designs can be utilized to reduce the temperature near the canister in a repository. As illustrated in Figure 5-11, one important consideration is to improve the heat transfer and thus reduce the excessive temperature rise across the crushed backfill and air gap. Other ways of reducing the maximum canister wall temperature, which occurs shortly after emplacement, are shown in Figure 5-12.(5) The canister shown in Figure 5-10 would have a wall temperature of 150°C in dry salt if it were emplaced after about 45 y. If the waste concentration in the original canister is reduced to one-third the normal content, the wall temperature would be 150°C after only about 10 y. Another factor is the areal loading in the repository. Figure 5-13 shows the rise in repository temperature above the ambient temperature as a function of the quantity of waste heat emplaced in each acre (plan view) of repository.(5,38) For example, an areal loading of 110 kW/acre causes an average rise of 100°C. If the ambient temperature is 35°C, the average salt temperature away from the waste canister is about 135°C (as shown in Figure 5-11). Reducing the areal loading obviously reduces the waste canister temperature.

The waste canister that contains the glass or other solidified form will also provide a barrier to the waste leaving the repository. Although the current Federal Regulation 10 CFR 50, Appendix F, does not require containment by the canister beyond 90 days after receipt at the repository, a typical container will certainly provide some longer-term containment. If a simple stainless-steel canister is used, some of the metal will crack due to chloride in the repository, which causes stress corrosion. However, this should not lead to total canister failure and the vessel should continue to hold the solid waste tightly together and provide a corrosion barrier. Leach rate data on stainless steel, Inconel, and carbon steel show relatively low attack at 250°C salt

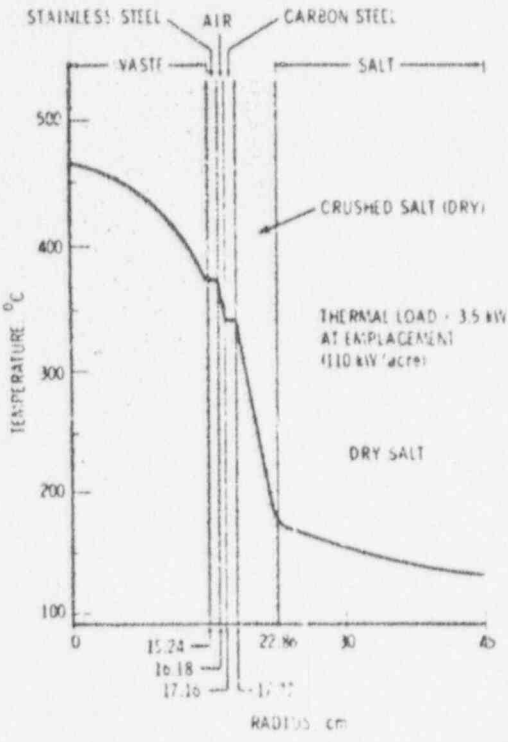


FIGURE 5-11. Typical Profile of Repository Temperatures Near a Waste Canister - An Early Salt Repository Design

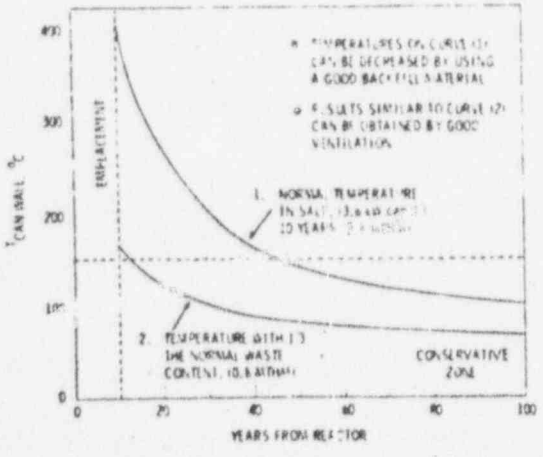


FIGURE 5-12. Reduction of Canister Wall Temperatures by Aging or Dilution of Waste

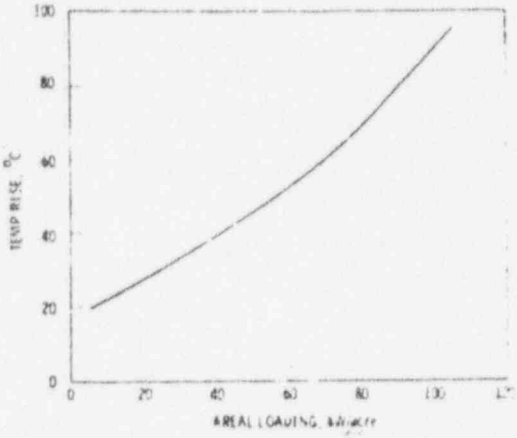


FIGURE 5-13. Average Temperature Rise at Waste Disposal Level

brine.(35) This information suggests a thin canister wall will provide some containment during the first 100 y. As discussed above, it is not likely that a wet high-temperature condition such as this would be present for this length of time.

In the Swedish KBS safety study, a canister is designed to last for 1000 y.(39) Borosilicate glass would be cast in a stainless steel canister during processing. Before the waste is placed in geologic disposal, a 6-in. layer of lead and a titanium outer skin would be applied. In the study, the canister is isolated in a wet granite repository at a temperature of 70°C. (Further details of this study are in Section 8.)

Studies underway for [illegible] are considering limiting the canister wall temperature to 110°C at time of insertion in the repository. High temperature repository conditions will not be present for existing defense and NFS high-level wastes, which are much lower in radioactive decay heat.

Other Solidification Activities

Alternative waste forms that might be used in place of glass are being developed at several laboratories and universities. These materials range from supercalcine at PNL (40) and Penn State University (41) to a cermet at ORNL.(42) The PAMELA process in Germany (23) is probably the most highly developed of these alternative processes. It consists of emplacing phosphate or borosilicate glass beads in a metal matrix.

Many of these alternative processes are being investigated as potential alternatives to glass because conceivably they may offer some improvement for immobilizing high-level wastes. This may result from either an improvement in the inherent physical properties of the waste form or by providing multiple layers of protection. An example of the former is the preference by some for a crystalline waste form. It is possible to modify the liquid waste with appropriate additives so that during processing, certain crystalline compounds are formed. In practice, since more than 30 elements are involved, only a few elements can be forced to crystallize in desired phases. Although direct control of the entire system is not feasible, it is possible, in principle, to establish that all phases formed have acceptable properties. The preferred synthetic mineral compounds would be quite thermally stable and could offer improved resistance to leaching. Examples of this type of waste form are supercalcine (41) and SYNROC.(43)

An example of the multiple layer waste form is the multibarrier waste form shown in Figure 5-14 that is being developed at PNL.(40) The inner core (primary barrier) might consist of a refractory crystalline form, such as supercalcine, or it could be a glass marble. The inner core could be coated with a leach- and oxidation-resistant material (second barrier), cast in a metal matrix (third barrier), and placed in the metal canister (fourth barrier).(3,40) The waste canister could also be designed to provide multiple layers of protection, such as the Swedish waste canister destribed above.

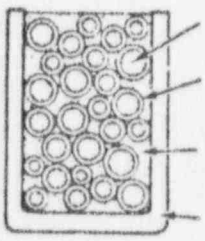


FIGURE 5-14. Multibarrier Waste Form

Another example of the multibarrier concept incorporates the SYNROC material recently conceptualized in Australia.(43) As with supercalcine, the waste elements are incorporated into synthetic mineral compounds, such as perovskite ($CaTiO_3$) and zirconia (ZrO_2). These are thermally-stable compounds that may improve resistance to leaching. In this concept it is proposed that the SYNROC could be encapsulated and transported in Ni$_3$Fe containers. These would be placed in deep holes in granite and surrounded with crushed magnesia and serpentine. Obviously, these multibarrier systems could become rather complex, so a realistic approach would be to only pick those barriers that provide significant benefit while substantially reducing the risk of loss of containment during storage and isolation.

Waste form development has to be undertaken in the context of the total system, including processing, storage, transportation and isolation. Factors that must be addressed include:

- process and waste form compatibility, including temperature and pressure requirements that affect corrosion and volatility
- adaptability to fully remote hot-cell operation
- ease of process scale-up to meet plant throughput requirements
- flexibility of the waste form to adapt to many different waste compositions and to withstand daily variations in the waste stream composition
- thermal and radiation stability of the waste form
- durability, including mechanical stability and solubility in environmental solutions.

With the exception of rapid thermal alteration under some extreme and unlikely repository conditions, glass and vitrification processes have been demonstrated satisfactorily in relation to all of these factors. While other waste forms may offer some improvements in thermal stability, it is not known whether they can satisfactorily accommodate all of the total system requirements. As an example, crystalline waste forms possess numerous phases, each of which has a different crystal structure. The formation and stability of these phases are highly dependent on waste composition control. The argument has been advanced that crystalline forms are preferable because they are more thermodynamically stable than glass.

However, this may be a moot point when all factors are considered:

- The crystals are not all stable to radiation and some will become amorphous (glass-like).
- If glass does crystallize (devitrify), the loss in leach rate performance is very small (less than a factor of 10).
- It is difficult to apply equilibrium thermodynamics to a system of complex structures whose overall composition is made of more than 30 components and which is subjected to an intense radiation field.

Multiple layers of protection can provide an improved waste form. Obviously, metal matrices would increase mechanical strength and improve thermal characteristics.(44) However, in general, these new alternatives require about the same processing temperature as current low-temperature glass (1050° to 1150°C) and will, in many cases, require more production steps. Also, data such as that shown in Figure 5-15 do not indicate that the leach rate performance improvements will be substantial. Figure 5-10 is taken from Jardine and Steindler (44) and is representative of 25 to 100°C conditions. Data on supercalcine has been added for comparison.(45) Recent data under severe hydrothermal conditions, 350°C and 135 atm in salt brine, indicate that leach rates for supercalcine are comparable to those for glass.(37) Therefore, selection between alternative solidification schemes may be dominated by the choice of the simplest process. However, it is wise to continue pursuing alternative technologies so that a suitable backup system is available and to continue to look for a system that may offer substantial improvements.

This philosophy was supported at a recent workshop where ceramic and glass radioactive waste forms were discussed. There was general consensus that the information available on glass provided a good basis to establish glass as a satisfactory waste form and that crystalline ceramics show promise as good waste forms if much more developmental work is completed.(46) A recent report by

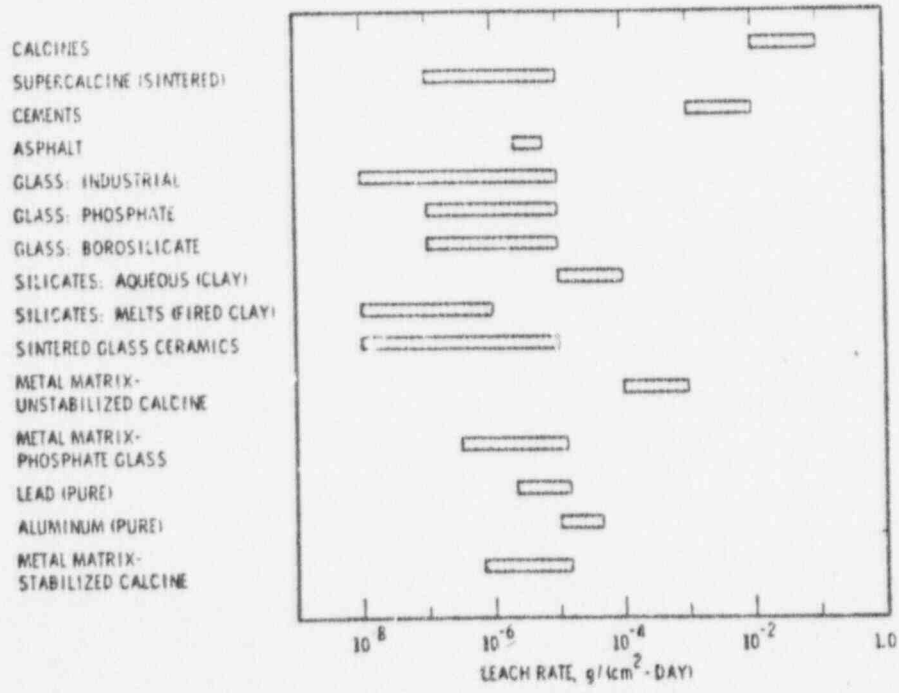


FIGURE 5-15. A Comparison of Leach Rates for Various Waste Forms

Mende. (25) describes the use of borosilicate glass as a satisfactory waste form for isolating high-level radioactive wastes.

