Chapter 1

High-Level Waste Management at the DOE Weapons Complex

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Chapter 1
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OVERVIEW

The first high-level defense waste was created as a byproduct of the production of plutonium in a natural uranium-graphite reactor at Hanford and the subsequent remote “reprocessing” of irradiated uranium fuel elements to recover plutonium. The byproduct was a highly radioactive, acidic, aqueous solution containing a variety of fission products with a wide range of half-lives, as well as residual uranium and some residual radionuclides with larger atomic numbers than uranium—the transuranics. It was recognized that this liquid high-level waste (HLW) required careful handling, as well as isolation from people and the environment for many years. HLW is generally distinguished from other radioactive waste types by its intense radioactivity coupled with the longevity of its hazard. Huge, underground, single-shell carbon steel tanks, eventually 149 in number, were built to store neutralized liquid HLW at Hanford. An early practice of discharging some of the liquid from the HLW tanks into “cribs” and then into the soil was subsequently discontinued. When some tanks began to leak, new tanks of double-shell design were added.

Today, most liquid HLW has been neutralized, forming mixtures of liquid, sludge, and salt cake, and is currently stored on-site in steel tanks, some of which have leaked and represent a potential threat to groundwater. Storage of waste in less expensive carbon steel, rather than stainless steel, tanks after neutralization of acidic HLW requires complicated waste handling and treatment. There is also concern about the possibility of fire or explosion in the waste tanks, accompanied by the release of radioactivity.

Four Department of Energy (DOE) sites have HLW: the Hanford Plant, the Savannah River Site, the Idaho National Engineering Laboratory (INEL), and West Valley, NY; the last, a nonweapons site, reprocessed some fuel commercially from 1966 to 1972. The prime contractors for the management of HLW at all four sites are subsidiaries of Westinghouse. Two sites have more than 90 percent of the HLW by both volume and radioactivity—Savannah River and Hanford—and are planning to begin operations to immobilize HLW in 1992 and 1999, respectively, although slippage of these schedules would not be unusual. The Savannah River vitrification facility was built at a cost of about one billion dollars. The Hanford facility is not yet constructed but plans call for it to be very similar to Savannah River. The West Valley site is also scheduled to begin vitrifying waste in 1996; the cost of all West Valley operations, including decontamination and modification of existing facilities to accommodate vitrification as well as new construction needed for the vitrification plant, will be on the order of one billion dollars. Canisters of vitrified waste (“glass logs”) are to be stored on-site, pending disposal in a deep geologic repository that is not expected to begin operation until the second decade of the 21st century. In contrast to the other three sites, for 25 years INEL has been converting liquid HLW from the reprocessing of highly enriched uranium-235 spent fuel, from naval and other reactors, to a powdery solid calcine and storing it in stainless steel bins; DOE has not made a final decision about the waste form for immobilization and disposal of INEL HLW.

At West Valley, DOE is reducing the volume of high-level tank waste to be vitrified by separating a portion of the waste that DOE believes qualifies as low-level waste, mixing it with cement, and temporarily storing it in drums above ground, pending a disposal decision through the Environmental Impact Statement (EIS) process. An analogous separation is planned for Savannah River and Hanford because it will greatly reduce the amount of waste to be vitrified and should substantially reduce disposal costs if the portion immobilized in grout or concrete can be disposed of on-site at or near the surface. At West Valley, DOE sought and obtained Nuclear Regulatory Commission (NRC) approval to perform such a separation; NRC has oversight authority under the West Valley Demonstration Act of 1980. However, there appears to be no such NRC authority at the weapons sites. Concerns have been raised by interested members of the public about the safety of such waste separation; the grouted waste at West Valley is reported to be “Class C low-level waste” containing technetium-99, a long-lived (210,000-year half-life) beta emitter. In South Carolina,

\footnote{In February 1991, a 2-year delay in operation of the Hanford Vitrification Plant was announced by DOE.}
Long-Lived Legacy: Managing High-Level and Transuranic Waste at the DOE Nuclear Weapons Complex

nevertheless, DOE is moving ahead, indicating that it has all necessary permits from the State to begin “saltstone” operations at Savannah River; by July 1990, those operations were underway but not using waste from the main high-level tank farm.

Uncertainty exists about the composition of HLW at DOE weapons sites. The uncertainty arises because of the variety of processes that have been used, the past mixing of wastes, and the heterogeneity of tank components after neutralization. Sampling is very difficult because of tank design, the high radioactivity levels, and concern about the possibility of tank explosions. Knowledge of waste composition is important in designing waste treatments and it is needed for proper glass-waste formulation for the vitrification process.

Historically, DOE has regulated HLW at weapons sites under the Atomic Energy Act. However, EPA has become a major factor in regulating waste management at the weapons sites through its jurisdiction over hazardous waste and application of the Resource Conservation and Recovery Act (RCRA) and the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) to these sites. State agencies have also become involved under RCRA and through mechanisms such as interagency agreements under CERCLA.

The basic thrust of the HLW management program of DOE is to move from the present less secure, less stable, less controlled condition to a more stable one by immobilizing the tank waste. HLW vitrification, if successful, should reduce the threat of groundwater contamination and tank explosions posed by liquid HLW stored in tanks. An objective of vitrification is to produce a waste form that will immobilize waste safely for hundreds or thousands of years; however, the process chosen, involving borosilicate glass, has yet to operate on a large scale in the United States, and long-term performance of the vitrified waste form in various settings is difficult to predict and hard to verify. If vitrification works as planned, the glass logs produced represent a potentially stable form for long-term storage on-site or in a monitored retrievable facility if the deep geologic repository should be delayed. DOE and most experts working within the DOE program believe that, at present, vitrification using borosilicate glass is the best available technology for geologic disposal. However, some concerns have been raised about whether DOE will be able to demonstrate that borosilicate glass will perform as required in the Yucca Mountain repository environment.

The Transition to More Stable Waste Form

A significant transition is beginning to take place from the less secure and more threatening storage of HLW in tanks to the more promising secure storage of immobilized HLW in solid, glasslike form. Bringing about this transition is a major and costly undertaking, and a successful outcome is far from being achieved. However, if it can be accomplished with minimal occupational risk to workers, it should greatly reduce if not remove the current, ever-present threat and concern regarding tank leaks and explosions. The nominal design lifetime of vitrified waste using borosilicate glass is such that even if a geologic repository were delayed significantly, the glass logs could be stored safely on-site at Savannah River and Hanford for hundreds of years, as long as the necessary institutional controls remain in place. Calcine, even without immobilization in glass or ceramic, also appears able to be safely stored for hundreds of years at INEL.

The legacy of past practices in which HLW was discharged into cribs or stored in 149 single-shell tanks at Hanford must still be dealt with; DOE has not yet decided how to accomplish the necessary decontamination and safe disposal.

Monitoring the Waste Forms

Because of the importance and the cost of vitrification to improve the safety and stability of HLW storage and disposal, it is essential to carefully monitor and regulate the integrity and hazard potential of the waste forms, including both vitrified and concrete products. Continuing studies and monitoring are required to resolve opposing claims that may arise concerning safety and health risks during storage, along with a continued strong research program on waste stability, container integrity, and radionuclide transfer through the environment.


\[^{3}\text{There is, however, considerable experience with commercial HLW vitrification in Europe, especially France, using a process somewhat similar to that built at or planned for these DOE facilities.}\]

\[^{4}\text{Factors concerning waste stability over the long term that need investigation include leaching, embrittlement, and corrosion.}\]
Form of HLW at the Idaho National Engineering Laboratory

At INEL, DOE followed a very different waste management approach and decided to produce a dry, calcined waste form for the storage of HLW. This decision provided considerable experience with an alternative to the approach used at Hanford and Savannah River. The calcined waste form has proved suitable for interim storage. For ultimate disposal, work is focusing on glass ceramic as a promising medium for immobilization of the calcine in order to reduce disposal costs relative to the use of borosilicate glass. Cost, although important, is but one factor to be considered. Environmental integrity in response to evolving, possibly tightened environmental standards for radioactivity is another.

Calcining appears to be a proven, relatively low-cost means of solidifying liquid waste and a viable medium-term (about 500 years) alternative to vitrification. More research on the calcined waste form and on bin hardening of calcine for disposal could supply data for use if some future treatment, storage, and/or disposal alternatives were considered.

HLW Repository

The U.S. approach to HLW disposal is to license and use a geologic repository to contain potentially harmful radionuclides for the tens or hundreds of thousands of years that may be necessary. The Swedish approach places more reliance on engineered barriers, including a thick container wall to provide the necessary isolation; other European countries are also focusing on engineered barriers. By contrast, in the United States, current policy places reliance on the geologic repository itself.

The U.S. high-level defense waste glass logs are to be formed in thin-walled canisters that meet NRC repository criteria; the canisters will then be put in containers before repository emplacement. One issue is whether to place more reliance on engineered barriers for isolating the waste, for example, by increasing either canister thickness or container thickness and backfill, or by some combination of these modifications for the geologic repository setting. Factors to consider include cost, interaction between barriers and the chosen repository environment, and the fact that both defense HLW and commercial spent fuel are now required to use the same container design.

Standards for HLW Disposal

Standards for disposal of HLW have been re-issued by the courts but are expected to be reissued for comment by the Environmental Protection Agency (EPA). These standards have important implications for defense HLW. The long-term trend has been to issue progressively more stringent standards for nuclear waste disposal as new information becomes available about health risks from radiation exposure. This trend may continue but the process is very slow and thus has affected planning for ultimate safe disposal.

Timeframe for Immobilization

Decisions about the urgency and rapidity with which liquid high-level tank waste should be immobilized are difficult to make because of the lack of good information on the contents of some tanks or on the movement of radioactive and hazardous materials that have leaked from tanks. In the absence of such information, and given continuing concern about the possibility of waste tank explosions, it may be prudent to move forward with vitrification projects as quickly as feasible and to make sure that technical, environmental, and policy questions or concerns are addressed promptly and effectively as well. Trade-offs between moving ahead with dispatch and moving ahead too precipitously require careful consideration. Among the current concerns is the possibility of explosion when disturbing or heating tanks or tank contents. Also, occupational radiation doses should be carefully controlled and monitored.

Airborne Releases

Airborne release of both radioactive and hazardous materials is a significant potential health threat from DOE weapons sites. The movement of contaminants in the air is direct and rapid compared with movement via groundwater. Reactors, reprocessing plants, and HLW treatment operations all involve some routine air emissions; in addition, there is the possibility of accidental release. Although air releases have been greatly reduced from the early days of the weapons program, attention to air emissions

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is continually important, both in setting standards and in monitoring waste management activity.

**Future of the PUREX Plant at Hanford**

The Plutonium and Uranium Extraction (PUREX) fuel reprocessing plant has been of concern because of its age, the large amounts of hazardous and radioactive wastes it produces, past atmospheric releases, and continued release of liquid effluents to the soil. In early 1990 DOE had plans to restart PUREX to reprocess backlogs of spent defense fuel over a 5-year period and then to close the facility permanently. A principal reason for this was concern about the 2,100 metric tons of metallic spent fuel from the Hanford N reactor awaiting reprocessing and currently being held in water basins at K plant, near the Columbia River. DOE needs to prevent any radionuclide release from failed fuel elements since these basins have leaked in the past.

In October 1990, DOE announced that it would not restart PUREX for at least 2 years, but would prepare an Environmental Impact Statement (EIS) to evaluate a variety of treatment and disposal methods for stored N reactor fuel. Some have interpreted this announcement as indicating that PUREX will never again operate. The EIS process requires full consideration with public input of the impact of alternatives to restart, including the consequences of continued storage of fuel elements in the K basins.

**Learning From International Experience**

Since the U.S. decision in the 1970s not to encourage reprocessing of commercial nuclear fuels, other nations have moved ahead in acquiring expertise in reprocessing that could prove useful to DOE’s fledgling efforts at waste minimization. Learning from these international sources could be part of a necessary upgrading of DOE waste minimization activity, particularly in planning for any new reprocessing capability in connection with modernization of the Weapons Complex.

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**INTRODUCTION AND DEFINITIONS**

High-level radioactive waste is a consequence of the materials and methods used by the United States to produce plutonium and tritium for nuclear weapons. Neither substance occurs in nature, and both are produced by neutron capture in nuclear reactors. For these reactors to operate, neutron chain reactions must occur, which require that fission takes place in one or more heavy isotopes of uranium. Fission produces numerous elements—strontium-90 and iodine-131 are two examples—that are radioactive. Each time a fission event occurs, two (or occasionally three) radioactive fission products are formed.

In the United States, the high-level waste (HLW) found in defense facilities differs from its spent fuel counterpart found in the commercial sector. Commercially, fission products, uranium and transuranic isotopes, are contained in irradiated fuel elements removed from reactors—so-called spent fuel—that (with the exception of a plant operating intermittently in West Valley, NY from 1966 to 1972) have not been subjected to reprocessing. In reprocessing, fuel elements are chopped up and dissolved; plutonium and, in some cases, uranium are separated from the fission products for reuse in a reactor. Spent fuel in the U.S. commercial sector is stored at reactor sites pending development of a deep geologic repository for its disposal. In this report, spent fuel is not considered to be a form of, or to fall within the definition of, defense HLW. However, spent fuel is present at some Department of Energy (DOE) weapons sites.

High-level waste is defined by a DOE order as “the highly radioactive waste material that results from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid waste derived from the liquid, that contains a combination of transuranic waste and fission products in concentrations requiring permanent isolation” (63). Box 1-A compares this and other definitions of high-level waste.

Defense HLW arising from reprocessing of fuel elements or irradiated targets (see figure 1-1) leaves...
the reprocessing plant as highly radioactive liquid that usually contains more than 99 percent of the nonvolatile fission products from the fuel elements or targets during their time in the reactor. It also contains roughly 0.5 percent of plutonium and 0.5 percent of the uranium that was present in the spent fuel, if both these elements are recovered in the reprocessing operation (3). The radioactive liquid is stored in tanks, pending conversion to a glassy solid by a process known as vitrification. The major exception to this waste form conversion is at the Idaho National Engineering Laboratory (INEL) where some HLW liquids have been subject to calcination, producing a powdery solid that is not generally considered to be in final form for disposal.

The only way to turn off the HLW source term (the ultimate in HLW waste minimization) is to stop producing plutonium and tritium, to produce them by methods completely different from those currently used, or to utilize a substitute for plutonium whose production does not require a nuclear reactor. For example, uranium-235, which occurs in nature could, in principle, replace plutonium if the need for new fissionable material arose. Plutonium might be recovered from existing materials in the civilian reactor program, but that would violate a long-standing policy of separation of military and civilian nuclear activities. Tritium, although not essential for weapons, makes them smaller and enhances their explosive yield; it could conceivably be made in a linear accelerator rather than a reactor.

The feasibility of alternatives to current nuclear weapons materials and production methods is not explored here. However, the intense radioactivity of HLW is a direct consequence of present production methods, and HLW minimization is difficult at current facilities. As of late 1990, no high-level defense waste was being generated anywhere in the DOE complex, although reprocessing facilities at Savannah River and Idaho were scheduled to resume operations and small amounts of research reactor fuel were being reprocessed at Savannah River. In January 1990, DOE agreed to perform a programmatic Environmental Impact Statement (EIS) of its plans for modernizing weapons facilities and that process began in late 1990 (25).

