APPROACH TO IMPROVED SAFETY OF HIGH-LEVEL RADIOACTIVE WASTE STORAGE BY THE U.S. DEPARTMENT OF ENERGY

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ABSTRACT

In mid-1990, two unreviewed safety questions were identified associated with high-level radioactive waste (HLW) storage at the U.S. Department of Energy (DOE) Hanford Site located near Richland, Washington. Admiral Watkins, the Secretary of Energy, and Leo Duffy, the Assistant Secretary for Environmental Restoration and Waste Management (EM), immediately initiated an aggressive program to resolve these two unreviewed safety questions and to improve the overall safety posture of HLW storage. This paper will first describe the DOE's approach to improve the safety of HLW storage and then discuss DOE's program to resolve the high-priority safety issues at the Hanford Site.

High-level radioactive wastes are stored at four DOE facilities: the Hanford Site, the Idaho National Engineering Laboratory, the Savannah River Site, and the West Valley Demonstration Project. The largest volume of waste is stored at the Hanford Site, while Savannah River Site has the greatest quantity of radionuclides. It is DOE's plan to treat and immobilize the high-level fraction of HLW at Hanford, the Savannah River Site, and the West Valley Demonstration Project as borosilicate glass for disposal in a geologic repository. To that end, vitrification facilities are undergoing operational testing at the Savannah River Site, being completed at the West Valley Demonstration Project, and are in the final planning stage at the Hanford Site. Unlike other sites, Idaho National Engineering Laboratory is storing its HLW either in acidic form or as calcine, and DOE is still in the early planning stage regarding selection of the appropriate waste form for disposal of this waste.

Of all the sites with HLW storage, Hanford poses the greatest challenge, given the age of the storage tanks, the large number of different production and waste processing campaigns, and design of the single-shell tanks; the majority of the HLW tanks at Hanford are single-shell tanks. A significant number of the single-shell tanks (66 of the 149) at the Hanford Site have either leaked or are suspected to be leaking. Compatibility of waste chemistry is also a major concern for both the single-shell tanks and double-shell tanks at the Hanford Site.

To address these safety concerns, DOE in 1990 established a HLW Tanks Safety Task Force. The charter of the Task Force is to identify safety issues, develop near-term mitigating measures, and recommend remediation measures for longer-term safe storage in tanks at all HLW sites. In addition, the charter includes the review of Safety Analysis Reports to determine adequacy of the safety envelope, a review of the conduct of operations to ensure safety of workers and safe operation of the HLW storage tanks, the development of a common database, and expansion of the scientific and technology base to address HLW storage and treatment(1). A HLW Tanks Advisory Panel was formed to provide technical guidance to the Task Force and to advise the Assistant Secretary for Environmental Restoration and Waste Management. Interface with oversight organizations both internal (the Office of Environment, Safety and Health; the Office of Nuclear Safety; and the Advisory Committee on Nuclear Facility Safety) and external (the Defense Nuclear Facilities Safety Board and the National Academy of Sciences) was also mandated.

DOE'S APPROACH TO IMPROVING THE SAFETY OF HLW STORAGE

A schematic of the DOE approach to achieving safe storage of HLW is presented as Fig. 1. Many of these steps are carried out in parallel, with many iterative loops.

Identification, Prioritization and Resolution of Safety Issues

The Office of Environmental Restoration and Waste Management coordinates the activities of the DOE Field Offices, facility operators, Task Force, and Tanks Advisory Panel to identify, evaluate, prioritize, and resolve safety issues at each of the facilities.

Preliminary criteria have been developed for establishing priorities for evaluation of HLW safety issues. They are:

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Criteria 1. Issues/situations which contain most of the necessary conditions that could lead to worker (on-site) or off-site radiation exposure through an uncontrolled release of fission products (e.g., flammable gas accumulation and periodic release that are observed in certain Hanford HLW tanks).