Considerable development is required before decisions can be made on the suitability of these waste forms and whether they offer a substantial improvement over currently available technology.

SPENT-FUEL TREATMENT

If unreprocessed spent fuels are to be temporarily stored (retrievably) in a geologic formation and eventually isolated there, appropriate spent fuel packages are needed. The spent fuel packaging program (47) in the U.S. is being managed for the DOE by the Battelle Office of Nuclear Waste Isolation. The baseline option is placement of spent fuel in canisters with only an inert gas (e.g., helium) fill. Other options being studied include filling the canister with a metal matrix, sand, or other glassy or ceramic materials, as well as encapsulating the spent fuel and canister when the fuel is declared a waste for disposal. Initially the sensitivity of the waste form to the geology for containment in a repository will be evaluated. Also, sufficient technical information is being accumulated on the packaging and encapsulating options to permit an assessment of the alternatives by 1979-1980. The study will consider whether any waste form other than the canistered spent fuel is desirable as a package for disposal of spent fuel, as opposed to retrievable storage which must keep open the options for reprocessing, continued storage, or disposal.

Experimental packaging and storage of spent fuel is planned in 1978, using facilities previously associated with the nuclear rocket program in Nevada. Several package. will be examined by Westinghouse-Nevada in the Engine Maintenance and Disassembly (EMAD) facility.(2)

A comprehensive study on packaging spent fuel was conducted by the Swedish project, Kaern-Braen-Slesaekerhit (KBS), set up in early 1977.(39) The containment proposed is a copper canister that will hold 500 fuel rods. After the fuel rods are placed in the canister, it will be filled with lead, and a copper lid will be welded in place. The final storage/disposal location proposed is in granite some 500 m below ground. Canisters will be emplaced in oversized holes completely lined with isostatically-compressed bentonite and quartz sand, as shown in Figure 5-16.(5)

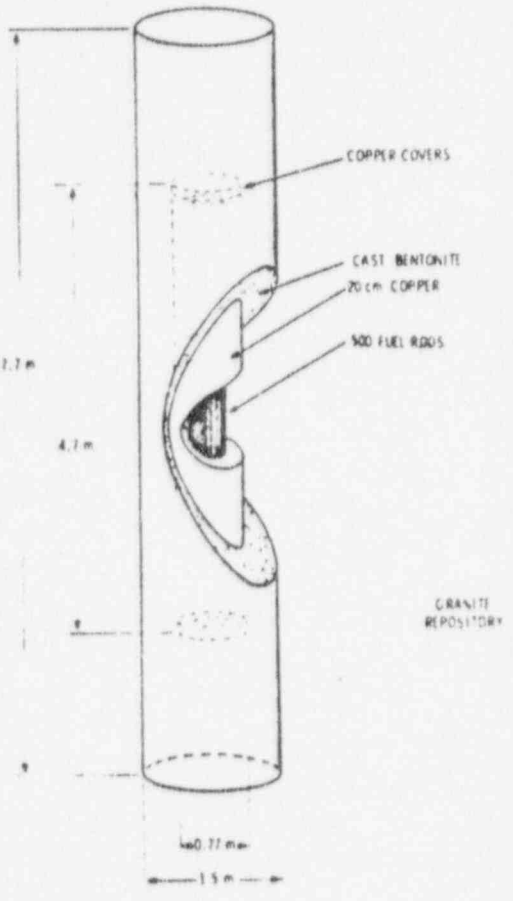


FIGURE 5-16. KBS Spent Fuel Isolation Concept

Granite formations investigated in the KBS project showed potential for small water flows (0.1 to 0.2 $\ell/m^2/y$) and this in part accounts for the sophisticated packaging and emplacement. Disposal formations essentially free of water are being sought in the U.S. and less sophisticated packaging may suffice.

The spent fuel packaging program is just getting underway in the U.S. However, data is already available on the leaching of irradiated fuel pellets in various solutions (48) and good progress is being made on storage of fuel and potential packaging concepts.(2,47) Furthermore, work

done to date on packaging HLW for isolation is directly applicat'e to spent fuel. Thus, there should be adequate time to develop packaging concepts since ultimate disposal of spent fuels is not needed for 25 to 30 y, and retrievable storage in a geologic formation is not scheduled until after 1985 even if the decision is made to test such storage in the Waste Isolation Pilot Plant (WIPP) in New Mexico.(2)

Section 6

OTHER FUEL CYCLE WASTES

Prior sections in this report have dealt primarily with HLW and spent fuels. This section discusses other fuel cycle wastes: cladding and hardware wastes, gaseous wastes, and other low-level wastes, both TRU and nonTRU. Mining and milling wastes will not be covered; techniques for stabilizing these wastes in place are being developed. Also, wastes derived from decontaminating and decommissioning wornout or obsolete nuclear facilities will not be covered. However, technology being developed for volume reduction, decontamination, and elimination of combustibles may also be applicable to these wastes.

CLADDING AND HARDWARE

In the first step of reprocessing spent nuclear fuels, end fittings are removed mechanically, and the fuel rods are sheared into small pieces (1 to 5 cm). The sheared fuel is then exposed to a dissolvent (HNO_3) which dissolves the core material. A residue of cladding and internal fittings is left, called cladding and hardware, or "hulls." This sequence is shown in Figure 6-1.

The cladding and hardware waste is radioactive due partly to neutron activation products generated in the metals and partly to a small amount of fuel material remaining on the inner surface of the cladding. Because of the amount of transuranic elements present, it must be considered a TRU waste.

Light-water reactor fuels are generally clad in zirconium alloy (Zircaloy) tubes which have stainless steel or Inconel end fittings and spacers. Figure 6-2 shows a sample of Zircaloy cladding residue.

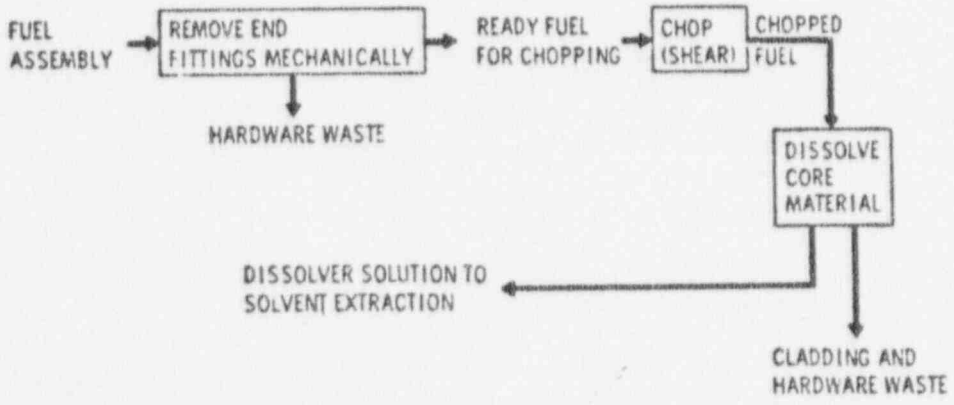


FIGURE 6-1. Source of Cladding and Hardware Waste

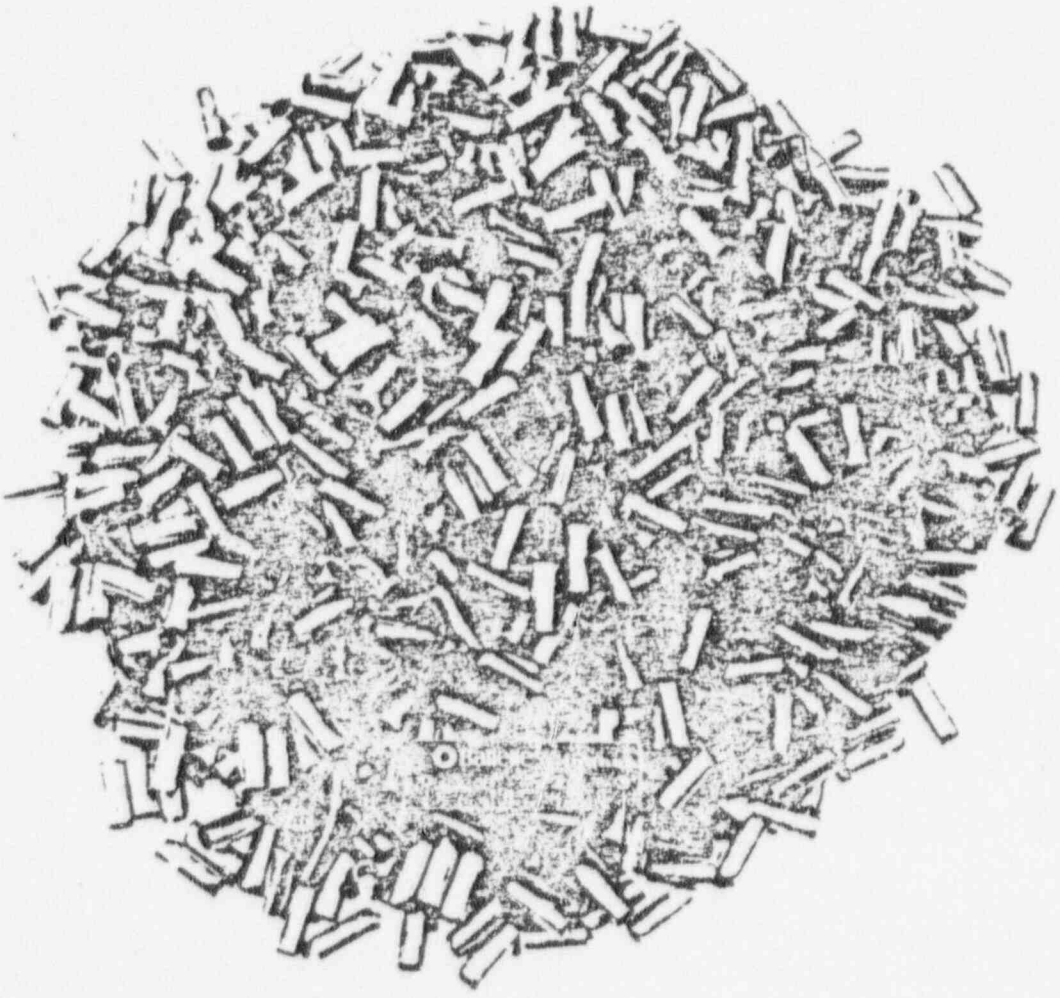


FIGURE 6-2. Zircaloy Cladding Residue

Typically, the residue weighs about 325 kg/tU for an LWR reference fuel.(49) Without compaction, it has a volume of about 0.3 m^3/tU (~10 m^3/GWe-y). Most of the material is Zircaloy (70 to 80%), 12 to 22% is stainless steel, and about 8% is Inconel. Principal activation products present at 5 y out of the reactor are ^{60}Co, ^{55}Fe, ^{125}Sb, ^{63}Ni, and ^{125m}Te. Their combined activity is about 900 nCi/g Zr. Zircaloy-2 and Zircaloy-4, the zirconium alloys generally used in LWRs, contain some tramp uranium (1 to 3 ppm). Neutron irradiation of this uranium produces alpha-emitting activation products. For typical irradiation levels, total alpha activity from this source is, at 5 y out of reactor, about 90 nCi/g Zr (85 nCi ^{241}Pu, 2 nCi ^{244}Cm, and 3 nCi, others). These alpha emitters are distributed throughout the metal and, consequently, cannot be removed by any surface decontamination technique. The cladding is estimated to contain 30 to 60% of the tritium generated during irradiation.

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Irradiated cladding has an external corrosion product layer up to 50 μm thick and an internal oxide layer a few μm thick. Following HNO_3 dissolution of the core material about 0.1% of the core material remains associated with the cladding primarily in the internal oxide layer.