Unlike civilian HLW which is “locked up” in solid fuel elements, defense HLW is currently stored in tanks, either as liquid or a form resulting from waste neutralization and liquid evaporation, including sludge, salt cake, and slurry. Some of these tanks were built in 1943; some have already leaked. The composition of waste in some tanks is not well known, and concerns have been raised about possi-

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**Box 1-A—Definitions**

**High-Level Waste:** The highly radioactive waste material that results from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid waste derived from the liquid, that contains a combination of transuranic waste and fission products in concentrations requiring permanent isolation.

**Spent (Nuclear) Fuel:** Fuel that has been withdrawn from a nuclear reactor following irradiation, but that has not been reprocessed to remove its constituent elements.

**Transuranium (Transuranic) Radionuclide:** Any radionuclide having an atomic number greater than 92.

SOURCE: DOE Order 5820.2A.

**Two Other Definitions of High-Level Waste**

**Nuclear Regulatory Commission:** 1) irradiated reactor fuel; 2) liquid wastes resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated wastes from subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuel; and 3) solids.

SOURCE: 10 CFR Part 60.


The highly radioactive material resulting from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid material derived from such liquid waste that contains fission products in sufficient concentrations; and B) other highly radioactive material that the [Nuclear Regulatory] Commission, consistent with existing law, determines by rule requires permanent isolation.

The essence of DOE’s strategy for managing HLW, described in the 1989 Five-Year Plan (52), is to move from the current situation in which most HLW is in tanks as liquid, sludge, or salt cake to a situation in which the waste is immobilized, put in stainless steel canisters, and eventually shipped to a deep geologic repository for disposal. According to DOE, “Vitrification and calcining are two demonstrated methods for treating HLW for storage and/or disposal” (52).

In its 1989 Five-Year Plan, DOE characterized vitrification as follows: vitrification produces a glasslike form with “long-term stability.” Extensive research undertaken in fiscal year (FY) 1983, which included consideration of about 15 different waste forms, resulted in DOE selection of borosilicate glass as a suitable final HLW form. After vitrification, waste is poured into stainless steel canisters that are sealed and stored until a geologic repository becomes available. Three facilities are planned to use this vitrification process: 1) the Defense Waste Processing Facility (DWPF) at Savannah River, 2) the West Valley Demonstration Project (WVDP), and 3) the Hanford Waste Vitrification...
Figure 1-2—High-Level Waste at DOE Facilities

DOE plans to pretreat the high-level liquid waste streams at Hanford, Savannah River, and West Valley to reduce the volume of waste to be vitrified. Pretreatment involves evaporation and separation of a "low-level" fraction to be disposed of by mixing with cement to make a concrete or grout that is subsequently placed in a disposal facility at or near the surface. Facilities for pretreatment have been constructed; some startup operations have begun at Hanford and Savannah River, whereas immobilization with cement is quite far along at West Valley. The "low-level" grout material at Hanford is subject to mixed waste regulation under the Resource Conservation and Recovery Act (RCRA). However, DOE contends that the Savannah River
grout is a “non-hazardous waste form and qualifies for disposal in an industrial waste landfill.”

Calcination solidifies liquid waste by spraying droplets onto hot particles, resulting in a granular end product that is transferred to stainless steel bins encased in near surface concrete vaults. In the process, the volume is reduced eightfold (9). Calcining operations began at the Idaho chemical Processing Plant (ICPP) in 1963. The newest facility, the New Waste Calcining Facility (NWCF) that came on line in 1982, has calcined more than 2 million gallons of liquid waste (73). The NWCF has had two extended shutdowns and was not operating throughout most of 1990 (35). According to the 1989 Five-Year Plan, although calcined waste is suitable for extended storage (400 to 500 years in the current stainless steel bin-concrete vault arrangement) (.12 “...DOE has not determined its acceptability for final disposal” (15). Calcined waste is “readily retrievable and, if necessary, will be immobilized for disposal.” Design of an immobilization plant is due to start in FY 2002 “if this is the decision of DOE”; the treatment method is unknown (52). DOE appears to be leaning toward a glass ceramic as the preferred final waste form.

Conditions of HLW storage is described in the 1989 Five-Year Plan (53). All HLW is mixed waste. Double-shell tanks at Hanford and Savannah River containing liquid waste meet RCRA requirements. However, new tank construction is planned for INEL. “Double-shell (double containment) tanks for liquids, salts and sludges at the Hanford Site and the Savannah River and stainless steel bins at the Idaho National Engineering Laboratory (INEL) provide high integrity storage pending final treatment and disposal of high-level waste. At INEL, storage tanks and concrete containment vaults were built about 35 years ago. They are still sound but do not meet current earthquake standards or RCRA secondary containment rules. A major effort is planned, beginning in FY 1991, to design and build four new 500,000-gallon stainless steel tanks for liquid waste at INEL, to replace some of the current ones (53).

Both DWPF and HWVP will include storage capability for vitrified waste, with the ability to add additional storage space until the geologic repository is ready to receive HLW for disposal. Plans for the final HLW form at INEL are still under development. “Shipments to the repository will fulfill DOE’s long-term goal of ending the need for interim storage of high-level waste” (54). “High-level waste is to be immobilized and disposed of using highly reliable isolation technology—a deep geologic repository” (55). Present efforts focus on a site at Yucca Mountain, NV. However, according to a 1989 Nuclear Regulatory Commission (NRC) assessment, such a facility may not be ready until the second decade of the 21st century. According to the 1989 Five-Year Plan, recent EPA restrictions on storage mean that if disposal is not accomplished, operations could be curtailed. As a waste producer, DOE must obtain approval from the NRC to place waste in a repository that will house the Nation’s spent commercial fuel. DOE must also share a cost of the repository (55).

Amount and Distribution

Figures 1-3 and 1-4 show the distribution of HLW at DOE sites by volume and radioactivity. Of the four locations with high-level waste, Hanford and Savannah River have a combined total of more than 90 percent of the waste, as measured by both volume and radioactivity. Idaho (ICPP) has 5.6 percent of the total radioactivity and 2.9 percent of the total volume. The fourth site, West Valley, has the smallest amount, occupying one large carbon steel tank and one small stainless steel tank. However, solidifying this ‘small’ amount, along with decontaminating the old fuel reprocessing facility and converting it to a vitrification plant, will cost about one billion dollars (45).

For purposes of comparison, high-level defense waste is estimated to contain 1.17 billion curies (Ci), some 40 times the radioactivity of commercial HLW but one-sixteenth the radioactivity of commercial spent fuel.“With the exception of 3,400 cubic meters (m³) of solid calcine stored in stainless steel bins at Idaho and a small volume of separated strontium-90 (Sr⁹⁰) and cesium-137 (Cs¹³⁷) in stain-

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14Spent fuel at DOE sites is not included in the definition of defense HLW, nor does the IDB provide any comprehensive information on the amount of defense spent fuel at DOE weapons sites.
less steel capsules stored underwater at Hanford, all HLW is stored in steel tanks. These exceptions amount to more when their radioactivity is considered: 4.8 percent of the total HLW radioactivity at Hanford, Idaho, and Savannah River is found in the Idaho calcine, and 15.1 percent is found in the strontium and cesium capsules.

Some 128,000 cubic meters (339 million gallons) of HLW at Savannah River is stored in carbon steel tanks. The radioactivity is distributed among four phases: liquid, sludge, salt cake, and precipitate, with sludge containing 60.5 percent of the radioactivity.

At Hanford, 149 single-shell tanks contain an estimated 157 million curies of radioactivity. Some of these tanks were initially designed to cascade (discharge) waste into cribs, followed by percolation through soils so that the radioactive material would be retained in the soil. This practice was discontinued after the 1950s, when evaporators

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\(^{15}\)The curie, a conventional unit of radioactivity, is equivalent to 37 billion disintegrations per second.
were built to concentrate the waste. About 66 tanks are believed to have developed leaks. The contents of these tanks, therefore, are subject to more uncertainty than indicated in the IDB. The salt cake has the largest volume, whereas the sludge contains the highest radioactivity. Also at Hanford are 28 double-shell tanks containing 111 million curies of radioactivity, reported in the IDB as ‘slurry.’

Idaho has 7,600 cubic meters of HLW liquid, more than twice the volume of calcine (3,400 cubic meters); however, the radioactivity of the calcine is 56.9 million curies, 5.6 times the radioactivity of the liquid (10.1 million curies). Calcination reportedly reduces the volume of HLW by a factor of eight (73). The volume reduction and radioactivity concentration that occur are indicated by the ratio of liquid-to-calcine volume per unit of radioactivity (12.6 to 1).

West Valley has three types of commercial waste: 1) 50 cubic meters (13,200 gallons) of acid liquid waste from reprocessing fuel containing thorium,
located in one stainless steel tank; 2) alkaline waste consisting of 2,020 cubic meters (534,000 gallons) of liquid and 46 cubic meters (12,200 gallons) of sludge, all in one carbon steel tank; and 3) 13 cubic meters (3,430 gallons) of solid zeolite loaded with cesium-137 and stored underwater.

Inventories of defense HLW, both past (historical) and future (projected), are presented in tabular form in the IDB; they are plotted in figure 1-5. From the end of 1980 through 1988, the accumulated volume of HLW at the sites increased by 30 percent; radioactivity decreased by 10 percent, and thermal power (the rate at which heat is generated due to radioactive decay) increased by 1 percent. The fact that the radioactivity decreased while the volume increased can be explained in part by the decay of short-lived radionuclides. However, other factors may also be involved.

DOE is constantly revising its estimates of the volume and radioactivity of defense HLW as a result of new information that permits either better estimates of already existing HLW or reclassification of HLW to other categories such as transuranic (TRU) waste or low-level waste (LLW). For example, in the 1989 IDB, the volume and radioactivity of the Hanford strontium and cesium capsules were each reduced by roughly 10 percent from values in the 1988 report. The reason given was that over the years, a number of capsules had been dismantled and the contents used outside of Hanford; these will not be returned to Hanford (61). In addition, substantial changes were made to estimates of West Valley waste. Finally, estimates in the IDB include no error bounds to help the reader judge the accuracy of information. In the past, questions have arisen about the inventory of plutonium in the HLW tanks at Savannah River (26). Characterization of the contents of the single-shell tanks at Hanford could yield changes in estimates of their volume, radioactivity and composition.

Projections of HLW through the year 2020 are also included in figure 1-5. Projections are based on a scenario for weapons production dictated by national nuclear weapons material stockpile needs. They assume that three reactors at Savannah River will be restarted during 1989-90 (they were not) and will operate through the year 2000; after 2000 the three reactors will be replaced by one new produc-

Current and Potential Problems

Liquid Tank Storage: Soil and Groundwater Contamination; Tank Explosion

The three DOE weapons facilities with HLW—namely, Hanford, Idaho, and Savannah River—as well as the site at West Valley, all have a number of tanks on site that contain highly radioactive liquids and associated physical forms requiring constant vigilance. Many of these tanks can hold on the order

\[ \text{One of the functions of the ICPP is to recover highly-enriched uranium from spent naval reactor fuel for use in the “driver” fuel elements of the Savannah River production reactor (31).} \]
Figure 1-6: Total Radioactivity of High-Level Waste in Storage by Site Through 2020 (figures projected for 1989 to 2020)
Figure 1-6—Total Radioactivity of High-Level Waste in Storage by Site Through 2020
(figures projected for 1989 to 2020)-Continued

Hanford Reservation

West Valley Demonstration Project

NOTES: a. Glass maybe in storage at the site, in transit to a repository, or in a repository.
b. Capsules contain either strontium fluoride or cesium chloride.

of 1 million gallons (3,800 cubic meters) of waste. Tanks at Hanford and Savannah River have been the object of scrutiny by States, environmental groups, and local citizens concerned with leakage and subsequent environmental contamination, and with the possibility of explosion. Tanks at Idaho, although used mostly for short-term storage prior to calcining the waste, are in concrete containment vaults that do not meet either current design basis earthquake standards or present-day RCRA secondary containment rules (53).

Hanford has 149 single-shell tanks and 28 double-shell tanks, both made of carbon steel as opposed to the more expensive stainless steel. Because of the highly acidic nature of liquid waste leaving the Plutonium Production and Extraction reprocessing plant, the waste must be neutralized to prevent reaction with the tank walls. The neutralization step precipitates sludge at the bottom of the tanks and complicates subsequent transfer. At Hanford, the single-shell tanks were built before the double-shell tanks, when there was less concern with environmental and health effects than exists now; the frost tank was put in operation in 1944 (see table 1-1 for chronology).

According to a 1989 General Accounting Office (GAO) report, DOE officials estimate that from 1959 through 1988, definite or possible leaks had occurred in 66 of the 149 single-shell tanks (5 leaking tanks were identified in 1988), releasing about 750,000 gallons of HLW as estimated by contractor personnel (42). Earlier, liquid waste from some single-shell tanks was deliberately released to cribs from which waste percolated into the soil as part of the disposal process. The first indication of a potential leak occurred in 1956; no new liquid has been added to the single-shell tanks since 1980 (see table 1).

The environmental and health impacts due to the movement of liquid waste from single-shell tanks into the soil are not well established. There is no unanimity about pathways through which the radioactive and hazardous substances travel or their ultimate fate. The GAO summarizes the situation as follows (43):

DOE officials have stated that the environmental impact of the single shell tank leaks will be low or non-existent and have cited several studies as a basis for their assessment. However, we believe the studies do not provide conclusive evidence about the degree of environmental impact attributable to tank leaks. Some studies indicated there would be limited environmental impact, but they did not analyze the impact of several mobile contaminants on Hanford’s groundwater. One study predicted groundwater contamination would exceed safety limits but did not project the impact on the Columbia River.

Information in a recent report by the Hanford Education Action League (HEAL) supports the case that groundwater contamination from a variety of sources on the Hanford reservation reached the Columbia River much more rapidly than was previously believed due to geological channels under the Hanford site or to the presence of organic chemicals that speed migration (11). DOE has yet to complete a comprehensive study of subsurface contamination at Hanford.

Since 1973, DOE’s strategy for limiting single-shell tank leaks has been to remove the liquid and seal the tanks to prevent penetration by liquids such as rainwater. A large volume of liquid has been removed by evaporation or by pumping to double-shell tanks. However, according to GAO, pumping has been delayed in part because of space limitations as a result of tank space being allocated to ongoing production programs. GAO recommended that DOE develop specific plans to place an interim ground surface material over the tank farms to slow water drainage through the soil (44).