Criteria 2. Issues/situations which present (or contain) some conditions that could lead to an uncontrolled release of fission products under extreme assumptions (e.g., seismic issues).

Criteria 3. Issues/situations which could lead to the future release of fission products if the tanks are viewed as intermediate storage (5-30 years) of HLW (e.g., corrosion/leakage, operating practices, buried single-wall transfer lines).

Based on the above criteria, facility operators will identify and prioritize their facility-specific safety issues. The Task Force supports EM's review of safety issues and the prioritization of issues across the DOE complex to be used in DOE program planning. Each facility operator develops a plan to resolve the safety issues at its facility. The resolution plan addresses both the need for short-term mitigation of safety issues to ensure that HLW is placed in a safe condition, and the longer-term remediation of safety issues to ensure that a safe condition will be maintained until the HLW can be treated for ultimate disposal.

The Tanks Advisory Panel reviews plans and activities to assist in the identification of safety issues and scientific needs, and to provide technical advice and recommendations, as needed.

The facility operators, Task Force, and Tanks Advisory Panel are also working together to develop an improved science and technology base for understanding HLW storage, and to ensure a timely and effective transfer of information among facilities and activities.

Validation of the Safety Envelope

Most of the Safety Analysis Reports that currently exist for the storage of HLW are not adequate in comparison with current standards for nuclear facilities. Since the time that the existing Safety Analysis Reports were written, safety standards have evolved that are more demanding and restrictive. Approaches to safety analyses have been broadened to include probabilistic as well as deterministic analyses. Guidance documents are in preparation to standardize the format and content of Safety Analysis Reports as well as analysis methodologies, definitions of unacceptable accident consequences, and to document review criteria and procedures. These modern approaches will be applied to new HLW tanks and to the revision of the existing Safety Analysis Reports to ensure the safety of continuing HLW storage.

To assist in the validation of the safety envelope, the Task Force conducts mini-reviews of facility safety documentation to guide the review and upgrade efforts of the facility operators and to perform independent reviews of safety documents meeting current standards. Facility operators have the responsibility for reviewing and upgrading their safety documents, and for comparing the upgraded documents with facility operations to ensure that the safety envelopes encompass tank operations. The Tanks Advisory Panel will review plans, activities, and outputs of the Task Force and facility operators, and provide guidance and recommendations, as necessary.

Evaluation of the Conduct of Operations

Current practice at DOE facilities utilizes Operational Readiness Reviews upon the start-up of new facilities, restart after significant facility modifications, or resumption of operations after shutdown for safety reasons. For the purposes of improving the safe operation of HLW facilities, Operations Reviews will be conducted to verify that HLW storage operations are in accordance with the limiting conditions and operating procedures defined for safe operation based upon the Safety Analysis Reports. Westinghouse Electric Company, whose subsidiaries operate all DOE HLW facilities, has agreed to take the lead to form a committee to ensure a common approach to conduct of operations, thus operational safety. EM, in conjunction with the DOE Field Offices, will develop consistent DOE conduct of operations programs and participate in joint Operations Reviews to ensure safe operation of the HLW facilities.

The Task Force and Tanks Advisory Panel support EM in developing Operations Review criteria and protocols for use by facility operators. The Task Force and Tanks Advisory Panel will also conduct independent Operations Reviews on selected operational areas. Facility operators conduct Operations Reviews of their storage operations in accordance with the criteria and protocols, and ensure that normal, off-normal, and emergency operations are in accordance with the conditions and procedures stipulated in the Safety Analysis Reports and encompassed within the established operating envelope.

Development of a Common Database

Attempts to remediate or mitigate the HLW issues will depend on accurate, accessible data. In order to share and effectively use this data, establishing site databases of known and defensible data, as well as an information network to link users to those databases, is of prime importance. Rapid retrieval of information from across the HLW storage complex will permit faster interchange of ideas, lead to a more effective use of resources for common and original problem solving, and provide DOE management with an important tool for monitoring and supervising the safe and efficient operation of its HLW tanks.