Zirconium and Zircaloy alloys can be pyrophoric if finely divided.(50) This property of the cladding must be considered in any proposed treatment, storage, or disposal of the waste.

To date, only a small amount of cladding and hardware wastes has been produced from commercial fuel reprocessing in the U.S. These wastes were simply dried, placed in steel barrels, and buried. Defense fuel reprocessing in the U.S. has involved chemical dissolution of Zircaloy cladding and subsequent storage and treatment of the aqueous waste. No solid metallic waste is produced.

Efforts toward improvements in the treatment and disposal of cladding and hardware waste have had four major objectives:

- providing a barrier to dispersal of the radioactive constituents
- reducing the volume
- reducing the potential for zirconium fires
- possibly reusing the constituents.

Many approaches have been proposed and studied for achieving these objectives. These approaches include sorting out the different metals for separate treatment, mechanical compaction, chemical and electrochemical decontamination, casting the metals as ingots, putting the waste in an inert matrix, chemical dissolution, burning Zircaloy to oxides, and volatilizing zirconium as the chloride. The Technical Alternatives Document (TAD) presents an excellent survey of these proposed schemes and the status of the technology involved.(51) Most of the technologies are in the small pilot-plant, laboratory, or conceptual stage and require further development. The most highly developed technology casts the waste as metal ingots.(49)

Unless spent fuels are reprocessed, technology is not needed for treatment and disposal of the cladding and hardware waste. This is the current situation in the U.S.(52) A PNL developmental program decontaminates Zircaloy and casts the cladding and hardware metals into ingots for disposal or possible reuse.(49) In this study, essentially complete decontamination of Zircaloy cladding (except for internal activation products) was obtained by treating the cladding with gaseous HF at approximately 600°C and then leaching it with an aqueous solution.(53) The metal can be melted and cast as an ingot.

Foreign R&D on cladding and hardware waste includes:(6)

France	• meltdown of Zircaloy cladding for consolidation and decontamination
Germany	• immobilization of hulls in concrete
India	• interim storage in cylindrical tiled holes in the ground
United Kingdom	• compaction and encapsulation of stainless steel and Zircaloy hulls.

Research being coordinated by the Commission of European Countries includes:(6)

Belgium	• incorporation in low-melting alloys
Germany	• incorporation in concrete
France	• decontamination and conditioning in glass
United Kingdom	• characterization of radioactivity in different cladding wastes.

OTHER LOW-LEVEL SOLID WASTES

Low-level solid wastes are generated in all phases of the fuel cycle, as well as in R&D activities. They include cleanup materials, failed equipment, containers, and contaminated clothing. These wastes encompass a wide range of materials, such as paper, rags, glass, plastics, wood, metals, and soils. They occupy a large volume, as shown by the data in Section 2. Some are TRU wastes and some are not. Under the U.S. reference waste management system, the non-TRU wastes may be disposed of in near-surface burial grounds, but the TRU wastes from commercial operations will be disposed of in a federal repository.

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Good management practices can help to minimize the volume of these wastes. This has been shown whenever a concerted effort to do so has been carried out.

Research and development efforts toward improved management of these wastes have much the same objectives as those for managing cladding and hardware wastes. Emphasis has been on the treatment of TRU wastes since these wastes represent a long-term risk to the biosphere and their disposal in a repository will be relatively expensive.

Solid wastes can be divided roughly into combustible and noncombustible materials. Some of the techniques for treating these wastes are:

1) Combustible Solid Wastes

Mechanical compaction

Incineration or oxidation in HNO_3-H_2SO_4

Fixation of residue in a matrix

Recovery of actinides from incinerator ashes

2) Noncombustible Solid Waste

Size reduction

Mechanical compaction

Melting of metals

Decontamination of equipment for reuse

Fixation in a matrix.

The TAD presents a good summary of past accomplishments in all of these areas.(51)

Incineration of combustible nuclear wastes has already been applied on a plant and pilot scale at numerous sites, both U.S. and foreign.(51) Currently, the DOE supports developmental studies on a fluid bed incinerator at Rocky Flats, (54) a controlled air incinerator at LASL, (55) and an HNO_3-H_2SO_4 digestor for oxidation of organic materials at HEDL.(56)

Excellent decontamination of metal surfaces by electropolishing was accomplished in PNL studies.(57) Other studies on immobilization of incinerator ashes and other TRU wastes in glass, concrete, ceramic, and cermet forms are in progress. The U.S. program is designed to demonstrate the required systems for processing, packaging, shipping, and isolating alpha-bearing wastes by the mid 1980s.(52)

GASEOUS WASTES

Certain fission products and neutron activation products generated during irradiation of nuclear fuels are inert gases or become gases during reprocessing operations. Those of most concern in waste management are ^{85}Kr, ^{129}I, ^{14}C, and ^{3}H (tritium). Data on these four isotopes are shown in Table 6-1.

So far, these nuclides, with the exception of ^{129}I, have been discharged to the atmosphere. Discharge limits effective in 1983 have been established by the U.S. EPA for ^{85}Kr (5×10^4 Ci/GWe/y) and for ^{129}I (5 mCi/GWe/y).(58) These correspond to decontamination factors of about 8 and 240

TABLE 6-1. Gaseous Isotopes of Concern in Waste Management(a)

	Half Life, Years	Source	Quantity per GWe-y (LWR)	Where Released	Chemical Form Discharged
^{85}Kr	10.76	Fission	3.3×10^5 Ci	Fuel dissolution	Kr
^{129}I	1.6×10^7	Fission	1.2 Ci	Fuel dissolution	I_2 or organic iodides
^{14}C	5730	Activation of C, N, and O	9-49 (fuel only) Ci	Reprocessing plant, some at reactor	CO_2 or carbonates at reactor
^{3}H	12.3	Ternary fission, some activation of impurities	$2-4 \times 10^4$ Ci	Reactor and reprocessing plant	HTO or HT

(a) Material taken from Reference 58.

for ^{85}Kr and ^{129}I, respectively, as compared to complete release. Discharge limits for ^{3}H and ^{14}C have not yet been set in the U.S.

Waste management R&D on these isotopes can be divided into two phases: 1) recovery of the isotope, and 2) conversion to a form suitable for storage and disposal. The status of waste management R&D for these isotopes is summarized below.

^{85}Kr

A plant for recovering ^{85}Kr from dissolver off gas by cryogenic distillation is in operation at INEL.(59) Other such plants are being developed in France, West Germany, Japan, and Belgium. A plant based on liquid absorption is in cold operation at Oak Ridge.(60) Long-term hot demonstration of the process is needed.

Storage forms investigated for Kr include: 1) containment in pressurized cylinders, and 2) incorporation in inorganic materials, such as glass, ceramics, zeolites, and metals.(61) Concern over long-term containment in pressurized cylinders prompts this second group of studies. To date, these technologies are still in the laboratory stage.

^{129}I

Techology exists for the removal of iodine from gas streams (61) by aqueous scrubbing ($NaOH$, $Hg(NO_3)_2$-HNO_3, ~20 M HNO_3), and by chromatographic sorption on solids (charcoal, silver-impregnated solids, and metal zeolites). Present R&D efforts are directed toward converting the iodine to forms adequate for long-term containment in geologic isolation (the half-life of ^{129}I is 1.6×10^7 y). The total quantity of iodine present in spent LWR fuels is relatively large (~0.25 kg/t), and the cost of expensive metals, such as Hg and Ag, required for recovery of the iodine is significant. Thus, there is an incentive to recover these metals for reuse. Research and development on storage/isolation forms of iodine are in the laboratory stage.

^{3}H

For stainless-steel-clad fuels, most of the tritium will have escaped through the cladding during reactor operation. Of that remaining, most (99%) will be released to the dissolver solution as HTO in the reprocessing operation. For Zircaloy-clad fuels, much of the tritium is contained in the cladding and is not released during dissolution. That which does enter into the dissolver solution may be released as aqueous effluents or as water vapor.

There are three main options for tritium management:

- Tritium can be evolved and recovered with an aqueous solvent before the solvent contacts fuel. The voloxidation process developed at Oak Ridge has given tritium releases >90% during heating of chopped fuel (450° to 500°C for four to six hours).(62)
- Through aqueous recycle in the plant, tritium can be removed and solidified in a small side stream. However, there may be significant in-plant exposure problems with this approach.
- Isotopes can be enriched and the HTO can be collected from the liquid stream.

Tritium as HTO can be incorporated in concrete but migrates out rapidly if moisture is present. A combination concrete-polymer material recently demonstrated in the U.S. shows promise for providing the necessary containment.(63) Collection of HTO on drying agents (e.g., silica gel, activated alumina, or molecular sieves) followed by encapsulation to prevent HTO release may produce an adequately stable form. Formation of metal hydrides, such as zirconium hydride, may be applicable. These processes are all in the laboratory or demonstration stage.

^{14}C

It is not clear at this time that collection and storage of ^{14}C will ever be necessary. If it should become necessary to control ^{14}C emission, existing technology for removal of CO_2 from gases could probably be applied. Because of the long half life of ^{14}C, geologic isolation is needed. Such isolation may be effected as a part of krypton isolation processes. One of several relatively insoluble carbonates would probably be an adequate disposal form.

Section 7

TRANSPORTATION CONSIDERATIONS

Any discussion of nuclear waste management must consider the problems of transporting nuclear materials. Transportation is a necessary link in all aspects of the nuclear fuel cycle and will become increasingly critical in an expanding nuclear economy. It is especially critical with highly radioactive materials.

HARDWARE

At present, TRU wastes are shipped only in DOE owned railcars which are neither licensed nor available to industry.(2) There are only about ten railcars and it would take decades using these cars to transport the defense TRU wastes expected by 1985 to the WIPP facility.

Spent fuels are transported, either by truck or by railcar, in specially-designed casks. The ability of such casks to withstand credible transportation accidents without breaching and loss of contents has been a point of concern. Recently the Sandia Laboratories have conducted full-scale crash tests on shipping casks.(64,65) Truck- and railcar-mounted casks have been crashed into virtually immovable concrete targets at speeds up to 136 km/h (84 mi/h) with only superficial damage to the cask and no rupture of contained fuel elements. These tests are reassuring.

Canisters of high-level wastes would be transported in casks similar to those used for spent fuels. One conceptual design, shown in Figure 7-1, proposes a railcar-mounted cask for waste canisters.(66)

The cask will hold nine waste canisters 30 cm (~1 ft) in diameter with a total waste volume of 1.58 m^3. Based on data from Pratt et al.,(5) this volume of HLW would correspond to about 0.36 GWe-y from an LWR fuel cycle. No shipping cask specifically designed for HLW canisters has yet been built. If spent fuels are never reprocessed, shipping casks for commercial wastes will not be needed. Casks for shipping defense HLW could differ considerably in design because of the low specific decay heat content of the waste.

The DOE Task Force for Review of Nuclear Waste Management(2) expresses concern about the availability of equipment for transporting HLW and TRU wastes. The current commercial cask inventory can transport only about one-third of the spent fuel output from currently operating reactors. Recently industry in the U.S. and abroad (67) has committed to build more casks. Currently, there are no special requirements for transporting low-level wastes.