In a 1987 DOE final EIS, the preferred alternative for dealing with the Hanford single-shell tank waste was to study the matter further and defer decision (59); this alternative was singled out because the need for action was believed to be less immediate than in other tasks to be performed, given that most of the liquid had already been removed from the tanks and that the remaining sludge and semisolids had limited mobility. The Hanford Federal Facility Agreement and Consent Order and the associated Action Plan of May 1989, entered into by DOE, EPA, and the State of Washington’s Department of Ecology, known as the tri-party agreement, codify deferral of disposal of single-shell tank waste (71), as proposed in the Hanford EIS. According to the tri-party agreement, a full-scale tank waste farm retrieval demonstration is not scheduled until the year 2004, with complete closure of all 149 tanks by 2018 (71). At present, effort appears to be focused on
Table I—I—Chronology of Major Events in the History of Single-Shell Tanks at Hanford

<table>
<thead>
<tr>
<th>Year</th>
<th>Event Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>1944</td>
<td>First single-shell tank went into service.</td>
</tr>
<tr>
<td>1956</td>
<td>First indication is obtained of a potential leak.</td>
</tr>
<tr>
<td>1959</td>
<td>First leak occurred.</td>
</tr>
<tr>
<td>1964</td>
<td>Construction was completed on last group of single-shell tanks.</td>
</tr>
<tr>
<td>1966</td>
<td>Last of single-shell tanks went into service. Total volume of waste in single-shell tanks reached ca. 77 million gallons.</td>
</tr>
<tr>
<td>1968</td>
<td>Construction of first double-shell tanks began.</td>
</tr>
<tr>
<td>1970</td>
<td>Groundwater monitoring well was drilled. Highly radioactive contaminants leaking from single-shell tanks were later detected in groundwater. According to DOE, migration to groundwater most likely occurred during drilling of well but might have been a natural progression through the soil.</td>
</tr>
<tr>
<td>1971</td>
<td>Double-shell tanks became operational.</td>
</tr>
<tr>
<td>1972</td>
<td>Pumping program was begun to transfer liquid from single to double-shell tanks.</td>
</tr>
<tr>
<td>1973</td>
<td>Largest single-shell leak occurred—an estimated 115,000 gallons.</td>
</tr>
<tr>
<td>1980</td>
<td>DOE stopped placing waste in single-shell tanks. Liquid waste levels in single-shell tanks were reduced to no more than 1 foot above solid waste. Plans were adopted to transfer the remaining 8.5 million gallons of single-shell tank waste that could be feasibly pumped into double-shell tanks by 1985.</td>
</tr>
<tr>
<td>1985</td>
<td>Planned pumping schedule was not followed, and scheduled pumping of single-shell tanks ended. Since August 1985, DOE had pumped liquids only from tanks it assumed had leaked, with the exception of about 16,000 gallons pumped from one tank in 1986.</td>
</tr>
<tr>
<td>1988</td>
<td>Five tanks were added to the list of assumed leakers.</td>
</tr>
<tr>
<td>1989</td>
<td>DOE, EPA, and Washington State signed an agreement in which DOE agreed to pump most of the remaining 5.3 million gallons of pumpable liquid waste from the single-shell tanks by the end of FY 1995. However, in accordance with the agreement, two tanks that may be susceptible to excessive heating and require supplement cooling are scheduled to be pumped by the end of FY 1996.</td>
</tr>
</tbody>
</table>

characterizing tank wastes. A National Academy of Sciences panel advises DOE on a continuing basis on single-shell tank waste. At a December 1989 meeting, panel members urged DOE and its contractors to conduct a systems analysis of alternative approaches for treating and disposing of tank wastes.18

High-level waste in Hanford double-shell tanks is to be vitrified, starting in 1999, at the Hanford Waste Vitrification Facility (HWVF); a $550-million contract for construction of the HWVF was awarded in December 1989 to United Engineers and Contractors (72). Waste from these tanks is to be pretreated prior to vitrification. Because there are only 28 of these tanks and they are very expensive to build, DOE is concerned with managing the waste in them to conserve storage space. Pretreatment serves to separate a portion of the original liquid HLW so that it can be treated as low-level waste (i.e., grouted), and to free up tank space for additional HLW. Although double-shell tanks appear to be a technological advance over single-shell tanks, the form (sludge and supernatant liquid) and intense radioactivity of their contents are given priority in the major milestones for disposal of tank waste in the tri-party agreement, which include initiation of pretreatment of double-shell tank waste in B Plant by October 1993 and initiation of HWVP operations by December 1999 (71). In February 1991, DOE proposed a two-year delay in the startup of the HWVP; startup of B-Plant may be delayed until the end of 1997.19

A potential problem that has received widespread public attention only recently is the possibility of explosions in HLW tanks. In the 1950s, potassium ferrocyanide was added to the single-shell tanks at Hanford to precipitate radioactive cesium and strontium so that liquid could be pumped from the tanks to create room for more high-level liquid waste. Ferrocyanide, mixed with nitrites and nitrates in the tanks, can be explosive if certain temperatures are exceeded. A "worst case" scenario, considered in a 1984 Battelle Northwest Laboratories report, indicates that the energy release in such an explosion could be equivalent to 36 tons of TNT (7). Concern about tank explosion has been heightened by reports of a chemical explosion in a nuclear waste tank at Kyshtym in the Soviet Union, that occurred in 1957 and resulted in the evacuation of 10,000 people and reportedly released 20 million curies of radioactive materials (15).

DOE’s position regarding the explosion hazards in the Hanford tanks has been that there is no immediate risk because temperatures in the tanks are well below those at which ferrocyanide explosions might occur.20 However, concerns have been expressed about uncertainties in characterization of the waste and about possible hazard if the waste was mechanically or thermally disturbed (e.g., through in situ vitrification, vitrification in the Hanford Waste Processing Facility, or cutting into the salt cake). Although DOE is studying the situation, further research has been recommended to learn whether any other materials present in the tanks represent an explosion hazard under certain conditions.21

In March 1990, DOE revealed that hydrogen gas has been building up in 20 of the HLW storage tanks at Hanford (19). Hydrogen arises from the decomposition of organic materials placed in the tanks and from radiolysis of water. Although Michael Lawrence, the DOE Hanford facility manager at the time, is quoted as stating that DOE believes the danger of an explosion is low and the potential for radioactive release even lower, according to Lawrence, "the worst case is any explosion that could cause the dome to collapse and send the contents up to the air . . . . I can’t sit here and say it’s not going to happen" (19). John Conway, chairman of the Defense Nuclear Facilities Safety Board, stated after a briefing on the Hanford tank situation in March 1990 that the board considers the danger of a hydrogen explosion potentially more serious than a ferrocyanide explosion (4). However, a subsequent statement in April 1990 quoted Conway as saying, "We don’t believe there is any kind of risk to the public" (18).

In a July 23, 1990 letter to the Secretary of Energy, DOE’s Advisory Committee on Nuclear Facility Safety (ACNFS) stated that: "The (high level) waste tanks are a serious problem. The possibility of an explosion of an unstable chemical (such as ferrocyanide) or a flammable, gas must be taken seriously because of the magnitude of the radioactive inven-

20FD Personal communication during visit to Hanford, November 1989.
tory available for dispersal. Although the ACNFS “found nothing to indicate that emergency action was required,” serious concerns were presented: 1) apparent lack of concern on the part of the operating staff for the tanks about the hazard; 2) apparent lack of attention by top DOE management and the contractor about achieving higher levels of tank safety; 3) an attitude at the DOE Richland Operations Office that because the tri-party agreement exists, waste issues are being efficiently resolved, even though the agreement postpones major decisions on waste handling and risks from operations. The apparent neglect of several factors that may call into question Hanford’s belief that there is “no deflagration or detonation hazard because of the differences between temperatures measured in the tanks and temperatures of the onset of reactions observed in the laboratory tests.” They go onto state that:

The Hanford tanks present a serious situation, if not an imminent hazard . . . much more effort must go into determining what is in the tanks, what is happening in the tanks, and what are the possible reactions that can occur . . .

Uncertainty surrounding the possibility of conflagration and explosion in the waste tanks does need to be resolved as rapidly as possible. The material presented to us at Hanford was weak, but did include the suggestion that the probability of conflagration may be low and that, even if it were to occur, the energy release might not rupture the tanks. However, the available information, the analyses and the experiments that have been done, leave wide margins of uncertainty.

Until this uncertainty is resolved, the ACNFS recommends establishing continuous monitoring and action plans for coping with the event of excessive pressures or temperatures, or for a release.

A significant airing of the tank explosion issue took place at a hearing on “Accident and Explosion Risks at Department of Energy High-Level Radioactive Waste Facilities” by the Senate Committee on Governmental Affairs on July 31, 1990. One of the witnesses, the DOE Director of the Office of Environmental Restoration and Waste Management, Leo Duffy, indicated that additional steps being taken by DOE include formation of a Headquarters Tank Advisory Panel to review waste tank technical issues at all DOE sites, and formation of a Senior Chemists Panel for Hanford to review waste chemical reactions. Two additional statements by Duffy shed light on DOE’s ambivalent attitudes toward external oversight as well as the apparent seriousness with which they now view the tank explosion issue:

All the oversight in the world will not do the job that we must do ourselves. We have had independent reviews of various pieces to this puzzle-first, the presence of ferrocyanide in the tanks, followed by the discovery of hydrogen gas generation, followed then by the realization that nitrous oxide was present in the tanks as well-all of which indicates a material weakness on the part of DOE and its contractors to understand the fundamental chemistry present in its HLW tanks.

As slow as we may have been to uncover these events and then link them together, our ‘discovery’ was not made with the help of the technical safety appraisals, which failed also to identify the potential hazards posed by the presence of these chemicals and failed to add to our understanding of the potential seriousness of the chemical reactions that were taking place.

... in my opinion, the issue of accident potential in the high-level waste tanks would not have surfaced without your (Senator John Glenn) question on ferrocyanide in the single-shell tanks. This question resulted in the subsequent examination of hydrogen generation, the presence of nitrous oxide and our need to understand tank chemistry issues, and the general lack of discipline and follow-up needed to resolve long-standing technical issues in the Hanford tank farm. It resulted also in the evolution of methods used in Technical Safety Appraisals, safety audits, and Tiger Team Assessments. I want to express to the Chair my appreciation to this Commit-

23Ibid.
24Ibid., pp. 5-6.
25Ibid., pp. 6-7.
27Ibid., pp. 15-16.
Long-Lived Legacy: Managing High-Level and Transuranic Waste at the DOE Nuclear Weapons Complex

...and to its staff for helping us to begin resolving these potentially serious issues.28

In October 1990, the Defense Nuclear Facilities Safety Board (DNFSB), which was established by Congress to provide oversight of DOE activities, indicated in a letter to DOE Secretary Watkins that DOE and Westinghouse Hanford Co. were not moving fast enough to implement safety measures at the Hanford tanks.29 These measures include installing new instrumentation, accelerating tank sampling, and developing plans for dealing with possible explosions.

The tank situation at Savannah River has received less media attention than that at Hanford. Savannah River has the largest amount of radioactivity of all sites in the form of HLW. According to the FY 1989 Savannah River Waste Management Plan, there are 51 large, subsurface tanks for storing and processing aqueous HLW (64). All are made of carbon steel and there are four different designs; none of the tanks appears to be equivalent to the Hanford single-shell tank. According to DOE, some leakage of tanks has occurred along with rusting of tank walls (64). A program to transfer waste from older tanks to newer, Type III tanks, which are believed to have good near-term integrity, is scheduled to be completed through 1995. There are 27 of these double-shell Type III tanks (64).

Problems with the HLW tanks at Savannah River figured prominently in a 1986 report (26) by the Environmental Policy Institute (EPI), namely concern about: 1) contamination of the shallow aquifers beneath Savannah River; 2) possibility of tank explosions; 3) potential threat of an earthquake, which the tanks were not designed to withstand (in an area that had a large earthquake in 1886); and 4) excessive radiation exposure of personnel working around the tanks or dealing with certain aspects of the DWPF vitrification plant when it begins operation (26). The highly critical tone of the EPI assessment contrasts with DOE descriptions of Savannah River HLW operations.

In EPI’s 1987 reply to Du Pent’s response to its Savannah River critique, the report states that the buildup of organic vapors in tanks has a larger likelihood of occurring than does buildup of hydrogen (27). Furthermore, EPI finds Du Pent’s estimates of the probability of a tank explosion to be considerably lower than its own (27). In October 1990 the condition of the tanks at Savannah River and the possibility of explosions and fires were being studied by a variety of groups including the Advisory Committee on Nuclear Facility Safety.

According to the 1989 Five-Year Plan, INEL’s HLW tanks, although stainless steel and still sound, are 35 years old and their concrete containment vaults do not meet either current earthquake standards or RCRA secondary containment rules. As a result, DOE plans in FY 1991 to design and eventually construct four 500,000-gallon tanks to replace five of the eleven 300,000-gallon tanks currently storing HLW at the Idaho facility (53); the new tanks are to be put into operation by FY 1997 (56). This appears to be the only facility in which new HLW tank construction is planned. If shutdown of the calcination facility continued for a long time, the liquid tank storage capacity required would increase.

In summary, HLW storage tanks represent potential sources of radioactive releases to the soil-and, hence, to water under the site and eventually off-site—or of more widespread release by explosion or natural disaster such as an earthquake. Many tanks were not designed for long-term storage, and the use of carbon steel necessitated neutralizing the waste, which has resulted in complex mixtures of liquid, sludge, and salt cake that are difficult to move and represent a potential hazard to those involved in sampling tank contents or in other operations. Ironically, new tanks are to be built at the one location of three in the Weapons Complex that has the smallest amount of HLW, where stainless steel rather than carbon steel tanks are present, and where calcining is used to solidify HLW, namely, Idaho. By contrast, at Hanford, no decision seems near on how to dispose of single-shell tank waste. A lot is riding on the success of the vitrification plants at Savannah River and Hanford to reduce the risk posed by high-level tank wastes.

Reprocessing Plants: PUREX

The high-level radioactive waste in interim storage as liquid slurry or calcine is generated by the reprocessing of spent fuel and irradiated targets. Before reprocessing, most of the radioactivity is associated with and contained in the confines of the solid spent fuel and targets. To recover plutonium...
for weapons use, these solids must be chopped up, dissolved, and processed with both organic and aqueous solvents. The net result is a huge increase in the volume of radioactive waste. Figure 1-7 illustrates some of the liquid streams from the Plutonium and Uranium Extraction (PUREX) plant at Hanford; the PUREX process with appropriate modifications has generally been adopted as the standard for reprocessing nuclear fuel, both in the United States and worldwide.

Some indication of the potential environmental impact of the PUREX plant is given by the following (40):

Reprocessing plants like PUREX and its predecessors, which recover plutonium from irradiated uranium, have been responsible for some of the worst environmental contamination at Hanford because they generate huge volumes of toxic chemical and radioactive wastes. The process that produces one kilogram of plutonium at PUREX also produces over 340 gallons of liquid high-level radioactive wastes mixed with hazardous chemicals, more than 55,000 gallons of low- to intermediate-level radioactive wastes discharged to cribs, and over 2.5 million gallons of cooling waters disposed to ponds.

Some advocate reprocessing fuel to recover and recycle transuranic radionuclides as a way of reducing the radioactive waste disposal burden. According to this approach, with recycling, long-lived transuranics such as plutonium-239 are contained within the reactor, reprocessing plant, and fuel fabrication plant or their immediate vicinity. Therefore, the waste to be disposed of contains fewer long-lived radioisotopes. However, reprocessing usually expands the volume of waste to be dealt with, and long-lived radionuclides are not totally eliminated by transmutation in the reactor.

In December 1989, the PUREX facility at Hanford was shut down in “mid-campaign,”30 that is, with highly radioactive materials dispersed throughout the system. Some 20 metric tons of material was present in the dissolvers and 70 metric tons elsewhere in the system.31 PUREX, which is more than 30 years old, went through a long shutdown period in the 1970s and early 1980s (9). In November 1989, Westinghouse and DOE officials at Hanford were hoping to perform a “stabilization” run on the

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30The term “campaign,” as used by DOE, means operation of the facility to process one batch of fuel from start to finish.
31Personal communication during visit to Hanford, November 1989.
system (i.e., operating the system to clean out material dispersed through it) as soon as possible, followed by a campaign to reprocess the 2,100 metric tons of fuel, including 350 metric tons of weapons-grade material awaiting reprocessing.\(^\text{[33]}\) Obstacles to restart included environmental regulation, concerned citizens groups, and the age and condition of the facility.