At the request of EM, Battelle Pacific Northwest Laboratory (PNL) is leading the effort to develop an integrated information network, to be implemented during fiscal year 1992, which will tie all HLW site databases together. PNL chairs a Database Working Group which has been tasked to define data, quality, system, and communications standards and requirements for the HLW tanks complex. The facility operators at the HLW Sites will take the lead in collecting data for their use.

Topical Workshops

EM is organizing a series of workshops to address different topics of importance to safe storage of HLW. Workshop topics include: surveillance and monitoring, waste characterization, safety issues, system structural integrity, establishment of safety envelope, staffing, planning for treatment and disposal, conduct of operations, and seismic design criteria. These workshops will serve to focus technical reviews on specific topics to encourage information transfer among facilities, to identify potential facility needs, and to identify areas of potential system-wide weaknesses for corrective actions.

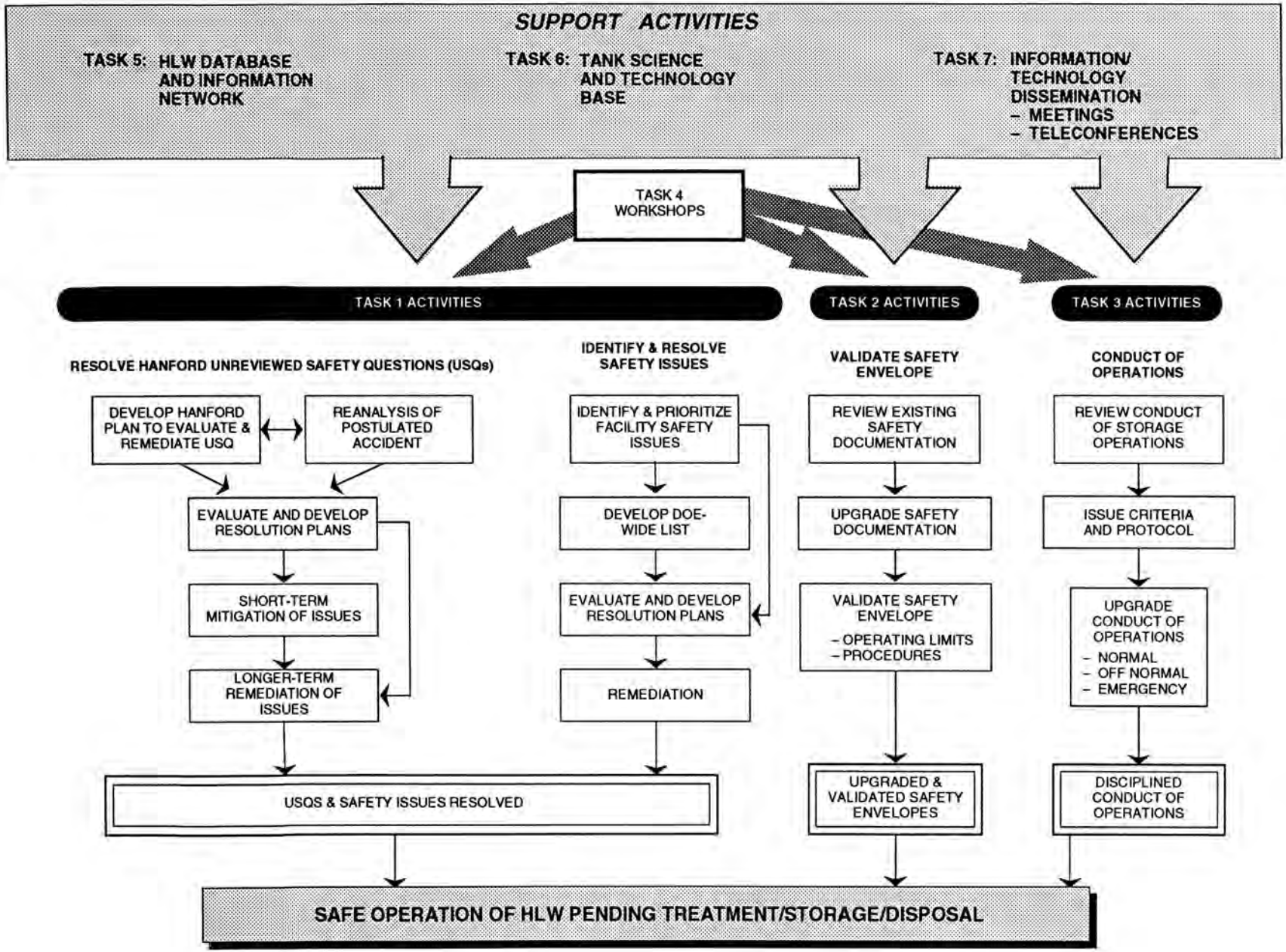

Fig. 1. DOE approach to HLW safety.

RESOLUTION OF HIGH-PRIORITY SAFETY ISSUES AT THE HANFORD SITE

Four Criteria 1 safety issues have been identified at the Hanford Site:

- flammable gas accumulation and periodic release in 5 double-shell and 18 single-shell tanks;
- ferrocyanide in 24 single-shell tanks;
- organics in 8 single-shell tanks; and
- high-heat content in 1 single-shell tank.

Detailed program plans for resolution of these safety issues are being developed by Westinghouse Hanford Company (the operating contractor at Hanford - WHC) working with the DOE Task Force. A Science Panel has been created by PNL to assist WHC in understanding the basic scientific mechanisms causing the accumulation and periodic release of hydrogen and nitrous oxide gases from 23 tanks and the potential for ferrocyanide compounds to react with nitrate and nitrite oxidants in 24 tanks. Work has begun on identifying potential mitigation methods in parallel with the basic science studies to speed up the resolution process.