REGULATIONS

In the past, nuclear materials have been moved around the country with relative ease and with only a few incidents, all of which have been minor. The prospects for greatly increased movements of these materials requires good communications to assure public acceptance. Transportation of nuclear materials in the U.S. is subject to regulations issued by the Department of Transportation (68) and the Nuclear Regulatory Commission.(69) Similar regulations developed by the IAEA are in use throughout the world.(70)

SECURITY

The present NRC, DOT, and DOE position is that guards are not needed during transport of spent fuels and TRU wastes. With adequate administrative controls, guards should be needed even less during transport of HLW. Risk analyses of transporting nuclear materials generally conclude that the risk to transportation personnel as well as to the public, is very small.(71-73)

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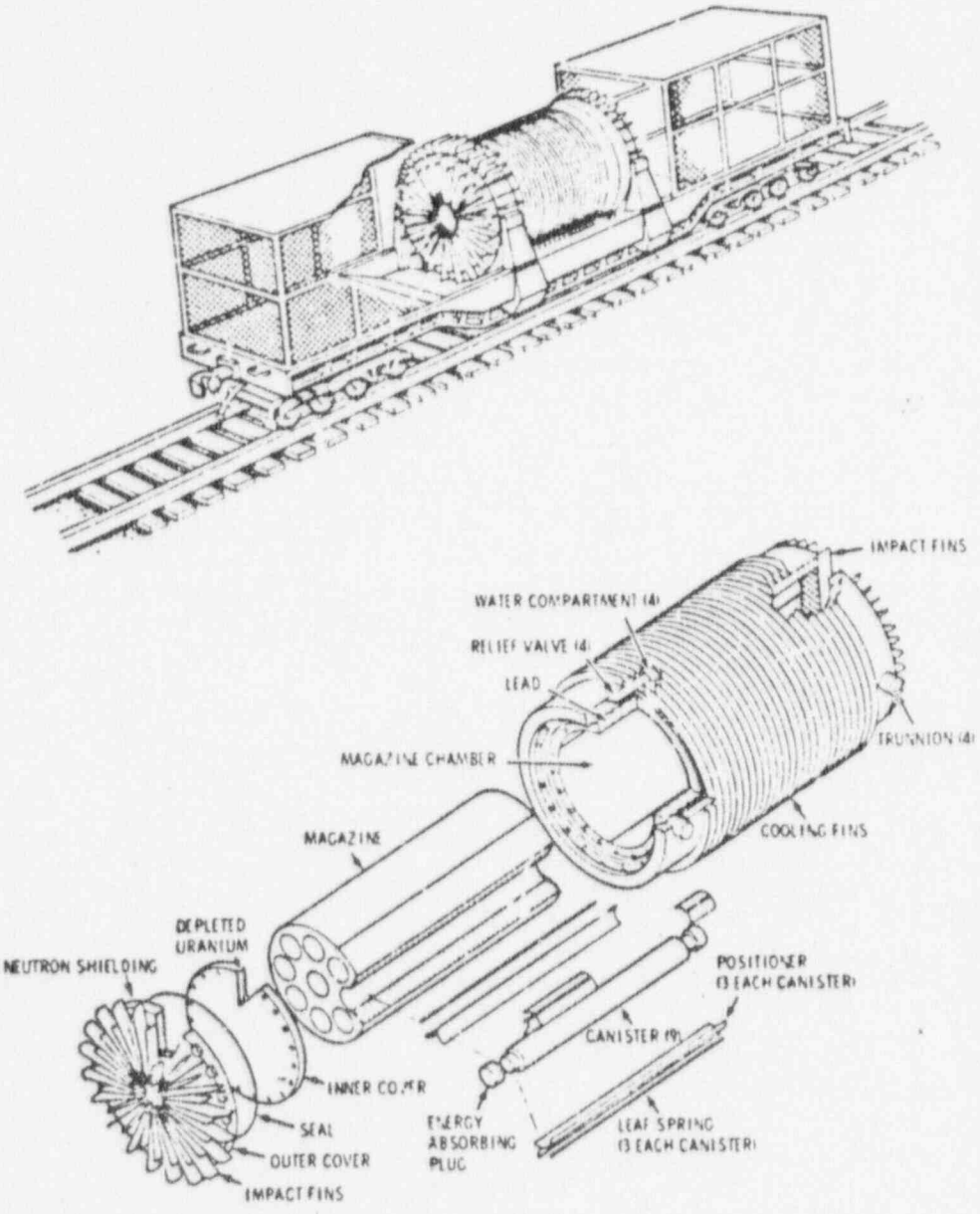

FIGURE 7-1. Conceptual Solidified High-Level Waste Shipping Cask and Railcar

However informative and reassuring these studies may be to the authors and their technical peers, the results must be available publicly in lay terminology if widespread transportation of nuclear materials is to be accepted by the public. A concerted public education effort is needed.

Section 8

WASTE REPOSITORY

Although various waste disposal alternatives are being studied in the U.S., current emphasis is on land (geologic) disposal. Several criteria have contributed to adopting geologic disposal in the reference U.S. nuclear waste management system:

- most available technology
- less transportation
- fewer emplacement problems
- most certainty as to long-term stability
- fewer problems with international agreements.

GEOLOGIC ISOLATION

Current activities in geologic site selection in the U.S. emphasize the use of salt beds; however, other deep, potentially dry formations, such as granite, shale, and basalt, are also being considered. A waste-isolation pilot plant (WIPP), intended as a final disposal site for defense TRU wastes and as a place for conducting R&D with other waste materials in salt, is scheduled for operation in 1985. A proposed site near Carlsbad, NM, has been chosen, and data are being collected for the necessary prelicensing documentation. A conceptual drawing of the WIPP facility is shown in Figure 8-1.(74) The facility will have two storage levels.(75) The upper level, ~2100 ft below the surface, will be for contact-handled TRU waste. The lower level, ~2600 ft below the surface will be used for remotely handled waste and HLW experiments. A DOE nuclear waste management task force recommends that capability for doing R&D on disposal of defense HLW and on a limited amount of unreprocessed commercial fuel be included in the design.(2)

Earlier emphasis in the U.S. on disposal of nuclear wastes was also on the use of salt beds because:

- By nature, salt beds are dry.
- They are considered geologically stable.
- Because salt creeps readily under pressure, cracks in the salt should tend to be self-healing. Extensive testing on a nonradioactive basis was done in a salt dome near Lyons, KS.(76)

Any permanent disposal of nuclear wastes must ensure that no event or combination of events, man-made or naturally occurring, can return the wastes to the biosphere in amounts great enough to cause a significant risk to the biosphere. Furthermore, the procedures of putting the wastes in appropriate forms for disposal, transporting them to the disposal site, and emplacing them in disposal must entail an acceptable risk to the biosphere.

Responsibility for the DOE program for isolation of commercial wastes has been recently shifted to Battelle Memorial Institute. The Office of Nuclear Waste Isolation (ONWI) at Battelle will most likely recommend a salt location for the first commercial repository. The DOE Task Force concluded that the earliest possible date for this candidate site selection is 1979 and for repository operation, 1988.(2) The ONWI program will also evaluate potential sites in basalt, shale, and granite.

The DOE site at Hanford rests on deep beds of basalt that are thought to be relatively dry and geologically stable. Characterization of the geology of the site and the basalt beds is in progress in a study conducted by Rockwell Hanford

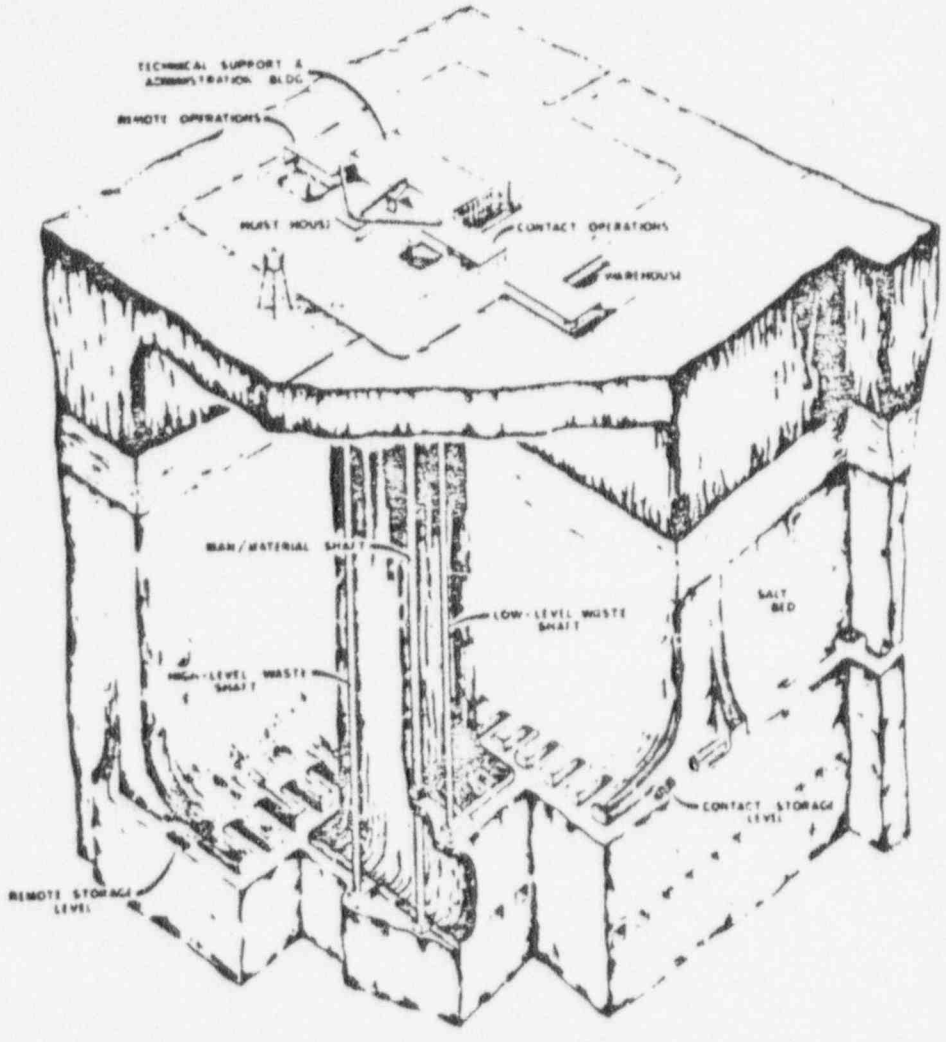

FIGURE 8-1. Waste Isolation Pilot Plant in New Mexico

Operations.(77) Research and development on the disposal of HLW and unreprocessed spent fuel in a basalt Near-Surface Test Facility (NSTF) is also underway.(77) Excavation of the NSTF began in July 1978.(78) The three tunnels shown in Figure 8-2 will be mined simultaneously in Gable Mountain at Hanford. Electric heater tests are scheduled to begin there in August 1979.

Work is in progress to evaluate the potential for nuclear waste disposal at the DOE Nevada Test Site (NTS). The principal contractor for the NTS studies is Sandia Laboratories. A major objective is to determine if disposal of nuclear wastes at the site would be compatible with predicted efforts of continued weapons testing.

A detailed study of the disposal of nuclear wastes in granite formations is in progress in Sweden. Tests using electric heaters to simulate waste are underway in the Stripa mine in Sweden. The U.S. is participating in these tests through joint funding with Sweden.

Tests of the behavior of domed salt formations under heat loading will be conducted in the Avery Island salt mine in

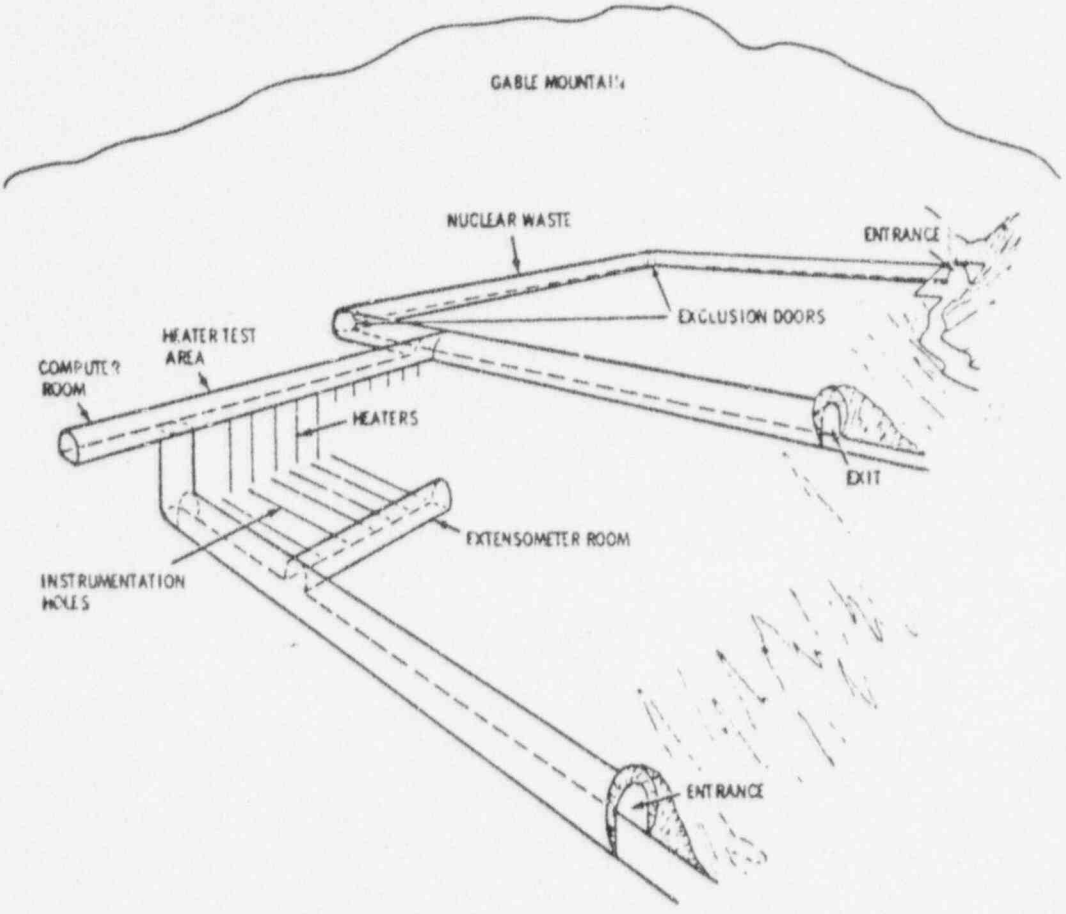

FIGURE 8-2. The Near Surface Test Facility at Hanford

Louisiana by RE/SPEC, Inc., and Union Carbide Nuclear Division.