Concerns about PUREX reported in the media or raised by environmental and citizen groups include the following:

1. Some reports claim that radioactive liquid effluent streams from the plant continue to be discharged into the soil, even though the fate of radioactive and hazardous materials in these discharges is not well known. The following material is from Associated Press (17): “According to the Hanford Education Action League, PUREX discharges up to 9,000 gallons of liquid per day (into the soil) when operating; even in a standby (shutdown) condition, it discharges 4,000 to 5,000 gallons per day into the soil. A DOE spokesman maintains that continued liquid flow is necessary during ‘idle’ for safety reasons. Although PUREX is only one source of liquid discharges at Hanford, it is the ‘biggest single liquid waste generator.’” Again, according to the Hanford Education Action League, of the 33 liquid waste streams identified at Hanford, the 19 most radioactive must be stopped, stored, or treated by June 1995 (in compliance with the tri-party agreement). A Hanford report estimates the cost to prevent dumping of liquid wastes into the soil to be $244 million.”

An article in October 1989 (40) reported that after PUREX was built in 1956, it took only 7 years for a radioactive tritium plume from its operations to reach the Columbia River, some 9 miles away; that plume results in about 4,000 curies of tritium entering the river annually, according to DOE-contractor water monitoring reports.\(^\text{[32]}\) Originally, it was expected to take 175 to 180 years for contaminated groundwater to travel to the Columbia River; the fact that movement is an order of magnitude more rapid is perhaps due to channeling effects under the Hanford site or to the presence of organic chemicals that speed migration.\(^\text{[35]}\)

2. An Associated Press article in December 1989 stated that shutdown of the plant in 1989, during a campaign, necessitated a stabilization run to blow out material that had settled in pipes and other equipment. A concern was that radioactive and hazardous liquids might be discharged if the plant were started up in this condition, with materials distributed throughout the system, especially if any equipment were to fail. Because of the age of the plant, equipment failure has been common in recent years. In December 1989, DOE and Westinghouse-Hanford began a “phased restart to stabilize chemicals” (75).

3. The ACNFS has expressed concern about the high turnover of workers and management at PUREX that could lead to a potential safety issue when the plant is restarted.\(^\text{[36]}\)

4. There is also some concern that DOE and Westinghouse-Hanford have not always provided information on occurrences at the plant in a timely and accurate fashion.

5. Finally, some have stressed a number of regulatory issues that center on hazardous waste streams in the PUREX facility, such as the use, treatment, and disposal of “listed wastes” (e.g., acetone, n-butyl alcohol, xylene, and toluene). These issues are important in considering the restart of PUREX.

Support for the restart of PUREX offered by DOE and DOE-contractor personnel at an OTA meeting at Hanford in November 1989 was based on the

\(^{32}\)Ibid.


\(^{34}\)Tritium is not produced deliberately by irradiation of lithium targets in the Hanford reactors. However, it is produced by fission in the fuel. Some tritium is released as a gas in the dissolution process, whereas the remainder follows the aqueous reprocessing stream and is released to the environment as tritiated water vapor or liquid. See M. Benedetti, T.H. Pigford, and H.W. Levi, Nuclear Chemical Engineering, 2d ed. (New York, NY: McGraw-Hill, 1981), p. 357.

\(^{35}\)For example, a concentrator failure led to an extended outage; it was one of several factors involved in the suspension of PUREX operations during 1989. Source: Viewgraph from briefing by D.H. Shuford, “PUREX Status, Plans and Issues,” Nov. 17, 1989. In addition, the New York Times, on Dec. 16, 1989, reported that on Dec. 3, 1989, an unexplained chemical reaction in a pipe of the shutdown PUREX plant “ruptured a gasket and sprayed acid into an area sometimes occupied by workers.”

following: Spent fuel from the N reactor awaiting reprocessing is being held in water basins at K plant, just one-quarter mile from the Columbia River. These basins have leaked in the past. The metallic fuel elements sustained some mechanical damage in being unloaded from the N reactor into storage pools; some 6 to 10 percent of the fuel is estimated to be in a failed condition.\textsuperscript{37} In this condition, radionuclides can be released to the water basins which, if they were to leak again, could cause groundwater and river contamination.

In March 1990, DOE provided the following information concerning the PUREX stabilization run and proposed restart.\textsuperscript{38}

PUREX was restarted on December 17, 1989, to complete a processing run that was interrupted on December 7, 1988. The processing run was successfully completed March 1, 1990. Only minor operational/equipment problems were encountered during the run. PUREX is now scheduled for an extended outage of approximately one year to prepare for the processing of the irradiated N reactor fuel now in storage. Activities scheduled during the outage include an inventory of special nuclear materials, maintenance and repairs, and the construction of waste disposal facilities. An Environmental Analysis (EA) will be issued in March, 1990. The EA will determine whether the previous environmental impact statement for PUREX needs to be updated.

A July 1990 analysis of whether or not to restart PUREX was prepared by the Institute for Energy and Environmental Research (IEER) for the Hanford Education Action League (HEAL).\textsuperscript{39} In a subsequent article, the situation is described as follows:\textsuperscript{40}

The environmental problem posed by the fuel is real enough. About 3-7 percent of the N-reactor fuel is damaged and is being corroded by contact with water. The spent fuel in K-West is sealed inside water-filled containers which contain the radioactivity, but the fuel elements in K-East are stored in open cans, and the water in the basin is highly contaminated. Workers do not enter the pool area without special radiation protection equipment. The pool itself leaked before it was repaired in 1980, and Westinghouse estimates that some 15 million gallons of water contaminated the surrounding environment with up to 2,500 curies of strontium 90 and cesium 137.

The IEER-HEAL report’s preliminary short-term recommendations were: 1) PUREX should not restart because the hazards of greatest consequence appear to be connected with reprocessing as opposed to other N-fuel management options; 2) to minimize the risks of continued storage of N-fuel in the K-basins, exposed N-fuel should be encapsulated as soon as possible; and 3) preliminary design of dry storage facilities for interim management of N-fuel should also begin as soon as possible.\textsuperscript{41}

The decision as to whether or not to restart PUREX is a significant one. It depends on factors such as the need for additional plutonium for the U.S. weapons stockpile and the desirability of keeping some form of production mission for the Hanford site. In October 1990, DOE Secretary Watkins announced his decision not to restart the PUREX plant for at least 2 years.\textsuperscript{42} During that 2-year period, a study will be conducted of environmental issues associated with PUREX. Although the option of restart after 2 years still seems to be left open, some observers interpret the decision as indicating that PUREX will never again operate.\textsuperscript{43}

Major chemical processing-reprocessing facilities also exist at Savannah River and INEL. Late in 1989, the ICPP, which processes fuel for naval reactors, was put on “temporary standby” because of concerns about underground piping leading to storage tanks. The piping is single-walled, and RCRA requires secondary containment such as double-walled piping. In addition, the New Waste Calcination Facility (NWCF) at the ICPP had not operated since October 1988. Calcination was to have been resumed, after completion of a “dissolution campaign,” on July 9, 1989, but was postponed pending

\textsuperscript{37} Information for this paragraph was obtained from DOE and DOE-contractor personnel during a trip to Hanford in November 1989.

\textsuperscript{38} S.P. Cowan, op. cit., footnote 1.


\textsuperscript{41} Ibid.


cessium capsules and 597 strontium capsules are stored in a water basin, pending additional packaging and disposal in a repository (61). The half-lives of beta-emitting strontium-90 and beta- and gamma-emitting cesium-137 are 28 and 30 years, respectively; thus disposal for several hundred years rather than tens of thousands of years should suffice to reduce the radioactivity to acceptable levels.

IDB 1989 estimates of the volume of strontium and cesium capsules stored at Hanford were reduced by roughly 10 percent over the previous year “to reflect the fact that over the years 43 strontium capsules and 227 cesium capsules have been dismantled and put to beneficial use outside Hanford. These radionuclides will not be returned to Hanford” (61). Even though no problems have been reported about the capsules stored at Hanford, some concerns have been raised about those that have been shipped elsewhere. For example, an Atlanta Journal article states that (39): Some 252 cesium capsules had been used by Radiation Sterilizers, Inc. (RSI) to sterilize medical supplies. That operation has been shut down since June 1988 when a leak was detected in one of the stainless steel capsules. Some 159 of the capsules were still in the RSI building. Removal of the capsules to Hanford was halted when cracks were found in nine of the lead-lined containers used to ship the capsules. DOE is reported to have assumed responsibility for removal of all capsules to Hanford, in contrast to the impression given in the IDB. That removal involves transporting the capsules from DeKalb County, Georgia to Richland, Washington.

The Atlanta newspaper account emphasizes that the situation at RSI poses no public danger. It highlights the tension that has arisen between officials of the Georgia Department of Natural Resources and DOE officials at Hanford. It also contains the following statement: “DOE officials now acknowledge that the cesium-fried capsules should never have been used for commercial purposes’ (39). An October 1988 Office of Technology Assessment (OTA) report concludes that theft and improper handling of sealed radiation sources have been responsible for 14 deaths and 4 major accidents...
in foreign countries over the last 25 years (47). One of these accidents, which occurred in Goiania, Brazil in 1987 and resulted in four deaths plus widespread contamination, has been called the second worst radiation accident in history (37). The situation in Goiania was caused by the removal of a stainless steel cylinder containing 1,400 curies of cesium-137 from a cancer therapy machine in an abandoned clinic (37).

Given the situation that has arisen at RSI and concerns about more serious problems, it appears that the strontium and cesium capsules at Hanford will probably not be destined for future beneficial uses but will remain as waste destined for geologic disposal. In 1990, DOE asserts that all capsules are accounted for. Capsules which are not disassembled for use as material sources are to be returned for storage pending eventual treatment prior to disposal in a geologic repository.47

In response to a follow-on question by OTA, DOE indicated that some strontium and cesium capsules were sent to the Office of Isotope Sales operated by Oak Ridge National Laboratory (ORNL). These capsules were disassembled and the isotopes sold to commercial ventures. Once sold, “regulating responsibility and oversight is transferred to the Nuclear Regulatory Commission. Disposition and accountability are then no longer the responsibility of DOE.”

Delays and Uncertainties About Vitrification

In November 1989, GAO completed a study of DOE’s program to prepare HLW for final disposal (45). GAO found that the Defense Waste Processing Facility for vitrifying HLW at Savannah River was 2 years behind schedule, and the West Valley Demonstration Project, also for vitrifying HLW, was 8 years behind schedule. The DWPF at Savannah River is now scheduled to begin operation in FY 1992.51 The DWPF has been under construction for several years and has benefited from a series of tests on smaller scale equipment as well as full-scale operation of similar technology in Europe. Even so, the DWPF is a very complex facility of the type that usually encounters startup problems. Further delays and uncertainties may be expected at DWPF. Delays have also been encountered at West Valley. Although it has been possible to utilize some existing facilities left over from the time of West Valley’s fuel reprocessing operations, decontamination added to the burden, as opposed to the entirely new building at Savannah River (70). It has also suffered continually from budget shortfalls.

DOE has selected a particular waste package for DWPF and WVDP, namely, borosilicate glass in relatively thin-walled (3/8-inch-thick) stainless steel canisters. If vitrification proceeds according to schedule, some waste canisters will be produced and stored long before a repository exists to house them. Providing such storage at vitrification sites should not be a major technical problem. Storage for about 5 years operation of the DWPF has already been constructed at Savannah River. Either in on-site or monitored retrievable storage, the waste form should guarantee isolation from the environment for a thousand years or more if long-term institutional controls are in place and the waste form performs as designed.50 The reported design lifetime of the DWPF canister is more than 8,000 years.51

According to present U.S. plans, no credit is allowed for the canister as a barrier to contact between the waste and the geologic repository in considering the acceptability of a waste package. The waste canister’s role is to “encapsulate the waste glass during on-site storage, shipment, and temporary storage at the repository prior to overpacking and final disposal.”52 The canister must be able to withstand a drop of 7 meters without failure and must fit in the repository overpack container, as specified by the NRC.

The U.S. approach of placing no reliance on engineered barriers is in marked contrast to the Swedish approach to waste disposal. An evaluation of the latter by the National Academy of Sciences states that utilizing thick-walled canisters should enable the life of the waste form package to be

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47 S.P. Cowan, op. cit., footnote 11.
48 Ibid.
49 At an October 1990 visit to the DWPF, OTA personnel were told that operations with radioactive waste might not begin until FY 1993 or 1994 due to budget uncertainties.
50 Should the repository delayed, however, DOE would need to consider the issues of longer-term institutional controls and local public acceptance of on-site storage.
51 S.P. Cowan, op. cit., footnote 11.
52 Ibid.
extended to more than one million years (28). Some thought is also being given in the United States to placing more reliance on the engineered waste form. One issue for future inquiry identified in the first annual report of the Nuclear Waste Technical Review Board (NWTRB) to Congress concerns the relative importance of natural and engineered barriers. Using longer-lived engineered barriers means that less reliance would have to be placed on geologic barriers or mathematical models of geologic performance when making licensing decisions (34).

If this were to occur, modification of existing designs might be required. Because the primary focus of the repository is commercial spent fuel, the NRC overpack container might have to be redesigned. This might have no effect on defense HLW canister design or it could necessitate changes. Current DOE plans and NRC requirements do not involve changing the waste canister, overpack, or container. If changes were anticipated, however, they would need to be balanced against the increased risk of delays in proceeding with vitrification of HLW tank contents.

Releases to the Atmosphere

The facilities involved in generating HLW—namely, nuclear reactors and reprocessing plants—as well as the facilities used to treat them, such as evaporators and calciners—sometimes release materials to the atmosphere. Early in the weapons program at Hanford, large releases of volatile fission products occurred when fuel was dissolved. More than 500,000 curies of iodine-131 was reportedly released to the air between 1944 and 1957 (41). Through the introduction of air filters and other off-gas handling equipment, releases of radionuclides have been reduced but not totally eliminated. At Savannah River in 1984, 1.7 million curies of radioactivity was released routinely into the atmosphere, most of which was tritium (790,000 curies) and krypton-85 (840,000 curies) (13). Accidental releases have also occurred. Fortunately, none has been as large as the Kyshym radioactive waste tank explosion in the Soviet Union, in which an estimated 20 million curies, roughly 40 percent of the radioactivity associated with the Chernobyl accident, was released (15).

Several panelists at an OTA Health Effects Panel Workshop in January 1990 stated that airborne release of both radioactive and hazardous materials could be a greater potential health threat than groundwater contamination. They also pointed out that the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA, “Superfund”) regulations focused attention on ground-water contamination but ignored air releases and that promulgation of standards for the release of radionuclides to air has become an object of contention between NRC and EPA.

TECHNOLOGIES FOR IMPROVED MANAGEMENT OF HIGH-LEVEL WASTE

Introduction

Efforts are underway to improve the management of HLW at the three major DOE sites and at West Valley. A principal objective is to convert the liquid and semisolid HLW now stored in aging tanks to solids that can be immobilized and dispersed throughout a rigid matrix material, encased in canisters, and placed in a deep geologic repository. This would eliminate the threat of groundwater contamination from tank leaks or radioactive releases from tank explosions. The matrix material selected by DOE is borosilicate glass and the process of choice is vitrification. At the Idaho National Engineering Laboratory (INEL), some liquid waste has already been solidified into a powdery form by a process known as calcination. INEL is considering a variety of solid waste forms besides borosilicate glass to immobilize the calcine for final disposal.