Thermochemical and mineralogical changes in heated argillaceous rocks are being observed in a test underway in Conasuaga Shale at Oak Ridge. The principal contractor is Sandia Laboratories.

The physical operations of emplacing nuclear wastes in a deep geologic formation present no problems new to the mining and nuclear industries. The basic concept involves constructing vertical shafts to the emplacement depth, excavating lateral tunnels to rooms where the canisters are to be emplaced, and boring the actual emplacement holes.(79) Special equipment is needed to transfer waste canisters from the transportation vehicle to mine-handling equipment at the surface, lower the canisters to the emplacement level, transport them to the emplacement site, and emplace them. Adequate shielding, decontamination equipment, and air control must be provided so that operators and the environment are protected from contamination. All of these safeguards are familiar to the mining and nuclear industry.

However, the fact that the wastes to be isolated contain radioactive materials that emit decay heat and represent risk to the biosphere for many thousands of years presents some new factors to be considered in the selection, design, and operation of a geologic disposal site. Some of these considerations are listed below:

- The site must remain intact and undisturbed for many years. No mining activities can be permitted in the vicinity.

- Geologic isolation in the U.S. has been previously based on the premise that the disposal site will be essentially free of water. Given a dry formation initially, water must be excluded during mining, emplacement, sealing operations and subsequent storage.

- Decay heat emitted by the wastes (see Figure 5-9) will heat the surrounding host material, causing expansion and possible chemical reactions, especially if high-temperature water is present. Excessive expansion can cause the host material to fracture, potentially allowing water to enter the repository from the surface or from underground aquifers.

- Intense radiation may produce significant physical and chemical changes in the wastes and surrounding media.

- If water enters the repository, corrodes the waste containers, and comes in contact with the waste, the rate at which the waste is leached will increase with increasing temperature of the waste-water interface. This becomes much less of a problem as the fission product heat decays.

- Geologic isolation is based on the premise that no phenomenon, man-caused or natural, will occur that will expose the wastes to fluids (ground water) that will carry them to the biosphere. But, since repository integrity and the exclusion of water for hundreds of thousands of years cannot be guaranteed, multiple barriers to the dispersion of the waste may be built into the system if necessary. For example, the HLW is converted to a relatively insoluble form, and the waste may be encased in one or more metal containers which corrode very slowly in the environment. Ion exchange materials, such as bentonite clays, may be packed around the waste to absorb waste elements from aqueous solution and retain them. The repository may be located where entering water must pass through a long path of absorbing materials before it can reach the biosphere.

- When waste emplacement in a terminal repository is completed, it must be backfilled and the shafts and tunnels leading to it must be plugged. How the backfilling is done will affect the thermal regime, as well as the expansion and subsidence in the repository. How the shafts are plugged may influence the long-term isolation of the repository.

The reader is referred back to Section 5 and Figures 5-9 through 5-13 where some of these repository designs issued are addressed. All of the above concerns affect the design of a repository for long-term isolation of the radioactive waste. To a large extent, the above concerns can be minimized if a conservative design concept is chosen. Many of the earlier design efforts for commercial waste repositories in the U.S. have been directed toward relatively high thermal loadings in waste repositories.(36,80) This high temperature has caused some concerns.(81) With lower temperatures, there would be less concern about predicting the long-term behavior of the geology. In addition, if the temperature in the repository at the rock-canister interface was at or below 100°C, the rate of attack on the waste package would be minimal. If the repository design allows temperatures of 300°C, then even though entering water would reduce the temperature, it may still be hot enough to rapidly attack both the waste and the host rock near the canister.

The DOE Task Force(2) states that the plan for geologic waste isolation and its implementation should emphasize technical conservatism. The overall system proposed in the Swedish KBS safety study is an example of conservative design and multibarrier protection directed toward minimizing the risk of system failure.(39) Key features of the system are listed in Table 8-1. The Swedish repository scheme does not have nearly as many concerns as were addressed above. This concept is shown in Figure 8-3.

It is likely that repositories can be designed, constructed, and safely operated without nearly as much conservatism as employed in the Swedish system. However, without some degree of conservatism, such as either the use of multiple barriers or lower temperatures, it is questionable that one could prove that a repository system is absolutely safe. In actual practice there will be an ipso facto temperature limit since the first wastes to be placed in geologic repositories will be low-heat

TABLE 8-1. Proposed Swedish Waste Isolation Scheme

Step	Feature
Reprocess the spent fuel 2 to 10 y after removal from the reactor.	The 10 y period decreases fission product heating and radioactivity.
Vitrify the HLW in borosilicate glass in stainless steel canisters with a 9% fission product loading in the glass. Store until 10 y out of reactor.	Borosilicate glass is stable and relatively insoluble. The 9% loading further decreases the thermal loading in the canister. The stainless canister is part of the vitrification process and provides adequate containment for interim storage.
Store the canister of HLW glass for 30 y in near-surface repositories.	This provides further decay of the fission product heat and decreases the radioactivity.
Place 10 cm of lead around the stainless steel HLW can and cover the lead with 6 mm of titanium.	The titanium provides a 1000-y barrier in the repository conditions. The lead shielding minimizes hydrolysis of water which could accelerate corrosion of the titanium.
Place the canister in a bentonite-quartz barrier within the granite repository at a depth of 500 m.	The bentonite is an ion exchange material that swells when wet to provide low water permeability.
	The granite repository will have water present, but the thermal loading is such that temperatures will not exceed 70°C, which should cause little, if any, change in the geology.

defense wastes and long-cooled commercial wastes.(25) Early operative experience would then provide a basis for future design.

In addition to studies noted above, several studies are in progress to provide the data needed for repository design.(82) The Office of Nuclear Waste Isolation is directing studies at many of the national laboratories and at several universities:

- heat transfer and thermal analysis programs to predict time-temperature distributions encountered in geological disposal of thermal radioactive wastes
- waste-rock interaction studies to characterize the chemical, physical-chemical, geochemical, and radiochemical reactions between emplaced radioactive waste and surrounding media
- rock-mechanics studies to predict structural behavior of a repository during construction and operation and long-term emplacement of nuclear wastes
- bore-hole plugging studies, including developing special cements, devising techniques to accurately measure properties of the cements, and verifying the techniques by field testing
- waste isolation safety assessment, to develop the methodology required for the assessment of long-term safety of geological disposal.

The other repository programs at Sandia, Rockwell, and Nevada Operations Office have similar studies underway. A discussion of the OKLO natural reactor and its relevance to geologic isolation of radioactive waste is presented in Appendix C.

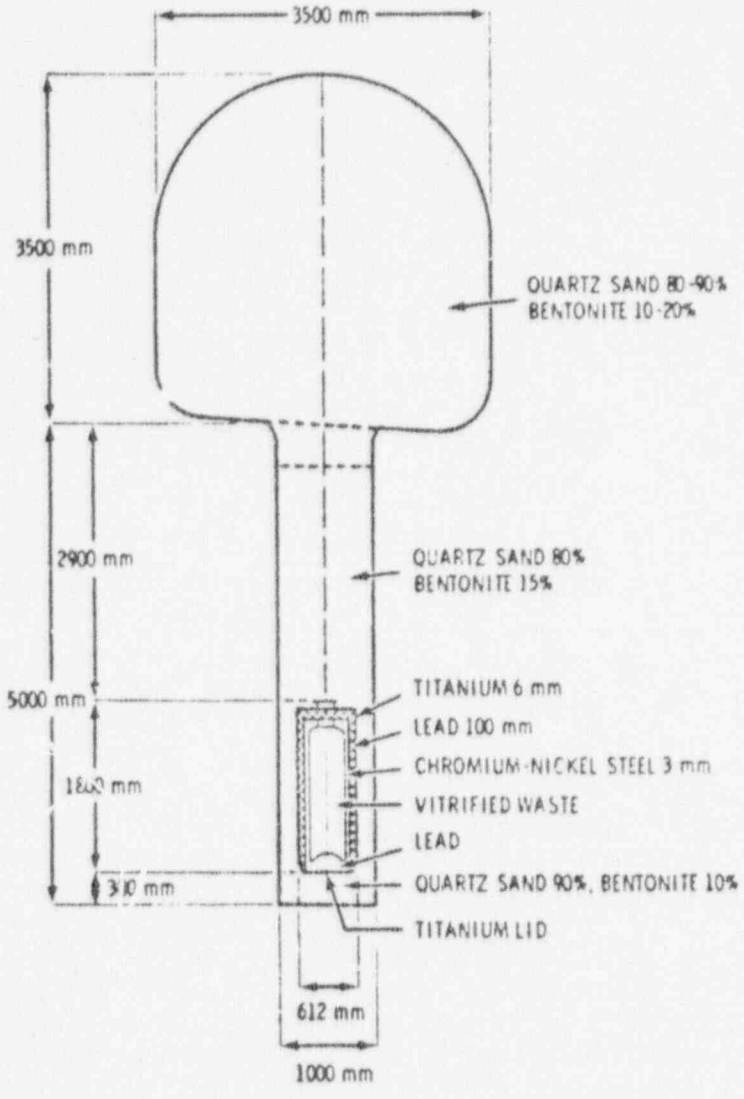

FIGURE 8-3. The Swedish KBS Waste Disposal Concept - A Cross Section in the Granite Repository

OTHER DISPOSAL ALTERNATIVES

Several alternatives to land (geology) have been considered for the ultimate disposal (isolation) of nuclear wastes. These include the following:

- Seabed--stable deep seafloor (abysmal plains), subduction zones, deep trenches, and rapid sedimentation areas (e.g., deltas)
- Ice sheets--the Antarctic ice sheet
- Extraterrestrial space--space orbits designed to ensure that the wastes can never reenter the earth's atmosphere.

A special case of nuclear waste management involves separating (partitioning) the long-lived alpha-emitting elements (TRU elements) and returning them to nuclear reactors where they are converted (transmuted) to shorter-lived or inert isotopes. The residual, shorter-lived waste might then be disposed of under less rigorous conditions.

A detailed study was made in the U.S. of these various disposal alternatives.(83) Site locations, appropriate waste forms, modes of transportation and emplacement, risk to the biosphere, and preliminary cost estimates were considered in the study. The TAD also addresses many of these alternatives.(79)

Section 9

WASTE DISPOSAL REGULATIONS AND CRITERIA

Establishing and implementing a nuclear waste management system involves: defining alternative approaches, risks, and costs; establishing criteria and regulations; and obtaining public acceptance. This section will discuss progress in these areas.