Three of the facilities that manage HLW pretreat55 it to some extent to reduce the volume that must be stored as liquid and then vitrified, and hence to

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53 The NWTRB was established by a 1987 amendment to the Nuclear Waste Policy Act of 1982, as an outside panel of experts appointed by the President to review decisions of DOE’s Office of Civilian Radioactive Waste Management (12).

54 DOE provided the following information on standards for air releases from reprocessing and other operations: DOE requirements for controlling and reporting air releases are set forth in various DOE orders; emissions of both radioactive and nonradioactive air pollutants must be maintained as low as reasonably achievable. DOE orders also specify the need for compliance with local, State, and Federal clean air laws or regulations where they apply. The National Emissions Standards for Hazardous Air Pollutants (NESHAPS) in 40 CFR 61 apply to radioactive emissions from all DOE sites, and the Prevention of Significant Deterioration (PSD) regulations in 40 CFR 52 cover nonradioactive pollutants. In general, NESHAPS requirements are enforced by EPA regional offices, whereas PSD regulations are enforced by State agencies. Source: S.P. Cowan, op. cit., footnote 11.

55 The term pretreatment is used in this paper to mean those steps taken to reduce the volume of HLW to be vitrified; it does not include calcination.
reduce waste management costs. A variety of technologies are used to treat and dispose of the ‘non-high-level’ fractions of what started out as HLW. Prominent among these are grouting (immobilization in grout or concretes), followed by near-surface or at-surface disposal.

Finally, some effort is underway at DOE to consider how waste minimization might be applied to all aspects of waste management operations. HLW minimization is discussed later in this section.

Vitrification

Vitrification or, alternatively, classification, as it is to be carried out at the Defense Waste Processing Facility (DWPF) at Savannah River, is a process in which high-level radioactive waste, after removal of mercury, aluminum, and other selected nonradioactive components, is mixed with ground borosilicate glass and sent to a melter that operates at 2,100 degrees Fahrenheit (74). The glass-waste mixture is transformed into molten glassified waste by the melter, which operates at a rate of 228 pounds per hour. The molten glassified waste is poured into stainless steel canisters with 3/8-inch-thick walls, in which the mass cools and solidifies into a hard glasslike substance, trapping the radioactive materials inside.

Each of the large Savannah River canisters weighs 1,100 pounds, and is 2 feet in diameter and almost 10 feet high. Each canister holds about 3,700 pounds of glass, of which approximately 94 pounds will be HLW; the radioactivity of the waste in an individual canister will be as high as 234,000 curies, generating heat at a rate of 700 watts (74). The canisters will be sealed, welded tight, and stored in a building near the classification plant pending shipment to a geologic repository. It is estimated that 6,000 to 8,000 such canisters will be required to hold existing and projected waste at Savannah River (74).

In 1983 the decision was made by DOE to adopt borosilicate glass as the waste form of choice for solidifying and immobilizing HLW at Savannah River (46). This decision, along with subsequent decisions to use the same vitrification process and waste form at West Valley and Hanford, has been the basis for major investments in the DOE Nuclear Weapons Complex. The Defense Waste Processing Facility has been constructed at Savannah River at a capital cost of about $930 million\(^\text{57}\) and is scheduled to begin operation in FY 1992.

Vitrification with borosilicate glass will be used in the West Valley Demonstration Project to immobilize HLW from commercial fuel and some Hanford fuel reprocessed at that location 20 years ago; after a series of delays, this plant should begin operating in FY 1995 at a cost on the order of one billion dollars, including decontamination and other operations (45). In November 1989, a $550 million contract was awarded by DOE for a third borosilicate glass vitrification facility, the Hanford Waste Vitrification Plant (HWVP), which is estimated to cost $1.4 billion when completed and will begin vitrifying waste around the turn of the century (6).

These plants, if successful, could go a long way toward eliminating the threat presented by HLW tank storage. In addition to immobilizing the waste by locking or “fixing” the radionuclides in a glass matrix, they substantially reduce the volume of waste (although not the radioactivity) and, if the waste package performs as designed, could shift concern about contamination from the present to hundreds or thousands of years in the future. Thus, a great deal depends, both substantively and in terms of financial investment, on the success of the HLW vitrification efforts at the Savannah River, West Valley, and Hanford sites. Furthermore, although DOE and its critics may disagree about specific technical decisions or factors, a general consensus is emerging that the move from liquid to solidified HLW is a good one.

Concerns about borosilicate glass vitrification fall into two main categories: 1) those that question the original decision to use borosilicate glass as the waste form and 2) those that accept the waste form decision and focus on improving the process. The original waste form decision is not discussed here, but is treated later in considering the choice of waste form at INEL, the one site that stores HLW as both liquid and solid (calcine) and has not yet selected a final waste form for repository disposal.

A long-time observer of the vitrification process has provided some insight into what might make it

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\(^{56}\) Despite the different terms used at different DOE facilities, this immobilization generally involves mixing the waste with cement and letting the product solidify. Terms used for the product are “saltstone” at Savannah River, “grout” at Hanford, and “cement” at West Valley.

\(^{57}\) Personal communication from DOE to R.P. Morgan during visit to DWPF, Savannah River Site, Oct. 26, 1990.
work.\textsuperscript{58} The French developed a vitrification process 20 years ago based on early work in the United States. Subsequently, the British bought and adapted the proven French process but the United States decided to go its own way. Two fundamental differences between the United States and the French approaches are: 1) the United States will use a ceramic melter, whereas the French use a metallic melter, and 2) the French employ three key steps one after the other---chemical adjustment to destroy free acid, calcining to drive off moisture, and incorporation with glass; the United States, however, incorporates all three steps in one operation within the melter itself. The ceramic melter would appear to have advantages over the metallic one in terms of operating temperature and useful lifetime. However, performing all three operations in the melter offers a difficult technical challenge; if something were to go wrong, difficult repair work could lead to considerable delay compared with the French approach. One specific problem that must be guarded against is plugging of the melter due to settling of heavy, noble metals (palladium, platinum, etc.) that can short out the electrodes.

Concern has been expressed that the waste processing facility at Savannah River will be operated without sufficient pilot-plant experience, and arguments have been made for running the plant initially like a pilot plant, with sufficient instrumentation to fully understand and carefully evaluate the operation\textsuperscript{(1)}. DOE contends it has sufficient experience, particularly with a key piece of equipment—the melter—to achieve successful operation.\textsuperscript{59} The only question still to be resolved, according to the 1989 DOE Research, Development, Demonstration, Testing, and Evaluation Plan (48), was how to dispose of the melter, a large piece of equipment that will operate for only 2 or 3 years and become highly radioactive. In October 1990 a storage vault was being constructed for the melter.\textsuperscript{60}

DOE has provided some insight into developmental decisions and work on the DWPF as follows:\textsuperscript{61} Savannah River built and operated a l/10th-scale joule-heated radioactive melter in 1977 and a half-scale melter in 1979. In 1980, a slurry rather than calcine was chosen as feed material, based on savings achieved through elimination of the calcine and significant canyon space reduction. A second l/10th-scale DWPF melter began operating in 1988 to test the full system. A replica half-scale melter was built and operated at Savannah River between 1980 and 1983, and a second half-scale melter was tested with simulated waste from 1986 to 1988.

According to Savannah River Site personnel, the largest melter that had been demonstrated with radioactive materials was l/100th scale. Overall system tests with radioactive materials had been demonstrated at l/200th scale; these tests established the equivalence of radioactive and non-radioactive processing. Larger-scale radioactive tests were not pursued because of cost; the smaller-scale tests fit within existing high-level radioactivity cave cells. The l/10th-scale Integrated DWPF Melter System (IDMS) (nonradioactive) was started up in December 1988. Tests involving mercury removal began in December 1989, and work on noble metals started in June 1990; deposition of noble metals has caused problems in German and Japanese tests but problems are not expected at DWPF. The IDMS will be operated during startup of the DWPF to test system elements and serve as an “early warning system” for possible problems. Some modifications of the DWPF design were made as a result of IDMS tests.\textsuperscript{62}

West Valley has operated a l/6th-scale melter at temperature (but without radioactivity) for 5 years, producing about 100 canisters of glass logs.\textsuperscript{63} WVDP officials also state that in one way or another, they have tested the whole system, including a l/6th-scale test of the tricky sludge mobilization step. The West Valley melter is about half the size of the Savannah River melter. In the last year or two, increasing exchange of information has occurred among DOE’s HLW vitrification projects, facilitated by a technical review group and a “glass producers’ club.”

\textsuperscript{513A} Schneider, Georgia Institute of Technology, personal communication to R.P. Morgan, March 1990.
\textsuperscript{59} Personal communication during high-level waste briefing, U.S. Department of Energy, Germantown, MD, October 1989.
\textsuperscript{60} Personal communication during visit of R.P. Morgan and P.A. Johnson to DWPF at Savannah River Site, October 26, 1990.
\textsuperscript{61} S.P. Cowan, op. cit., footnotes 11 and 46.
\textsuperscript{63} Personal communication during trip to West Valley Site, Feb. 21, 1990.
Vitrification plants are large facilities and will handle very large amounts of radioactivity. Some of the operations will involve transferring molten liquids, transporting slurries of radioactive waste, and mixing waste with glass. Some equipment will become highly contaminated, with radioactivity. Equipment breakdown could occur. Given the nature of the facility, the health and safety of workers involved in plant operations—both routine operation or maintenance and unanticipated shutdowns or emergency procedures—must be protected. According to DOE, all major operations will be performed by remote control, and if equipment breaks down, it will be repaired in place with mechanical arms or removed and replaced with an overhead crane. Thorough equipment testing; reinforced concrete walls separating the process, maintenance, and control areas to provide radiation shielding; and personnel training during 2 years of simulated (nonradioactive) operation are some of the elements being employed to ensure worker safety and protection from radiation (74).

Requirements for interim storage of glass canisters at Savannah River have been based on certain assumptions about an opening date for a deep geologic repository (see figure 1-8). Figure 1-8 was prepared in April 1989; as of March 1990, some changes had been reported by DOE. The start date had been delayed from January 31 to June 30, 1992; production rates had been reduced from 800 to 400 canisters per year from 2012 through 2020. Furthermore, shipments to a Federal repository are now not anticipated by DOE and NRC to begin before 2010. Therefore, HLW storage capacity may need to be expanded beyond that currently planned. The one existing Savannah River storage building cost $55 million in 1983 dollars. At West Valley, glass logs will be stored in a decontaminated cell area of the old plant. Source: Personal Communication during trip to West Valley site, Feb. 21, 1990.
signed to last 1 million years or longer, compared with several hundred to 1,000 years for the U.S. canister. The United States could also consider increasing the design lifetime required for the canisters or overpack containers. Currently, no standards are in place for long-term disposal of HLW since previous Environmental Protection Agency (EPA) standards were remanded by a court in part due to concern that disposal standards for HLW were not consistent with more restrictive drinking water standards. Further, a recent National Academy of Sciences report questions whether HLW disposal standards can be met with the current approach.  

If canisters or overpack containers with a longer design lifetime prove to be an attractive option, one question is when they should be redone. At this time, it is not clear whether canister designs for one or more of the three planned vitrification facilities can be changed, without considerable effort and cost. Certain approaches could be studied, however, such as keeping the existing canisters but redesigning the overpack containers to provide greater assurance of long-term isolation of waste via engineered barriers.

Finally, the management structure for the vitrification activity could be questioned. One major corporation, Westinghouse, with its associated companies, is the contractor for work at all four HLW vitrification sites. This should facilitate communication and result in the experience gained at one facility being readily available to the others; in fact, cost projections for the Hanford Waste Vitrification Facility (HWVF) assume savings based on utilizing experience gained at Savannah River. However, this also means that one company has a monopoly on the technology and might be less willing to innovate and less receptive to learning about advances in vitrification technology outside the United States.

Calcination

Calcination of HLW has occurred at the Idaho National Engineering Laboratory periodically for over 25 years. This is a process in which liquid waste solutions are sprayed as a fine mist into a vessel containing heated granules about the size of coarse sand. The granules and waste solution are brought in contact with air that flows through and circulates the material in the vessel, an operation known as fluidization. In the hot fluidized bed, heat evaporates water and deposits dissolved aluminum and fission product nitrates as coatings on the granules. Small fragments chip off from the granules during agitation as particle size increases. Some fragments are carried aloft where they enter an off-gas cleanup system consisting of scrubbers, silica gel absorbers, and falters. Other fragments remain in the vessel to nucleate new granules. The solid, nonfragmented granules, or "calcine"—a dry, white, powdery substance that contains most of the radioactive material—is blown by air through a shielded underground tube to be stored in stainless steel bins inside reinforced concrete vaults (73).

Calcination began in the Waste Calcining Facility (WCF) in December 1963, following developmental work at two pilot plants in the 1950s; the WCF operated intermittently until March 1981, calcining approximately 3.9 million gallons of radioactive liquid waste. In October 1982 the New Waste Calcining Facility (NWCF) began operating; this facility, built at a cost of $92 million, can handle 3,000 gallons of waste per day. It has calcined at least 2 million gallons of HLW since it began operation (73). In December 1989 the NWCF, which had not operated since October 1988, was placed on a longer “temporary standby” condition, pending a review of ways to bring single-wall piping from some older storage tanks into compliance with Resource Conservation and Recovery Act (RCRA) standards (35). Calcination was not expected to resume at INEL until late 1990.

Calcination might be considered an intermediate step between liquid HLW and vitrified waste. The solid form of the calcine renders it less prone to tank leaks than liquid waste; furthermore, the volume of calcine, one-eighth that of the liquid, requires less storage space. The design lifetime of stainless steel storage bins for the calcine, 400 to 500 years, is perhaps an order of magnitude longer than that of the liquid HLW storage tanks, but perhaps two orders of magnitude shorter than required for a repository. All of the factors that caused INEL to pursue calcination, whereas Hanford, Savannah River, and West Valley chose vitrification, are not clear, but different
organizations may simply have developed their own approaches.69

Given the seemingly modest cost and the proven nature of calcining compared with vitrification, what—if any—are its liabilities? First, gases from the calcining step must be cleaned up to meet EPA standards before release. Second, the powdery nature of the calcine could result in airborne dissemination if it is not handled properly or if storage bins are breached. In response to an inquiry about storage of calcine, DOE stated, “We know of no technical problems at this time to preclude long-term storage of calcine in the bins, based on their design and on measured corrosion coupon results. Observed corrosion rates support the design lifetime of 500 years. However, the calcine is a radioactive hazardous mixed waste whose long-term storage may not meet regulatory requirements such as the Land Disposal Restrictions (LDR). 70 If DOE will be producing more HLW in the future, more attention should be given to the relative merits of the calcination process compared with tank storage followed by vitrification.

**Alternative Waste Forms for the Idaho National Engineering Laboratory**

Final choice of the longer-term solid form for HLW at INEL has not yet been made (49). Figure 1-9 shows some of the alternatives being considered in October 1989. Among these are the use of a glass-ceramic material for the matrix of the waste form in which the radioactive calcine would be embedded; that should permit higher radioactive waste loading (i.e., immobilization of a larger amount of radioactive material per canister) than borosilicate glass. The cost for disposal of one canister in a repository is cited by DOE as $350,000 (49). During a visit to INEL in July 1989, the following information was obtained: INEL anticipates a large increase in the fuel processing rate, resulting in a large HLW volume. If borosilicate glass vitrification is used to immobilize the calcine, production of 38,800 canisters is projected by the year 2020, corresponding to a disposal cost of $14 billion. However, if a glass-ceramic matrix being developed is used, an increase in the radioactive material loading will reduce the number of canisters by 2020 to 16,300, at a disposal cost of $6 billion. That cost might be lowered to $4 billion by changing the geometry of the canisters. INEL anticipates that the durability of the glass-ceramic will be similar to that of glass.71 72

At present, given the economic incentive, most research and development on the long-term form for INEL HLW seems to be devoted to glass-ceramic. However, one alternative is not immobilizing the calcine in a glass-ceramic matrix but hardening it in storage bins, that is, using the hardened calcine itself within an engineered barrier as the final waste form. Such an alternative might not prove attractive for repository disposal, given the large volume of calcine, but if delays in opening the repository become lengthy, necessitating a de facto shift to on-site, monitored retrievable storage, or if problems arise in vitrification efforts, bin hardening of the calcine might well be worth a harder look. A March 1990 DOE communication considers bin hardening a “subordinate alternative for long-term storage of calcine. . . . Based on current calcine leach data and the fact that the Snake River aquifer is located below the INEL, the bin hardening option is not feasible. Bin hardening will be addressed as the No Disposal Action option in an EIS (Environmental Impact Statement) for the immobilization project.”73 A decision on a reference strategy and form for Idaho Chemical Processing Plant HLW is currently planned for FY 1993-94.74

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69 Evidently, INEL fuels require large amounts of chemicals, including hydrofluoric acid (HF) to get them to dissolve. A substantial amount of aluminum is required to protect the tanks from HF. Thus, in hindsight, it could be argued that calcination might best have been employed at Hanford or Savannah River on acid liquid waste stored in stainless steel tanks, thus eliminating the difficulties in dealing with neutralized waste sludge and slurry; instead it was used on the more dilute waste at INEL. (Source: A.G. Croff, Oak Ridge National Laboratory, personal communication to R.P. Morgan, June 28, 1990.). Such an approach was seriously considered in the late 1950s at Hanford. Idaho (and also the French and British) had the proper foresight not to neutralize the HLW and store it in acidic form in stainless steel tanks; thus greatly simplifying the subsequent selection of a solidification process. By contrast, the neutralized HLW at the three other U.S. sites cannot be calcined without complicated pretreatment processes.

70 R.P. Cowan, op. cit., footnote 11.

71 This statement, of course, will need to be verified, probably by means of an extensive test program similar to that done with borosilicate glass over the past decade. Glass-ceramic and other ceramic materials had their supporters at the time the decision was made to use borosilicate glass for vitrification at Savannah River. A synthetic rocklike material called Synroc, also received attention about a decade ago.

72 J. Solecki, op. cit., footnote 12.

73 S. P. Cowan, op. cit., footnote 11.

74 J. Solecki, op. cit., footnote 12.
### Technologies for Pretreatment of High-Level Waste

At the three sites gearing up to vitrify HLW, Savannah River, Hanford, and West Valley, the HLW streams coming from the reprocessing plants are subjected to one or more steps before vitrification, which OTA has referred to as pretreatment. Two objectives of pretreatment are to reduce the volume of liquid that must be stored in tanks and to remove that portion of the streams that can be disposed of as other than vitrified HLW. Both these actions are driven in part by economic incentives; in their absence, the cost of waste management would increase because of the need for additional storage tanks and because of the high cost of vitrification relative to cemented waste forms disposed of near the surface.

Figure 1-10 illustrates treatment methods for HLW in tanks and canisters at Savannah River. Note that evaporation is an important element in the system, reducing the volume of liquid in the tanks. According to a 1988 document, without evaporation, 69 additional waste tanks valued at more than $33 million each would have been required (65). Prior to 1989, some water from the evaporator, not totally free of radioactivity, was discharged to seepage basins; in 1989, OTA staff observed operation of the new effluent treatment facility, which uses filtration, reverse osmosis, and ion exchange to clean up evaporator discharge (66).

Two main operations emerge after a series of pretreatment steps, as illustrated on the right-hand side of figure 1-10. One is the vitrification operation itself. The other involves mixing a decontaminated salt solution from the waste streams with cement to form a substance called “saltstone,” which will be disposed of in above-grade vaults. Making the saltstone is essentially a grouting or cementing operation of the kind used or planned for the disposal of some low-level waste (LLW). An approach that is similar overall but has some different steps to separate low-level streams is planned for Hanford HLW.

Various chemical operations plus radioactive decay with the passage of time are utilized to achieve low levels of radioactivity in the solution to be mixed with cement; some 99.9 percent of the aged waste supernatant will reportedly be removed (67). Among factors of interest to appropriate regulatory agencies are the amount and nature of the remaining

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**Figure 1-9—Alternative Long-Term High-Level Waste Management Strategies for the ICPP**

<table>
<thead>
<tr>
<th>Alternative Strategies</th>
<th>Process Options</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste</td>
<td>Immobilization Process</td>
</tr>
<tr>
<td>1. Dispose of waste in a geologic repository</td>
<td>Calcine and liquid waste</td>
</tr>
<tr>
<td>2. Dispose of waste in a near surface facility</td>
<td>Calcine and liquid waste</td>
</tr>
<tr>
<td>3. Dispose of annually generated waste in a geologic repository and dispose of existing calcine in a near surface facility</td>
<td>Annually generated liquid waste</td>
</tr>
</tbody>
</table>

radioactivity, and the mixed (i.e., hazardous plus radioactive) nature of the saltstone. The amount of saltstone to be produced at Savannah River is very large, about 3 million tons over 28 years; long-lived isotopes include 60,000 curies of technetium-99 (14). DOE reports that the saltstone facility started operating in the summer of 1990, all necessary permits having been granted by EPA and the State of South Carolina. Furthermore, saltstone has evidently been ruled a nonhazardous waste form; that is, in contrast to Hanford grout (see below), it is not considered a mixed waste. According to Savannah River officials, the saltstone contains much lower concentrations of organic material compared with the Hanford grout. This may account for the markedly different regulatory treatment.

The saltstone developmental process has not been without problems. In late 1987 it became evident that the organic chemicals used to decontaminate the salt solution created a flammability hazard; furthermore, the amount of benzene in the decontaminated filtrate sent to the saltstone facility did not meet new environmental standards. Modifications were made that should be in place (68). Further insight into the complex number of steps required prior to and in parallel with vitrification is given in the Savannah River Waste Management Operations Program Plan of 1988 (69).

The saltstone facility at Savannah River began operation in June 1990. As of that date, the only radioactive materials that had been immobilized and placed in the saltstone facility were streams from the

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76 S. P. Cowan, op. cit., footnote 11.
77 Ibid.
Effluent Treatment Facility and some wastes from tests of the in-tank precipitation (ITP) process that were carried out in 1983 and 1984. It will take about one year to fill one vault of the saltstone facility once operation of the pretreatment operations begins on the HLW tank wastes.

One key technical step that is new and has been the cause of some problems is precipitate hydrolysis, to be carried out within specially designated waste tanks, and referred to as ITP. The purpose of this process is primarily to remove radioactive cesium from the waste stream that will be sent to the saltstone facility. As of October 1990, Savannah River personnel were hopeful that radioactive operations of ITP would begin in April 1991; this would signal the start of pretreatment of HLW tank waste and immobilization of a non-high-level component in the saltstone facility.

The precipitate hydrolysis process at Savannah River involves the use of benzene, which emerges as a radioactive mixed waste. Plans call for building an incinerator to burn the benzene; until this occurs, it will be necessary to store the benzene in tanks.\footnote{Personal communication during trip to Savannah River Site, Oct 26, 1990.}

At other DOE HLW sites, there are some parallels to, and differences from, what is planned for Savannah River. At Hanford, the double-shell tank wastes are complex mixtures from a variety of operations, including a plutonium finishing plant (62). In 1990, Hanford began producing grout in the facility to be used in connection with the vitrification plant. The grout for the first run was “low-level” waste that was not “mixed” in nature. The grout facility is designed for mixing 1 million gallons of liquid with cement and producing 1.4 million gallons of grout,\footnote{Personal communication during visit to Hanford, November 1990.} the conversion actually increases waste volume. Like Savannah River, Hanford has been concerned about obtaining the necessary permits for grouting a portion of what started out as HLW. It has also shut down a key evaporator because of the presence of hazardous wastes in process condensates and the disposal of hazardous wastes directly in cribs, which violated either EPA or Washington State Department of Ecology regulations.\footnote{Viewgraph obtained during visit to Hanford, November 1989.}

Mixed-waste regulatory issues are being addressed through the process of obtaining a RCRA Part B permit from the State of Washington Department of Ecology.\footnote{S.P. Cowan, op. cit., footnote 11.}

At West Valley, processing of high-level alkaline liquid waste began in May 1988 to remove cesium-137 from the supernatant by ion exchange so that some of the waste can be treated as low-level waste (62). As of February 1990, more than 99.9 percent of the cesium-137 had been removed from more than half of the liquid in the larger of the two West Valley waste tanks; the liquid with the cesium removed had then been mixed with portland cement and stored on site in specially designed, easily stackable 7 l-gallon square drums in a storage building about 200 yards from a public road.\footnote{Personal communication during trip to West Valley site, Feb 21, 1990.} The cesium is captured by inorganic ion exchange on zeolite, which is stored for subsequent vitrification. Thus, West Valley leads other sites in the pretreatment of waste destined for vitrification by grouting a low-level fraction. The regulatory basis cited by DOE for this pretreatment is the West Valley Demonstration Act of 1980; the NRC is accorded special status in this act because of the commercial origin of the fuel that was processed into HLW. NRC does not have such a role in weapons sites such as Hanford and Savannah River. At West Valley, DOE sought and obtained NRC approval for the immobilized waste form from the pretreatment process.

The West Valley Demonstration Project was sued by citizens groups when it attempted to convert the building containing the drums with grouted waste into a permanent disposal facility. According to DOE, that building is the only “certified Class-C (low-level waste) cement farm in the country.”\footnote{Ibid.} From the DOE-WVDP point of view, radiation at drum surfaces was lower than anticipated because of better than anticipated cesium removal. Nevertheless, in an out-of-court settlement, DOE and WVDP agreed to study the matter and use the National Environmental Policy Act (NEPA) process to decide upon disposal by preparing an Environmental Impact Statement (EIS) not only for the drum building but also for the entire West Valley site. In
the meantime, on-site disposal is precluded; presumably, temporary storage is not. The EIS process should provide a forum for those concerned about above-ground storage of Class C low-level waste (38). Issues that might be scrutinized include the presence of technetium-99, a very long-lived, somewhat mobile, radionuclide in the grout at levels of 30 nanocuries per gram85; the integrity of the grout; and its ability to contain radionuclides over time.

At INEL, examination of a simplified flow sheet depicting HLW treatment indicates that to date, acid HLW flows directly to the calciner without pretreatment (60). An elaborate off-gas treatment system operates to control the radioactivity of effluent from the calciner. In planning further treatment of the calcine, consideration is being given to removing “inerts” (nonradioactive materials) from the calcine prior to immobilization (48). The efficiency, environmental impact, and regulatory requirements of this separation must all be considered; such a separation would appear to be somewhat analogous to HLW-LLW pretreatment separations being implemented at other sites.

**Waste Minimization**

Waste minimization is receiving increased attention at DOE. The charter of DOE’s Waste Reduction Steering Committee defines waste minimization as “any action that minimizes the volume or toxicity of waste by: 1) avoiding its generation, or 2) recycling” (60). Given this definition, if defense HLW alone is considered, minimization is intimately involved with production levels and methods for producing plutonium and tritium.

Several scenarios might be considered. If no more plutonium or tritium is produced, no more HLW will be generated.86 If one or more of the Savannah River reactors is restarted to produce plutonium or tritium, the radioactivity generated should probably be compared with the radioactivity that would be created if the same amount of material were produced in a new reactor designed to maximize the ratio of plutonium or tritium production to that of fission. If only tritium is produced in a reactor, DOE might consider whether a higher ratio can be achieved by producing only tritium.

Although an examination of reactor technologies could indicate possible HLW minimization, the savings are unlikely to be significant. Major reductions in HLW generation for this case appear to be possible only through reduction in plutonium and tritium production in nuclear reactors, production of tritium by a radically different method such as a linear accelerator, or substitution of uranium-235 for plutonium.87 This conclusion arises because HLW is an inevitable outcome of producing weapons fuel in a nuclear fission reactor.

However, for the second case in which spent fuel is not included within the definition of HLW, then one way of minimizing HLW generation is to reprocess only the lithium targets used to produce tritium and not to reprocess either the driver fuel elements or the depleted uranium targets used for plutonium production. According to this definition, processing to recover plutonium produces HLW whereas processing to recover tritium does not. An argument for proceeding in this manner is that the material more likely to be in short supply, namely tritium, could be produced in this manner without creating additional liquid HLW tank waste that must be dealt with. Furthermore, it might be argued that although both HLW and spent fuel will eventually be disposed of in deep geologic repositories, spent fuel is more cheaply, easily and safely treated and/or stored than HLW on an interim basis. On the other hand, the cost of running reactors fueled with enriched uranium for tritium production might increase if the fuel elements were not reprocessed. Furthermore, whether or not the spent fuel is reprocessed, the radioactivity to be dealt with will be the same.

At the next step in production-reprocessing—certain advantages may be derived from the use, reuse, and handling of hazardous materials because it could possibly make the waste management system simpler. Despite modernization of certain components and subsystems, most reprocessing plants and technology in the United States are 30 to 40 years old. New approaches that build on experience in other fields and possibly outside the United States, where commercial reprocessing activity has been pursued that utilizes smaller and more modern equipment, might well be possible.

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85 *Personal communication during trip to West Valley Site, Feb. 21, 1990.*
86 *This ignores the fact that HLW could still be produced from spent fuel that has not been reprocessed at certain DOE sites.*
87 *This assumes that uranium-235 is obtained by a process such as gaseous diffusion or centrifugation that does not involve its recovery from spent fuel in a nuclear reactor.*
Any efforts that minimize the generation of HLW have intrinsic merit because of the threat posed by the intense radioactivity associated with it. Moreover, because HLW and spent fuel are at the top of a pyramid that broadens to include other waste types (i.e., low-level, mixed, and transuranic), decreasing HLW generation will also reduce the problems created by waste in these other categories.88

REGULATORY FRAMEWORK

Introduction

Historically, the regulatory framework and standards for high-level waste (HLW) at Department of Energy (DOE) weapons sites have long been the province of DOE and its predecessor agencies under the Atomic Energy Act.89 The primary vehicle for specifying the definition, handling, and treatment of defense HLW has been by DOE order, in which the Secretary of Energy has the final authority. However, the Nuclear Regulatory Commission (NRC) becomes a factor in establishing licensing criteria for disposal of defense HLW. According to current plans, vitrified HLW is to be placed in the same deep repository as spent fuel from commercial reactors; criteria for the repository and for the waste forms to be placed therein are governed by the NRC.89 Environmental standards for repository disposal of HLW are the responsibility of the Environmental Protection Agency (EPA). Furthermore, EPA’s role in radiation protection standard setting has been growing and affects DOE’s HLW management activities. EPA is the lead agency in a Federal interagency committee to prepare Federal guidance on radiation protection of the public. EPA is also primarily responsible for setting environmental radiation standards for specific practices or sources, although criteria developed by NRC for the commercial sector or DOE for the defense sector can apply if they are more stringent than EPA standards (23).