CRITERIA, REGULATIONS, LICENSING AND OPERATION

In the efforts to meet all the requirements for safe and reliable disposal, one key element is the performance criteria which are used to evaluate the systems. These criteria are required so that "safe" is safe enough. Although these criteria are not yet established, it appears they will be finalized in the near future. Both the EPA and the NRC have important roles in providing or regulating criteria.

The EPA has responsibility for setting radiation protection standards for radioactive wastes with: 1) broad responsibility to provide Federal radiation guidance for all radiation directly or indirectly affecting health, and 2) specific responsibility for setting generally applicable environmental standards outside the boundaries of sites which possess radioactive materials subject to regulation under the Atomic Energy Act. The EPA standards are neither method nor site specific. The NRC will regulate waste management operations, assuring that EPA standards are met. The DOE is responsible for developing the waste management technology and for operating HLW sites in conformance with NRC regulations. Thus, the three agencies can be differentiated by their responsibilities:(84)

- The EPA sets radiation protection standards.
- The NRC regulates waste management operations to meet EPA established limits.
- The DOE operates sites in conformance with NRC regulations.

The EPA plans to publish general environmental criteria in 1978. These will be quite broad and applicable to all forms of radioactive waste. Workshops were held in 1977 in Reston, VA, and Albuquerque, NM, to achieve broad public participation in preparing these criteria. The timetable for EPA's HLW program is designed to coordinate with the needs of the DOE to design disposal facilities and of the NRC to create a regulatory structure. The EPA has formed an Interagency Working Group made up of representatives of DOE, NRC, and USGS to exchange both technical and program information.(84) Six criteria for radioactive waste were provided by EPA for comment in November 1978: 1) definition of radioactive wastes, 2) length of isolation period, 3) assessment of risk, 4) unacceptable risks, 5) locations for radioactive waste disposal, and 6) procedures that provide additional protection.(85)

Currently, the regulation most applicable to high-level waste is 10 CFR 50, Appendix F.(18) This regulation states that HLW must be solidified within 5 y after reprocessing and be shipped to a Federal repository within 10 y after reprocessing. The regulation also requires that the solidified waste be chemically, thermally, and radiolytically stable to the extent that the internal pressure not exceed the safe operating pressure of the canister for the period up to 90 days after receipt at the repository.

The NRC is currently preparing a new regulation, 10 CFR 60, which will deal with disposal of high-level radioactive wastes in geologic repositories. A draft of this regulation may be available for public comment in early 1979. The proposed regulation will be in two parts.

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One part sets forth the general rules governing the licensing of DOE to receive, store, and dispose of high-level waste in a geologic repository. The second part sets forth performance objectives and technical criteria for site suitability, design, and operation of a geologic repository. The regulation is directed to geologic repositories because: 1) it is the DOE's currently intended direction, and 2) with the possible exception of permanent surface storage, geologic disposal is believed to be the only present method which reasonably assures that the waste can be disposed of with no undue risk to public health and safety. An NRC EIS will provide technical arguments that support this belief.

Currently there are two major performance objectives proposed in 10 CFR 60. These are:

- Facility Design and Operation: The facility and its systems shall be designed and operated so as to assure the control of radiation releases and exposure, as required by 10 CFR 20 including ALARA.

- Long-Term Performance: The sealed repository shall be shown by state-of-the-art procedures and analysis to have sufficient multiple natural and engineered barriers to waste migration to conservatively prevent the transport of radionuclides to the biosphere in amounts exceeding acceptable releases.

RISK STUDIES

Several investigators have used the "toxicity" or "hazard index" approach in an attempt to put the potential risk of nuclear wastes in perspective with the risk (radioactive or chemical) of naturally-occurring ores:

- Hamstra (86) showed that, after 300 to 500 y of decay, the toxicity of the high-level waste from LWR reactors is less than the ore from which the uranium came.

- Cohen (87) compared the projected release of radionuclides from a geologic repository to the natural release of radioactivity from the earth's crust. He concluded that nuclear power is a method of "cleansing" the earth over the long term, because the toxicity of the waste was less than that of the radium from the natural uranium ore deposits.

- Tonnessen and Cohen(88) showed that the toxicity of LWR and LMFBR wastes is less after 200 to 300 y than some chemically toxic ores. Their results are shown in Figure 9-1.

While these analyses are helpful, they have several limitations:

- These analyses do not address the transport of radionuclides in geologic and biologic media.

- They distribute the risk over all of society instead of to individuals living near the repository.

- Finally, they assume that the repository will be equivalent to the natural geology in preventing nuclear wastes from reaching humans.

More detailed studies are in progress to provide an adequate basis for setting criteria.

A study has been completed by Burkholder which considers the waste form/repository/geologic and biologic site as a system.(89) (Several other studies are also underway.) This study determined the importance of the various barriers to radionuclide movement, assuming that some event had occurred which permitted flowing water to contact the waste. The nuclide retention and leach resistance barriers were found more important than the containment barrier, but the containment should prevent water from reaching the waste until the waste temperature is low enough to prevent rapid leaching. This condition can be made more certain by proper selection of the geologic formation and by proper design of the repository.

A similar study (90) performed the same analyses for spent fuel, cladding waste, and various other solid wastes (TRU, LLW, ore tailings). Some conclusions were:

- Spent fuel requires greater nuclide isolation than HLW because more U and Pu are present.

- Good isolation conditions, compared to storage in the biosphere, can greatly reduce the projected maximum dose consequences to individuals. Poor conditions can increase it.

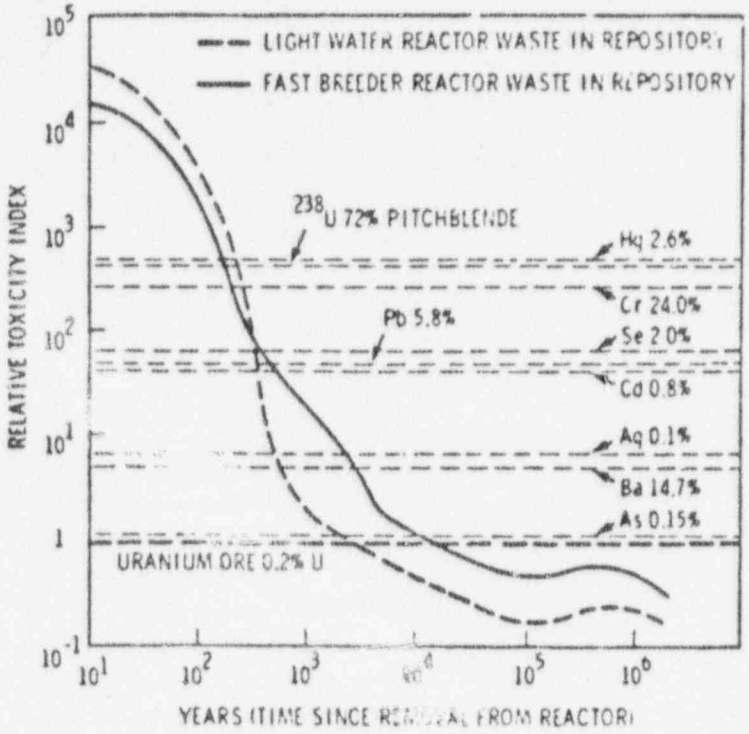

FIGURE 9-1. Relative Toxicity of Nuclear Waste Over Time, Compared With That of Average Mineral Ores of Toxic Elements

- Isolation at multiple sites can reduce the requirements for leach resistance of some waste forms.

Work is continuing on barrier and system studies at Lawrence Livermore and Sandia Laboratories (NRC-sponsored), the University of New Mexico and Arthur D. Little (EPA), and the Pacific Northwest Laboratory (DOE). Relatively little has been reported but the reports available suggest that significant progress is being made.(91-95)

A discussion of the OKLO natural reactor and its relevance to geologic isolation of radioactive waste is presented in Appendix C.

SUPPORTING DOCUMENTATION

Important links in the decision-making chain include defining the various options that may exist for nuclear waste management and determining the relative cost of these options, both in dollars and in environmental impact. Documentation completed or in progress in this area is highlighted below:

Technical Alternatives Document Completed

A study has been completed which characterizes and classifies various technologies for managing the nuclear wastes produced in reprocessing LWR fuels. This report, the Technical Alternatives Document (TAD), will serve as a reference document for the preparation of environmental impact statements and other documents required for licensing the construction and operation of waste management facilities.(24)

Commercial Waste Management Statement - Prepared

A generic environmental impact statement (GEIS) on the management of commercially generated nuclear wastes has been prepared. Recently, PNL completed a modification of this document, a Commercial Waste Management Statement (CWMS) (96), based on guidelines from the report by the DOE Task Force for review of Nuclear Waste Management(2). The study was completed in September 1978

and is for use by the government in preparing a policy statement on waste management. Several documents have been prepared to support the CoMS.(97-99)

Technical Alternatives Documents - Prepared for DOE Sites

Each of the DOE sites, SRP, INEL, and Hanford, has prepared a technical alternatives document characterizing the various technologies for managing the HLW stored (present and future) on the site.(19,21,22) Environmental impact statements based on these technologies are in preparation and will serve to guide programmatic decisions on the long-term management of the wastes.

Technical Alternatives Document-Prepared for WS

A technical alternatives document has been prepared for managing the small amount of commercial HLW in storage at the WS plant in West Valley, NY.(20)

International Fuel Cycle Evaluation Program - Underway

At the request of President Carter, a worldwide International Fuel Cycle Evaluation Program (INFCE) was established.(11) The purpose of this program is to conduct a thorough evaluation of the complete fuel cycle to identify how to best minimize the risks of proliferation, and at the same time, to assure that civilian nuclear power remains a viable energy source. Forty countries and four international organizations are participating in this program. An Organizing and Plenary Conference was held in October 1977. Working groups were established and co-chair countries for the groups were assigned. Most of the working groups have held two meetings. While the results of the INFCE will be advisory only, the outcome could well influence the nature of the future fuel cycle and, consequently, the composition of the high-level waste which would be produced.

DOE Review of Nuclear Waste Management - Complete

Early in March 1978, a DOE Task Force under the direction of J. M. Deutch completed a review of all nuclear waste management programs in the DOE.(2) Their report endorses geologic disposal of high-level wastes and spent fuels and recommends early testing of geologic disposal of a limited number of spent fuel assemblies. It also recommends government ownership and operation of all nuclear waste sites.

Interagency Review Group Established to Provide Recommendations to the President

The DOE Review of Nuclear Waste Management was studied by an Administration-established Interagency Review Group. Open meetings were included to get public response. A report on the review is due to the President in December 1978. (100,101) It will include a set of recommendations to be used in decision making and implementation.

Technology Exchange Meetings on Solidified Waste Forms - Underway

Several important technical meetings on solidified waste forms were planned in late 1978 and early 1979. The results of these meetings will be documented in proceedings to be published in 1979. These meetings include a symposium on Science Underlying Radioactive Waste Management, held in conjunction with the 1978 Annual Meeting of the Materials Research Society (November 28 - December 1, 1978, Boston, Mass.); the 5th Conference on High-Level Radioactive Solid Waste Forms (Denver, October 1978); and the upcoming Department of Energy/American Ceramic Society sponsored International Symposium on Ceramics in Nuclear Waste Management (to be held in Cincinnati, Ohio, April 30 - May 2, 1979).

PUBLIC ACCEPTANCE

Public concern over environmental quality has increased tremendously in the last few decades. The recent referendum on nuclear facilities in California, the current threat of a referendum in New York, and the many protests over nuclear construction are ample evidence of this concern. At the same time, when the issue of deciding on a nuclear moratorium was put before the voters in five states, by a two-to-one margin, those who voted favored continued development of nuclear power. However, the public concerns are evident and they conclusively show that public acceptance must be won if nuclear energy is to continue to grow. The Interagency Review Group study has instituted open meetings of the proposed U.S. nuclear waste management plans. The EPA has conducted similar open meetings on criteria.

Government agencies recognize that the public must be heard and informed if these plans are to be accepted.

Surveys of public attitudes toward nuclear power have been conducted [102,103] and the data have been analyzed.[104,105] Over 100 surveys were analyzed by Mazur and Ramsay.[105] They concluded that through 1976 a clear majority of the public favored the use of nuclear energy. In national probability studies conducted primarily during 1975 and 1976, on the average, approximately 60% of the public supported nuclear power, 23% of the public opposed nuclear power, and 17% of the respondents were undecided. On the issue of radioactive wastes, about 22% of the public felt that radioactive wastes are so dangerous that we cannot afford to risk producing them, while 25% were not sure, and 53% believed that modern technology can find ways to store wastes so there will never be a problem.[105] These results indicate that, to maintain the current pronuclear balance of public opinion, the technical community must have convincing evidence of the ability to safely manage nuclear waste. Such evidence must be readily available, written in lay terms that the general public can understand.

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APPENDIX A

HISTORICAL OVERVIEW OF NUCLEAR WASTE MANAGEMENT

Nuclear wastes are waste materials which are contaminated with radioactive elements. They are generated in all phases of nuclear operations: mining and milling uranium and thorium, fuel fabrication, reactor operations, irradiated fuel reprocessing, uranium enrichment, and fuel refabrication. Radioactive wastes are also generated in weapons manufacture, in R&D, during medical treatment, and in some industrial operations involving radioactive materials. Furthermore, the facilities where radioactive materials are generated--reactors, chemical treatment plants, laboratories--become contaminated with such wastes. Sooner or later these facilities become worn out or obsolete and must be disposed of in some manner; thus, more nuclear wastes are generated.

Management of these wastes to prevent unwanted dispersal of radioactive materials and to maintain safe levels of exposure to workers in the field and the general public began with large-scale production of wastes during World War II. This was almost entirely in connection with defense activities.

Early nuclear waste management can be characterized as storage, burial, or release, depending on the activity and form of the waste. It was based on the best technology available at the time. Waste management was influenced by World War II and postwar needs, and the view that the overall problem was to be confined to defense activities.

High and intermediate-level liquid wastes were stored in underground tanks; low-level liquid wastes were discharged to the ground at a safe level. Very low-level liquid wastes were discharged to ponds or streams. Solid wastes were buried in near-surface trenches. Iodine-131 was considered the major gaseous waste. Interim fuel storage time to allow decay before processing was the primary control on the discharge of ^{131}I.

Some of the waste management practices which were followed in post WW II times would not meet Environmental Protection Agency (EPA) criteria or Nuclear Regulatory Commission (NRC) requirements today. These requirements have progressively become more severe as the technology has advanced. However, it has not been shown that any significant risk to the environment was created by the past practices.

Early R&D efforts toward better waste management involved improving storage tanks and conditions, reducing the volume of stored wastes and of wastes produced, decontaminating streams to permit release, reusing contaminated materials, and developing silver reactors to remove iodine from off-gas streams (largely to permit processing after a shorter cooling time).

As the defense activities continued to grow and as the potential for a nuclear power economy was recognized concern over the long-term safety of nuclear wastes increased. This was reflected in 1970 by the U.S. Atomic Energy Commission (AEC) ban on further burial of wastes containing significant amounts of long-lived alpha-emitting nuclides. Because the storage of liquid wastes is expensive and involves long-term risks, calcination was developed. To make high-level wastes even more inert to water, technology was developed for converting these wastes to glasses, and concepts were studied for converting them to cements, ceramics, and metal matrices. Then concepts were studied for destroying the radioactive nuclides (transmutation), getting them away from the earth (extra-terrestrial disposal), or isolating them from the biosphere in a geological formation (sea bed, ice sheet, geologic

[illegible]

[illegible]

- [illegible]

- [illegible]

- [illegible]

Origins of these various wastes in a typical plutonium recycle fuel cycle and in a once-through fuel cycle are shown in Figure A-1.

The present U.S. overall waste management reference system for commercial wastes is shown in Figure 1-1 in the main body of this report. It provides for isolation of HLW and TRU wastes in some location remote from the biosphere and in a manner where uncontrolled reentry into the biosphere is very unlikely. Because both wastes contain significant amounts of long-

[illegible]

Isolation of HLW and TRU wastes in geologic formations depends on a multiple barrier system to prevent reentry of radioactive wastes into the biosphere. Figure 1-2 (main body of report) shows some of the barriers which may exist or be built in this multibarrier system. Corrosion-resistant containment of the waste will delay contact of the wastes with entering water. A relatively insoluble waste form, e.g., glass, will retard leaching of radioactive constituents if water enters the storage location. Surrounding the waste canister with engineered barriers, such as materials impervious to water or which adsorb and retain radioactive elements will aid in delaying dispersal. Remoteness of the repository from the biosphere and the slow movement of radioactive constituents through rock and soil paths to the biosphere provide natural isolation barriers.

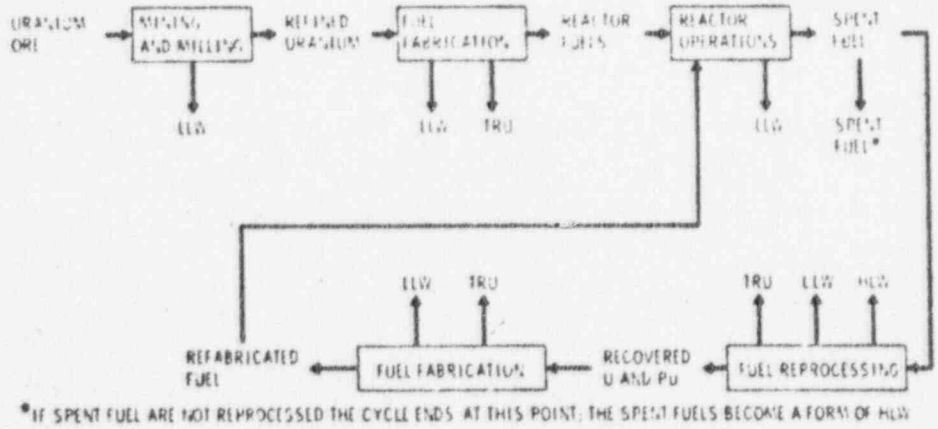

FIGURE A-1. Sources of Nuclear Wastes in the Fuel Cycle

APPENDIX B

OVERVIEW OF FUEL REPROCESSING [illegible]

[illegible] reprocessing of [illegible] fuels began with the recovery and purification of plutonium for weapons from fuel irradiated in Hanford reactors. The first process, the Bismuth Phosphate process, involved a series of batch precipitations to separate and purify plutonium. Uranium was not recovered; it was [illegible] as a waste and large volumes of waste were produced.(1)

CONVENTIONAL FUEL REPROCESSING

Many techniques have been studied on a laboratory scale (in some cases, pilot-plant scale) for separating and purifying plutonium and uranium from irradiated nuclear fuels. These studies include solvent extraction, fluoride volatility, ion exchange, adsorption and zone refining. Today, virtually all nuclear fuel reprocessing is done by solvent extraction although other techniques may be used for specific steps (e.g., ion exchange for plutonium purification). Figure B-1 schematically shows a typical solvent extraction process.

Dissolved fuel (in HNO_3) flows into Column 1A where it is contacted with an organic solvent. Both uranium and plutonium are extracted into the solvent while ~99% of the fission products and other transuranium elements are left in the aqueous raffinate. This raffinate is the HLW. Uranium and plutonium are stripped from the solvent in Column 1A. Columns 2A, 2B, and 2C separate and further purify uranium and plutonium.

Solvent extraction has several advantages:

- It is readily made a continuous process.
- The solvent can be easily recovered for reuse.
- [illegible] largely [illegible].
- Waste volumes are relatively small.
- Uranium and plutonium recoveries are [illegible].
- Product purity is high.

This process meets the spent fuel reprocessing needs [illegible] plutonium recycle [illegible] very well.

REPROCESSING AND RELATED EXPERIENCE

The technology of nuclear fuel reprocessing is well advanced; it has not changed much in the last few years. Many plant-years of successful operation have been logged in both domestic and foreign plants. Tables B-1 and B-2, updated from Dau and Williams (2), show the plant-years of experience in the U.S. and foreign plants. Experience in the U.S. has been largely with reprocessing defense fuels. The only commercial fuels reprocessed in the U.S. have been at the NFS plant. This plant has not operated since 1972. The General Electric Company built a small (300 t/y) plant at Morris, IL, but it has never been operated. Spent fuels are stored in both the NFS and GE storage basins. A 1500 t/y plant was built by Allied-General Nuclear Fuels Services (AGNS) at Barnwell, SC. To date, a plant-operating license has not been issued for the AGNS plant. Exxon Nuclear Company has designed a fuel reprocessing plant proposed for construction near ORNL in Tennessee. In view of the present indefinite deferral of commercial nuclear fuel reprocessing in the U.S., the future of these plants is unknown. However, AGNS was funded by the U.S. government in FY-1976 to study alternative operating schemes.

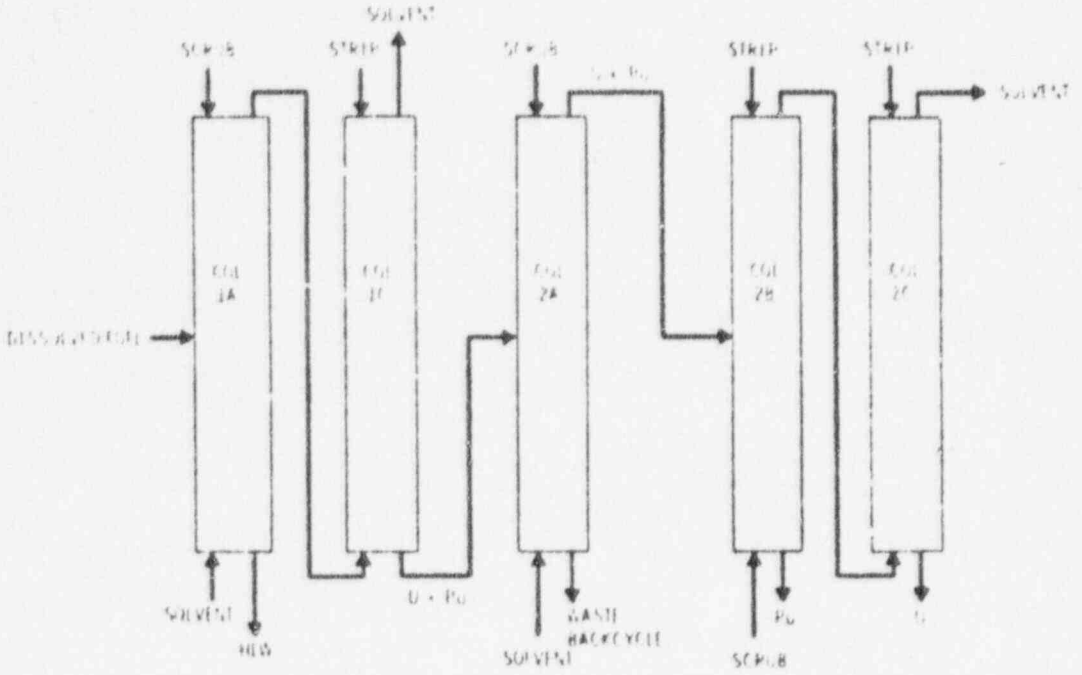

FIGURE B-1. Schematic of a Typical Solvent Extraction Process

Commercial nuclear fuel reprocessing has not been deferred in foreign countries. Reprocessing plants continue in operation abroad and new plants are being planned, as shown in Table B-3.(3) Most notable among the new operating plants are those at La Hague in France and at Windscale in Great Britain. Other countries are now planning significant reprocessing facilities.

ALTERNATIVE REPROCESSING SCHEMES

The U.S. is presently committed to an indefinite deferral of spent nuclear fuel reprocessing. The government is encouraging other nations to do the same until the role of nuclear energy in supplying the world's energy needs and approaches to minimizing the potential for nuclear weapons proliferation can be better understood. If spent fuels are not reprocessed to recover plutonium and uranium, the energy represented by them is lost. Furthermore, fuel reprocessing is required to support a breeder reactor program which some believe is necessary to assure meeting long-term energy needs.