EPA regulates waste management practices at DOE weapons sites through its jurisdiction over hazardous wastes and the application of the Resource Conservation and Recovery Act (RCRA) and the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA). Its role in regulating hazardous waste and the hazardous components of mixed waste is broadly based. It is mainly with regard to hazardous materials that regulatory pressure and actions are being brought to bear on DOE. Two examples at Hanford are: 1) shutdown of the 242-A evaporator in April 1989 because process condensates contained hazardous ‘‘listed wastes’’ (e.g., acetone) and 2) prolongation of the shutdown of the Plutonium Production and Extraction (PUREX) reprocessing plant, beginning in December 1988, in part because acetone and other listed wastes were being used and discarded. The latter two actions are in response to RCRA, which governs ongoing waste management operations.

Strictly speaking, all HLW is mixed waste; that is, it has both hazardous and radioactive components. It might be argued that because the health threat represented by the radioactivity of HLW far outweighs the health threat associated with the hazardous component of that waste, any actions that DOE takes to provide adequate protection against radioactivity would provide more than adequate protection against the hazardous component. However, this argument does not appear to be accepted in the regulatory sense, nor need it prove correct in all situations having to do with storage, treatment, transport, and disposal of HLW.

State agencies have also become factors in regulation through interagency agreements. For example, at Hanford, the Federal Facility Agreement and Consent Order, the so-called tri-party agreement, entered into in May 1989 by the Washington State Department of Ecology, EPA, and DOE, governs the Hanford cleanup (71). In the Hanford tri-party agreement, milestones are set forth for vitrification of HLW from double-shell tanks, and a more expanded schedule involves further study before any action is taken for single-shell tanks. A 1989 report sets forth the very complex set of regulations that apply to HLW management at Hanford.

88This discussion of HLW minimization focuses on radioactivity, the overwhelming contributor to its toxicity. Often, analyses of waste minimization concentrate on reducing the volume of the waste; accordingly, the volume reductions achieved by vitrifying or calcining a given amount of HLW might have been compared. However, reducing the volume after the waste has been generated does not fit the concept of waste minimization as defined by the DOE Waste Minimization Steering Committee; nor, in the case of HLW, does it really get to the heart of the problem. Volume reduction considerations are relevant to pollution prevention and cost savings, and appear throughout DOE planning and this report.

89Reg. 1970, the first regulatory definition of high-level waste was developed by the U.S. Atomic Energy Commission in 10 CFR Part 50, app. F.

90This authority is vested in the NRC by the Nuclear Waste Policy Act (NWPA) [Pub. L. 91-190, 93 Stat. 2201 (1983) (codified) at 42 U.S.C. §§10101-10226] and also applies to the case where defense HLW is not commingled with spent fuel but placed in a separate repository.

91“Listed wastes” are Substances that have been placed on the RCRA-based list of hazardous materials and are thus subject to EPA regulation. In this report, the term “hazardous” is used in this sense; in other words, radioactivity, although a hazard, is not “hazardous.”
regulations that might govern the treatment of single-shell tanks and tank wastes (22).

**Definition of High-Level Waste**

The definition of *high-level waste* contained in both the DOE 1989 Five-Year Plan (55) and the draft DOE Order No. 5820.2A (63) is as follows:

...the highly radioactive waste material that results from the reprocessing of spent nuclear fuel, including liquid waste produced directly in reprocessing and any solid waste derived from the liquid, that contains a combination of transuranic waste and fission products in concentrations requiring permanent isolation.

The last five words of this definition introduce the idea of a definition based on the concentration of radionuclides in the waste and not solely on the source of the waste, namely, liquid waste produced directly in reprocessing. In recent years, the issue of a source-based v. a concentration-based definition has arisen because strict application of the former would require the treatment and disposal of much larger amounts of liquid HLW currently stored in tanks, thus greatly increasing overall costs. If some waste of lower radioactivity could be separated and treated as low-level waste, costs would decrease not only because of reduced disposal costs but also because of more effective use of existing HLW tank storage space. On the other hand, if such a redefinition is not allowed and a strict interpretation of the existing source-based definition promulgated by the NRC and listed in the *Code of Federal Regulations* is adhered to, DOE’s cleanup plans could be affected very substantially. Presumably, the definition is based not only upon cost considerations but also upon consideration of health effects and potential health risks.

The issue is particularly relevant to the waste at Hanford and Savannah River. Both of these facilities expect to reduce the amount of HLW by separating a large “low-level” waste component from the reprocessing streams prior to vitrification. At Savannah River, this involves separation of a salt solution that has been decontaminated of at least 99.9 percent of its radioactivity; the salt solution will then be mixed with cement to form saltstone and disposed of on-site in above-grade vaults (see figure 1-10). At Hanford, HLW is to be pretreated by a series of steps that will separate “low-level” liquid streams to be grouted and then disposed of on-site in near-surface vaults. The specific technical steps differ, but the net result is the same. Figure 1-11 illustrates the reason for interest in this approach: the cost of HLW treatment and disposal by vitrification is about two orders of magnitude greater than the cost of LLW treatment and disposal.

In November 1989, the NRC gave tentative approval to a DOE plan to grout and then pump into concrete vaults some of the high-level tank waste at Hanford. According to a State of Washington estimate, “As much as 10 percent of Hanford’s HLW could be put in low-level vaults because technical difficulties prevent the separation of all the high-level material from less radioactive components . . . ’ (3). Officials of the State of Washington called for an independent assessment of that decision. However, a DOE spokesman said that DOE has a reasonable, cost-effective plan for dealing with the waste (3).

These conflicting views can be interpreted in the context of how HLW is defined and who has regulatory authority over it. NRC is involved because of its responsibility for the HLW repository. Evidently DOE argued, and NRC concurred, that the material to be grouted, regardless of its source, had concentrations that resemble low-level waste and should be governed as “incidental waste” under a rule from the 1970s. 93

The HLW definitional issue, as perceived by environmental groups, arose in what they viewed as a DOE effort to redefine Hanford single-shell tank waste so that it could be left in the tanks as a cost-saving but potentially dangerous means of disposal. 94

The source-based definition of HLW still appears to be the official one, and the NRC interpretation does not appear to permit redefinition of the Hanford single-shell tank waste. However, it does appear to

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92 *Code of Federal Regulations. 10 CFR 0.735-I, Title 10, Energy, Chapter I, Nuclear Regulatory Commission, Part 60, Subpart A, P. 542.*

93 *Personal communication during visit to Hanford, November 1989.*

94 “What DOE may have had in mind is treating single-shell tank waste in situ, that is, at the site by vitrification or some other means. In situ treatment is believed by some to have two major advantages over removal and treatment: namely, it should be considerably less costly and should pose less of a health risk to workers. Whereas this may be an appealing alternative technically, the political difficulties associated with it are significant.”
Wow redefinition of a portion of Hanford HLW for disposal as low-level waste by grouting.\(^\text{95}\)

The grouting of a “low-level” fraction of high-level tank waste is currently underway. As of mid-1990, more than half of the low-level fraction at West Valley had been grouted. The Savannah River saltstone operation has begun operating with waste from the effluent treatment facility. The low-level fraction of Savannah River tank waste is expected to be treated at the saltstone facility starting in mid-1991. Hanford is also proceeding to get necessary approval, in accordance with the tri-party agreement, to grout portions of tank waste. Thus the definitional issue may be of only academic interest. It could resurface, however, if questions arise about disposal of the grouted “Class C low-level waste” resulting from the current approach, which appears to be happening at West Valley.

At West Valley DOE moved vigorously forward with a program of pretreating the HLW to reduce the HLW disposal burden. There was at least one attempt in the mid-1980s to redefine the various waste categories (2). That effort did not succeed however and pretreatment is now being carried out as planned.

In December 1990, the States of Washington and Oregon petitioned the NRC to initiate rulemaking to redefine HLW as follows: \(^\text{96}\)

HLW means: (1) Irradiated reactor fuel, (2) Liquid wastes resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated wastes from subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuel, and (3) Solids into which such liquid wastes have been converted; provided that if, prior to disposal, defense reprocessing tank wastes are treated to remove the largest technically achievable amount of radioactivity on a tank-by-tank basis ..., the treated residual fraction shall be considered an incidental waste and therefore not HLW.

The States apparently took this action because they feared that wastes in the Hanford tanks could be designated as low-level waste and disposed of in a facility conforming to EPA requirements but unlicensed by the NRC. \(^\text{97}\)

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\(^{95}\) The question might be raised as what authority NRC has over this matter. There appears to be no explicit authority as in the case of West Valley.


\(^{97}\) Weapons Complex Monitor, Dec. 31, 1990, p. 3.
Regulations Affecting Single-Shell Tanks

Regulatory requirements important to waste management decisions for the Hanford single-shell tanks are summarized in a Pacific Northwest Laboratory report (22). The number of regulations and regulatory bodies that will govern tank cleanup is large, and the process is complex (see figure 1-12 and table 1-2). Uncertainties and conflicts could very well arise, some of which may be resolved as tank contents are better characterized. Many issues have not yet been resolved, involving waste definitions, mixed-waste disposal, and groundwater protection requirements. RCRA may not provide sufficient quantitative criteria to assess the performance of proposed disposal systems, and variances from some applicable RCRA regulations for tank storage systems may be needed to remain in compliance. Some emphasis is needed to determine quantitative criteria, other than those in RCRA, that can be used for guidance in areas such as groundwater protection (22). Attention should be paid to regulatory requirements as the waste characterization process continues.

Taking core samples from tanks to characterize the waste and analyzing these samples can expose workers to radiation that exceeds ALARA (as low as reasonably achievable) limits. Modifications have been proposed to the EPA guidelines for testing methods to be used in the evaluation of solid hazardous waste (21). These modifications specifically focused on sampling and analysis procedures for the highly radioactive single-shell tank waste. In early 1990, EPA did propose to amend testing and monitoring regulations for hazardous wastes under Subtitle C of RCRA.

Regulations Affecting Restart of PUREX

The regulations external to DOE that affect restart of the PUREX plant are concerned mainly with hazardous wastes. First, a major waste management evaporator at Hanford, the A-242 evaporator, was placed on temporary standby because hazardous waste was being disposed of in cribs and because process condensates contained hazardous waste, as defined by the State of Washington Department of Ecology regulations. DOE and Westinghouse-Hanford believe that restart of the evaporator is essential in reducing the volume of liquid waste to be accommodated in double-shell tanks so that new waste from the PUREX restart can be pumped to those tanks.

The PUREX plant itself is shut down for a variety of reasons. The initiating event in December 1988 was a “limiting condition of operation” violation in which the steam pressure in a line fell below the 185 pounds per square inch required for operation. In addition, the PUREX process uses a variety of organic materials and solvents that are hazardous wastes regulated under RCRA.

Concerns were expressed about how the plant and its aging components might behave during restart, after having been shut down for an extended period in mid-campaign. In December 1989, DOE undertook to operate the plant for several weeks to clean out materials that had lodged in the system during shutdown and to stabilize the plant for an extended shutdown of about a year, prior to restart for normal operations (75). In October 1990 DOE announced that it would not restart PUREX for at least 2 years but would prepare an Environmental Impact Statement to evaluate a variety of treatment and disposal methods for stored N reactor fuel.

Finally, it should be noted that the regulation of hazardous wastes at DOE sites under RCRA gives EPA and authorized States a mechanism whereby they can exert some regulatory control over DOE’s waste management activities. In certain instances, the conditions involving hazardous waste that need correction may not appear very threatening, compared with the dangers “posed by the radioactive components of the system. Nevertheless, the regulatory agencies have used the only authority available to exercise control over these DOE activities.

Regulations Affecting Vitrification

It seems reasonable to assume that under the current framework, DOE will have major regulatory authority for the vitrification process itself. However, there is an important interface with the NRC which, in turn, interfaces with EPA. NRC is...
Figure 1-12—Regulations for Management and Disposal of Nuclear and Hazardous Waste
(Hanford Single-Shell Tanks)

Abbreviations:
DSHS: (Washington State) Department of Social and Health Services.

The performance of the waste form in the repository will be governed by standards promulgated by EPA for the management and disposal of spent fuel and of transuranic and HLW. Such standards were established in 1985 (40 CFR Part 191); however, they were vacated by the First Circuit Court in 1987 and remanded to EPA for further proceedings (16). At present, no new formal proposal has been published by EPA. Disposal standards for HLW were expected to have appeared for public comment late in 1990.

At a briefing at Savannah River in October 1990, the following information about oversight and monitoring of the vitrified waste form was obtained. The DOE’s Office of Civilian Radioactive Waste Management (OCRWM) has setup specifications for acceptance of the vitrified glass product. A Waste Form Compliance Plan has been submitted to the OCRWM and a Waste Form Qualification Report is being prepared; these reports are reviewed by an internal DOE Technical Review Group. Later, a Production Records report will be written to provide data as to whether the glass form complies with specifications. The OCRWM is the organization that interfaces with the NRC in connection with the repository; OCRWM will transmit these reports to the NRC.

The glass specifications are being based on the NRC technical criteria for the repository under 10 CFR 60 rather than the EPA disposal standards for the repository promulgated at 40 CFR 191. New methods are being developed to obtain and characterize product samples; these methods are being submitted for review by the American Society for Testing Materials, a national certification organization. EPA’s role in regulating the waste form is, at the moment, unclear. The HLW to be vitrified has RCRA hazardous materials and is therefore a mixed waste. In mid-1990, EPA designated vitrification as Best Demonstrated Available Technology (BDAT) for mixed HLW. Although DOE and Westinghouse-Savannah River officials have met with EPA officials to discuss the matter, it still appears to be an open question as to whether or not EPA will require...
proof of vitrification. DOE seems to be proceeding on the basis that EPA may be a factor in monitoring the waste form and is developing a test that might be substituted for the EPA Toxics Characterization and Testing Protocol (TCTP); the latter can not be performed in a hot cell.106

Whether or not EPA monitors the vitrified waste form after production and during storage appears to be an open question. The answer may depend more upon the inclination and actions of the principal organizational players, namely DOE, EPA, and perhaps NRC, than on clear-cut existing regulatory requirements.

Associated with the vitrification process and usually one step ahead of it is the production of an immobilized grout or “saltstone.” The saltstone facility at Savannah River has been granted a permit by the State of South Carolina as a nonhazardous waste facility. Thus, it is apparently not subject to EPA regulation. This is in contrast to the Hanford grout facility where continuing EPA presence seems assured by the larger component of hazardous materials in the waste. DOE monitors the saltstone product and produces monthly reports. Internal oversight is provided by DOE’s Office of Environment, Safety and Health.107

DISCUSSION

Definition of High-Level Waste

In recent years, attempts to redefine high-level waste (HLW), moving from a source-based to a concentration-based definition, have occurred and have caused some controversy. The definition used can have a substantial impact on cleanup and waste management operations and costs. The Department of Energy (DOE) has proceeded to follow a concentration-based definition in pretreating HLW prior to vitrification so that a portion of tank waste can be disposed of as low-level waste. The current DOE definition of HLW uses the qualitative phrase “in concentrations requiring permanent isolation.” Because this definition could lead to several different interpretations, it may need reexamination. Also, any concentration-based definition may need to be reexamined in view of the fact that the Nuclear Regulatory Commission (NRC) source-based definition remains part of U.S. Code. Finally, it may be desirable to arrive at a single, consistent definition of HLW and other waste categories that is adopted by DOE, NRC, and the Environmental Protection Agency (EPA) and is acceptable to State agencies and public interest groups. However, irrespective of definition, the waste must meet EPA disposal standards that have to be reissued; the definition and standards are interrelated.