These considerations have prompted interest in alternative reprocessing-refabrication schemes and alternative fuel cycles which would provide low potential for nuclear weapons proliferation. An example is the CIVEX process.(4,5) This process would permit reprocessing nuclear fuels, to permit using the energy values while minimizing the potential for proliferation of nuclear weapons. No weapons-usable material would be produced. Instead, plutonium would be left with uranium and a small percentage of waste products, which make the product highly radioactive. Subsequent separation of the plutonium is impossible without complex and highly sophisticated equipment. The product could be refabricated into fuel rods by remote operation.

In 1977 and 1978 the DOE was supporting work on development of alternative fuel cycle technologies.(6) The DOE was also supporting studies on fuel cycles that do not directly involve uranium or plutonium. A program at Savannah River Laboratory was evaluating the use of thorium fuel in LWRs as a possible solution to the potential proliferation problem.(7) Conceptual designs were being prepared for the thorium fuel cycle facilities and supporting R&D efforts were underway to solve new and different problems associated with processing thorex fuel.

TABLE B-1. U.S. Nuclear Fuel Reprocessing and Related Plant Operating Experience

Plant	Type of Process	Operating Time	Plant Operating Years
Hanford Operations Plant Experience			
B Plant	Precipitation	1945-1952	7
T Plant	Precipitation	1944-1956	12
U Plant	Solvent extraction	1952-1967	15
REDOX	Solvent extraction	1954-1966	12
PUREX	Solvent extraction	1956-1972	16
Z1 Plant	Precipitation	1952-1955	3
308 Plant	Precipitation, melt refining, fabrication	1944-1975	27
waste Conversion	Solvent extraction, ion exchange	1965-1970	4
Hot Semi-works	Solvent extraction	1953-1957	5P*
WSEP	Calcination/ vitrification	1966-1972	4P
			113
Nuclear Fuel Plant Experience			
H Plant	Solvent extraction	1955-1971	21
F Plant	Solvent extraction	1954-1971	24
			47
Idaho National Engineering Laboratory Plant Experience			
Chem. Separations	Solvent extraction	1953-1965	25
waste Calcination	Calcination	1963-1974	15
EBR-II Reprocessing	Melt refining	1964-1969	5P
			45
Oak Ridge National Laboratory Experience			
TRU Plant	Solvent extraction, ion exchange	1966-1969	12P
Metal Recovery	Solvent extraction	1952-1961	9P
Radiochemical Pilo. Plant	Solvent extraction, volatility	1959-1963, 1965-1976	5 26P
			52
U.S. Private Reprocessing Experience			
NFS	Solvent extraction	1966-1972	6
TOTAL PLANT OPERATING YEARS			121

*P designates pilot-scale operation = 0.1 to 0.5 t/day.

TABLE B-2. Foreign Nuclear Fuel Reprocessing and Related Plant Operating Experience

Country	Plant	Type of Process	Years of Operation	Plant Capacity (tons/yr)
United Kingdom	Windscale I	Solvent extraction	[illegible]	12
United Kingdom	Windscale II	Solvent extraction	[illegible]	15
Scotland	Dounreay I	Solvent extraction	[illegible]	[illegible]
Scotland	Dounreay II	Solvent extraction	[illegible]	[illegible]
Belgium	Eurochemic	Solvent extraction	[illegible]	60
France	La Hague	Solvent extraction	[illegible]	[illegible]
France	Marcoule	Solvent extraction	[illegible]	[illegible]
Germany	Wak	Solvent extraction	1971-1974	35
Italy	Eurex I	Solvent extraction	1971-1973	[illegible]
Italy	ITREC	Solvent extraction	1974-1977	5
Norway	Kjeller	Solvent extraction	[illegible]	20
India	Trombay	Solvent extraction	[illegible]	100
France	PIVIC	Vitrification	1969-1973	40
France	AVM	Vitrification	1972 -	60
United Kingdom	[illegible]	Vitrification	[illegible]	30

P designates pilot-scale operation - 0.1 to 0.5 T/day.

[illegible]

Country	Plant	Status	Major Milestones
[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	
[illegible]	[illegible]	[illegible]	
	[illegible]	[illegible] metal fuels	
	[illegible]	[illegible] kg/y operational pilot plant [illegible] fuels. Reprocessing breeder fuels	
	[illegible]	[illegible] t/y, operational, [illegible] for LWR fuels	
	[illegible]	[illegible] reprocessing plant for LWR fuels. [illegible] to service foreign customers	Startup of first [illegible]. Startup of second [illegible]
Germany (FRG)	WAK [illegible]	40 t/y operational pilot plant for R&D	
	[illegible]	1400 t/y commercial plant for LWR fuels	Startup--1990
	JUPITER (Jülich)	2 kg/d pilot plant for HTGR fuel	Startup--1980
India	Trombay	60 t/y operational pilot plant for U metal or oxide fuels	
	Tarapur	100 t/y, oxide fuels	Startup--1977
	Kalpakkam	Planned--to handle CANDU and thorium FBR fuels	
Italy	EUREX (Saluggia)	30 kg/d, operational MTR and oxide fuels	
	ITREC (Trisaia)	15 kg/d, operational U-Th and Pu-U fuels	
	Commercial	1000-1500 t/y	Startup--late 1990s
Japan	Tokai	210 t/y, operational, oxide fuels	Startup--1977
	Commercial (two plants)	Plants are planned	Startup of first--late 1980s
Pakistan	Commercial	Capacity--100 t/y range	
Spain		Lab-scale, 2000 kg/y--in design stage	Commercial plant--late 1990s

Nation	Plant	Status	Major Uncertainties
United Kingdom	Windscale	[illegible]	
	Dorset (Winfrith)	[illegible]	[illegible]
	Dounreay	[illegible]	
United States	West Valley, NY	[illegible]	
	Morris, IL	[illegible]	
	Barnwell, SC	[illegible]	
USSR	Virgin Radium Institute	Experimental (several kg/d) reprocessing facility	
		[illegible]	

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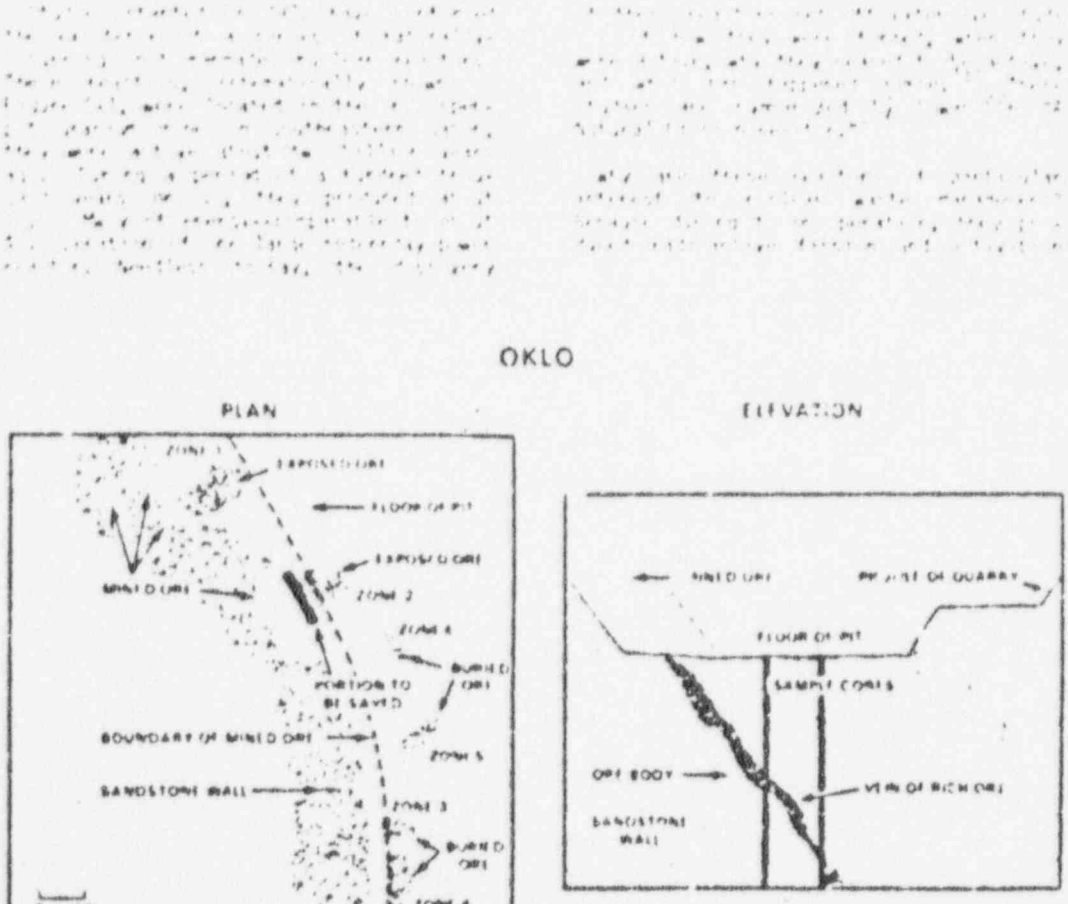

SIX REACTOR ZONES AT OKLO WERE FOUND IN LENSES OF EXCEPTIONALLY RICH ORE WITH AN AGE CONCENTRATION OF URANIUM FROM 20 TO 30 PERCENT THE URANIUM IS STRONGLY DEPLETED IN U 235

ORE BODY AT OKLO SHOWN IN A SCHEMATIC PROFILE OF THE MINE LIES ABOVE A SANDSTONE WALL WITH AN AVERAGE SLOPE OF ABOUT 45 DEGREES THE REACTOR ZONES ARE ROUGHLY A METER THICK AND HAVE A LENTICULAR CROSS SECTION

FIGURE C-1. Schematic of OKLO Open-Pit Uranium Mine

products much as are present in the nuclear wastes whose disposal is of concern now. And they were contained in a geologic formation in a manner somewhat similar to current plans for isolating nuclear wastes. Here, then, is an excellent opportunity to study the fate of radioactive materials, comprising the same chemical elements of concern to us now, when placed (formed) in geologic isolation.

If nuclear wastes are placed in a dry, or nearly dry geologic formation, a major concern is: What happens if water somehow gets into the repository where it may react with the wastes and possibly carry them back to the biosphere? Mathematical models, based on what we know about the behavior of waste constituents in aquifers, are constructed to predict what will happen, how long it will take for specific constituents to travel and how far and in what amounts. But the times of concern are long compared to present experimental capability and it would be helpful if there were a real system with which to compare calculated results.

The OKLO reactor sites are very helpful in this regard. It is postulated that water had to be present in the uranium deposits to carry and concentrate the uranium there and to moderate the neutrons to maintain the necessary chain reaction. Also, it is probable that the uranium deposits during their active reactor period were buried deeply beneath the surface, and the event occurred long ago, giving an adequate time frame.

What has been learned so far that is helpful?(3)

- Most of the rare gases (Kr, Xe) are gone from the reactor sites.
- Most of the monovalent metals (Rb, Cs) and the bivalent metals (Ba, Cd, Sr) have migrated away.
- Most of the molybdenum is missing.
- Most of the ^{90}Sr apparently remained in place until it decayed to ^{90}Zr.
- Most of the higher valence metals have remained in place during the nearly two billion years since the deposits were reactors. This includes the long-lived transuranium elements which we wish to isolate for many thousands of years.

How can this information be applied to the concept of geologic isolation of nuclear wastes? In general, the results agree with current technology on the behavior of various elements in an aquifer in contact with soils--high valence metals tend to be retained; low valence elements tend to move. Retention of the long-lived transuranium elements even in the presence of water is encouraging because even if water enters an initially dry repository, these long-lived nuclides will not become dispersed over very long time periods. The observation that most of the ^{90}Sr decayed to ^{90}Zr before strontium migrated indicates that migration of the bivalent metals was slow even with water present (^{90}Sr has a 30-y half life). If an initially dry repository remains dry, or the wastes are protected by engineered barriers, for several hundreds of years, the short-lived nuclides will decay in place to levels that represent little biosphere risk.

Care must be exercised in extrapolating these results to any given geologic repository. The exact geologic history of the OKLO site is not likely to be duplicated by a modern waste repository. Migration rate data used for the various isotopes must be site-specific for a proposed repository. But the OKLO data do lend credence to the concept of geologic disposal and encouragement that conditions can be found or engineered to assure long-term retention of critical radionuclides.

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