Repository Delays and Contingency Planning

DOE’s Five-Year Plan is predicated in part upon the availability, starting about 2010, of a deep geologic repository for disposal of HLW, as mandated by Federal legislation. In accordance with this thinking, DOE is moving forward, at three of four sites that have HLW, with vitrification facilities for converting HLW from a liquid to a glasslike solid in a form acceptable for repository disposal. This strategy has the major advantage of reducing the potential threat to public health and the environment, in the short term, that is posed by more mobile tank waste, albeit at some increased occupational risk. However, more consideration must be given to facilities and requirements for storing solid waste if the repository opening is delayed.

For several decades, or even longer, de facto, on-site, monitored, retrievable storage of vitrified waste should not be technically prohibitive, provided vitrification goes well.108 However, the institutional controls and monitoring needed for such storage require further attention. There appears to be no substantial contingency planning underway to allow for the possibility that vitrification might not succeed or might encounter major delays; the calcination work at the Idaho National Engineering Laboratory (INEL), which produces a powdery solid that is stored in bins with a lifetime of 400 to 500 years is an alternative that could be examined. The political trade-offs associated with planning for various contingencies must also be considered. For example, shipment of canisters with vitrified HLW to a geologic repository may be opposed by some along the transport route; on the other hand, long-term on-site storage of those waste canisters may not

106 Personal communication during visit to Savannah River Site, Oct. 26, 1990.
107 Ibid.
108 European countries plan to allow their vitrified high-level waste to cool on-site for 50 years before further action is taken. The benefit of letting the waste cool (i.e., undergo some radioactive decay prior to repository disposal) merits consideration in the United States, which has not planned for such a long cooling period but may, in fact, be accommodating to one.
be acceptable to residents and officials of the State in which the site is located. In addition, changing from the current policy of building a deep repository in favor of monitored retrievable storage at the weapons sites would require extensive study and debate. These issues could be scrutinized further in the Programmatic Environmental Impact Statement (EIS) that DOE now has underway.

**Urgency of High-Level Tank Waste Treatment**

The urgency of solidifying high-level tank waste is difficult to quantify. For example, although DOE generally asserts that HLW currently stored in tanks at Hanford, Savannah River, Idaho, and West Valley poses no imminent threat to public health, current or potential groundwater contamination due to tank leakage is a matter of debate. Concerns about the possibility of tank explosions with ensuing large releases of radioactivity also continue to arise. Accurate characterization will require time and money—perhaps a decade and hundreds of millions of dollars—if current regulatory guidelines are adhered to. The characterization process might be speeded up by more focused sampling and attention to suspected environmental pathways.

Current DOE plans indicate that vitrification of liquid waste from double-shell tanks at Savannah River will begin in FY 1992 or 1993, followed by vitrification of Hanford double-shell tank waste commencing in FY 1999. Decisions about the treatment or disposal of single-shell tank waste at Hanford have been deferred. The tri-party agreement calls for closure of single-shell Hanford tanks during the period 2005 to 2018; the agreement also calls for the removal of all pumpable liquid waste from these tanks by 1996 (42). A General Accounting Office (GAO) report advises that this latter date should not deter DOE from removing the liquid sooner, if possible, given GAO’s conclusion that DOE’s current monitoring efforts do not provide sufficient data to adequately trace the migration of leaks or to fully assess their effects (42). GAO also advises placement of new ground cover material over the tank farms to slow water drainage through the soil. The major concern is the danger of contamination of groundwater and the Columbia River by leaking high-level tank waste.

At Hanford, priority was given to early treatment of liquid HLW in double-shell tanks, in response to a number of factors, possibly including the relative ease of treatment of double-shell waste compared with single-shell waste; the less mobile condition of single-shell waste; and the fact that the double-shell tanks are required for new waste storage. This priority is now codified in the tri-party agreement. While it is important to move ahead with a treatment system for the double-shell tank waste, it is also important to give attention to the single-shell tank problem and to reach a decision on how to improve conditions of waste storage there as soon as possible.

At a December 1989 meeting of the National Academy of Sciences Panel on Hanford Single-Shell Tanks, panel members urged DOE and Westinghouse-Hanford to take a more systematic overall approach to exploring alternatives for treatment and disposal of single-shell tank waste rather than simply focusing on specific tasks, such as taking core samples, without any concept of the “big picture” for treatment and disposal. The panel also called for more evaluation of the potential for tank explosions due to the ferrocyanide that had been added to some single-shell tanks, as well as a determination of other tank contents that might constitute an explosive hazard. If this evaluation should reveal the possibility of tank explosion, immediate corrective action would have to be considered. Indications, based in part on DOE statements to date, are that the possibility of explosions involving ferrocyanide will be of greater concern during treatment operations than during storage.

More recently, reports of the presence of hydrogen gas in some of the HLW tanks at Hanford have raised the possibility of fires or explosions. The matter is under more intensive study by DOE following a number of oversight investigations and hearings in 1990. This situation increases the urgency of proceeding with the solidification of high-level tank waste. However, the trade-offs between moving ahead with dispatch and moving ahead too precipitously require careful consideration. In early 1991, Secretary of Energy Watkins indicated that a two-year delay in the start of construction of the Hanford Waste Vitrification Plant was needed; two reasons were to complete a risk assessment and to develop more data on the contents of the tanks to ensure safe pretreatment. Such a delay would require modification of the Tri-Part Agreement.

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**Technologies for High-Level Waste Treatment**

Two technologies are central to DOE’s plans for treatment of liquid high-level tank waste over the next decade: 1) vitrification of the “high-level” component of the waste with borosilicate glass, followed by placement in canisters and disposal in a deep repository, and 2) immobilization in grout or concrete of the “low-level” component of the waste, followed by on-site disposal either at or near the surface. Both of these technologies require major financial investments, especially vitrification. It is important that technical work be performed well and monitored carefully.

Vitrification of the HLW component is scheduled to commence in the Defense Waste Processing Facility (DWPF) at Savannah River in 1992 or 1993. The technology will have to be carefully demonstrated over a period of time, first with cold runs (i.e., with no radioactive waste). These cold run tests began in the fall of 1990. Careful balance is required between the need to move as rapidly as possible in getting high-level tank waste into the more stable vitrified form, and the need to proceed carefully and cautiously. If all goes reasonably well at Savannah River, the Hanford Waste Vitrification Plant (HWVP) should be less problematic because it will use a technology similar to that of the DWPF.

The operation of grout facilities at both Savannah River and Hanford could reduce the volume of high level tank wastes to be managed in the future. Grout or concrete has the advantage of being a solid form, in contrast to current tank waste liquids and sludges. However, given the concerns that have been raised about treating some fraction of the high-level tank waste in a “low-level” reamer, some questions concerning this practice must be resolved. A key question is how long will the grout last (i.e., keep radioactive or hazardous components contained)? DOE may need to investigate the lifetime of these waste forms more extensively.

**Rethinking the Waste Form and Package**

DOE has decided to use a waste form and package for the disposal of vitrified HLW that involves using borosilicate glass and relatively thin-walled (about 1-centimeter-thick) stainless steel canisters. Whereas this approach was chosen to assure a lifetime range of several hundred to 1,000 years, the canister itself is not expected to last the 10,000 or more years required to isolate the long-lived transuranic and other radionuclides from the environment. Therefore the current approach provides for long-term integrity to be ensured in large part by the integrity of the deep geologic repository.

In contrast, Sweden places more reliance on the waste package to ensure that radionuclides will not escape into the environment. Plans call for the Swedish waste package to be thick walled (about 10 centimeters) and for the copper canisters to be filled with either molten lead or copper surrounding the spent fuel. A National Academy of Sciences panel that reviewed the Swedish plans believes that, in this manner, canister lifetimes of 1 million years or more can be achieved (28). This means that less reliance has to be placed on the geologic repository.

The need to achieve a 10,000-year or 100,000-year lifetime for waste isolation in a repository has created a difficult regulatory problem. EPA’s HLW disposal standards were struck down by a court in part because some controls were not deemed to assure control for a sufficient length of time (16). Also, there is the possibility that stricter radiation protection standards may be needed in response to the findings of increased risks of cancer from radiation (29). Finally, if a deep geologic repository for HLW disposal is delayed for a long time, and monitored retrievable storage were required during that time, a waste package with a long design life could be useful.

Given the delays in repository development in the United States, it might be useful to review storage and disposal options for defense HLW with particular attention given to strengthening the engineered barrier performance of the waste package. Although some elements of DOE’s HLW management strategy (such as the DWPF vitrification plant at Savannah River) may be too far along to change, others (such as the HWVP) may not be. Also, since DOE’s canister must fit in the same NRC-approved container as commercial spent fuel before being placed in the repository, the canister design might not have to be changed; instead, the overpack...
A key question to be considered is whether the major additional cost of proceeding with engineered barriers is justified by the benefits to be derived. Another question is whether current performance assessment methodology for the combination of engineered barriers and repository allow assessments to be made in which one can have confidence. This may depend on whether knowledge of interaction between the engineered barrier and the repository environment is sufficiently advanced so that a barrier can be designed that will perform its function for the required period of time.

The choice of borosilicate glass as the waste form for vitrification of HLW is consistent with the waste form selected in other countries. OTA found that a high level of confidence in this choice was held by DOE and its contractors; however, this view is not shared by all. Concerns have been raised about the long-term performance of borosilicate glass in the Yucca Mountain repository environment, the adequacy of the scientific program to demonstrate that borosilicate glass will retain defense HLW for the necessary duration, and the lack of adequate R&D on second generation waste forms after DWFP.

Producing a qualified waste form from the wide range of input waste feed to the vitrification process is a challenging technical assignment. The development of a theoretical framework with which to predict long-term waste form performance appears to be an ongoing process and DOE, in cooperation with the regulatory agencies, will be participating in this process for many years to come.

Waste Form for the Idaho National Engineering Laboratory

Cost reduction appears to be a major factor in the search for a waste form other than borosilicate glass at INEL; each canister of vitrified borosilicate HLW is very expensive to produce and dispose of in a repository (50). If more waste can be loaded onto the matrix by the use of glass-ceramic instead of borosilicate glass, costs for immobilizing the HLW at Idaho might be reduced by billions of dollars.

Idaho also has 25 years of experience in calcining HLW. Currently, the HLW calcine at INEL is stored in stainless steel bins within reinforced concrete vaults having design lifetimes of 400 to 500 years. The possibility of hardening the calcine within the existing storage bins could be an alternative under consideration.

The INEL waste form decision represents an opportunity to reexamine this area in light of what has been learned since the decision in favor of borosilicate glass a decade ago. Such reexamination could include economic, environmental and political factors. An independent technical review panel might be useful in this regard if it had the resources to do the level of evaluation needed.

Releases to the Atmosphere

Nuclear reactors, reprocessing plants, and facilities such as evaporators and calciners for treating HLW sometimes release radioactive or hazardous materials to the atmosphere. Through the introduction of air filters and other off-gas handling equipment, releases of radionuclides have been greatly reduced, but not totally eliminated, since the early days of the weapons program. In addition to releases from routine operations, concern persists about potential releases during accidents such as HLW tank explosions of the kind that occurred in 1957 in the Soviet Union. Air emissions, including radioactivity, from DOE sites are subject to National Emissions Standards for Hazardous Air Pollutants (NESHAPS) promulgated by EPA under the Clean Air Act.

Unlike groundwater, the air exposure pathway of the offending materials is direct and known. At the Office of Technology Assessment (OTA) Health Effects Panel Workshop in January 1990, several panelists stated that they believed airborne release of both radioactive and hazardous materials to be a greater potential health threat than groundwater contamination. Further, they pointed out that the Resource Conservation and Recovery Act (RCRA) and the Comprehensive Environmental Response, Compensation and Liability Act (CERCLA) regulations focus attention on groundwater contamination but ignore air releases. Setting standards for the release of radionuclides to air has been the object of


\[112\] Ewing, R., personal communication to R.P. Morgan, March 5, 1991.

contention between NRC and EPA (33), and the monitoring of emissions presents some technical difficulties. Although EPA has authority to set air release standards for radioactivity from DOE sites, implementation and enforcement remain with DOE. It may be useful to pay more attention to regulating air emissions, to implementing and enforcing air release standards, and to monitoring DOE activity in this area.

Future of the PUREX Plant at Hanford

In December 1988, the PUREX fuel reprocessing plant at Hanford was shut down in mid-campaign due to a low steam pressure condition. The plant has caused concern because of its age, the large amounts of hazardous and radioactive wastes it produces, past atmospheric releases, and continued release of liquid effluents to the soil even in its shutdown condition. In December 1989 the plant was restarted for a short time to stabilize the situation by flushing out material that had settled in pipes and other equipment during the sudden shutdown. After this stabilization run, DOE had planned to restart PUREX in late 1990 to reprocess the backlog of spent defense fuel over a 5-year period and then permanently to close the facility. However, a decision was made in 1990 not to restart PUREX for at least 2 years but to prepare an EIS and evaluate options for handling the stored fuel.

The decision not to restart PUREX may have been reached for a number of reasons, including: 1) the U.S. plutonium stockpile is widely reported to be sufficient; 2) citizen groups and state officials had increasingly raised questions and expressed concern about environmental impacts of operating PUREX; 3) regulatory constraints imposed by RCRA had already shut down the plant and there was also the threat of pending legal action if restart was attempted; and 4) outside independent analysis coupled with DOE’s own work suggested that encapsulation and storage of spent N-reactor fuel could be an alternative to reprocessing with environmental benefits.

The future of PUREX will continue to be an issue of intense public concern as well as requiring sound technical analysis. It will be a challenge for DOE to resolve while actively involving the public in the EIS process.

Waste Minimization; Tritium Production; International Cooperation

The radioactivity of HLW generated from reprocessing spent fuel and irradiated uranium targets is strongly related to weapons material production requirements. Within current production practice, it will be difficult to reduce HLW other than by reducing production. However, it may be possible to produce tritium without producing HLW if no reprocessing of the spent driver fuel elements is performed. There is no minimization of radioactivity if such a change in operations were to be adopted; to a first approximation, the total radioactivity associated with the fission process should be the same, whether the radionuclides are contained in the spent fuel elements or released from those elements and contained in waste tanks, or eventually, in glass logs.

Hazardous waste and certain types of radioactive and mixed waste other than HLW might well be reduced by technological improvements to reprocessing. The U.S. decided in the 1970s not to pursue reprocessing of commercial nuclear fuels. Other nations may have acquired certain expertise in reprocessing that might prove useful to DOE’s efforts at waste minimization. DOE has already supported several important programs in international technology exchange. Since the DOE waste minimization program is currently in a very early stage, its design could profit by a wide range of input. Learning from international experience and expertise should be a strong element of the DOE effort.

Scenarios for Future HLW Production

HLW is an inevitable consequence of the fission process that occurs in current nuclear weapons production practices. At present, no DOE weapons production reactors are operating. This pause in reactor operations provides an opportunity to pursue cleanup at the Weapons Complex during a time of reduced levels of HLW generation. The Department of Energy has recently begun to prepare an Environmental Impact Statement of its plans for modernizing the Weapons Complex. Such an analysis should provide valuable insights and help DOE in its efforts to focus greater attention on the environmental consequences of various production scenarios.

In January 1991, DOE issued a Reconfiguration Study for the Weapons Complex which appears to represent a useful step towards facilitating the PEIS process. In that study, alternative configurations