In addition to the work by WHC and PNL, scientists at the Los Alamos National Laboratory and Brookhaven National Laboratory are conducting, at the request of EM, a "bottom-up" reanalysis to better understand possible chemical reactions affecting tank contents, potential accident initiators and consequences, and probability of occurrence. The Hanford and Los Alamos National Laboratory/Brookhaven National Laboratory efforts together will assist EM in developing approaches which are scientifically sound and technologically feasible to make the tank contents safe and to reduce the risks to acceptable levels.

The four highest priority safety issues at the Hanford Site are discussed in the following sections, and current status of the resolution of each issue is briefly described.

Flammable Gas Accumulation and Periodic Venting

Five double-shell tanks and eighteen single-shell tanks have been identified as having a significant potential for flammable gas generation/accumulation and periodic gas releases. The simultaneous presence of a flammable gas mixture and an ignition source would pose the potential for a deflagration that could lead to increased consequences of the release of radioactive waste. Operational restrictions to reduce the potential for sparks and ignition sources have been imposed for the 23 waste tanks. Of lower potential risk is an unfiltered release of radioactivity to the environment from overpressurizing the ventilation system during a periodic gas release.

The waste tank that has been the most active, Tank 241-SY-101 (referred to as Tank 101-SY), has released, during several of its venting periods, concentrations of hydrogen that for a short period of time exceeded the amount necessary to support combustion. During these releases, overpressurizations have been measured without resulting in any contamination spread. The venting of gases is expected to *reoccur periodically* until some form of mitigation or remediation is implemented.

Potential accident sequences in Tank 101-SY have been extensively analyzed. The worst postulated accident in this tank is a hydrogen deflagration which could ignite the crust on the waste's surface, leading to large releases of radioactivity. However, data from analyses of crust samples indicate that the moisture content of the crust is sufficient to preclude wide-spread crust ignition. Further, it is believed that the probability for hydrogen combustion is low because operating procedural controls are adequate to prevent the possible presence of potential ignition sources in tanks, and temperatures are well below those required to initiate chemical reactions.

Several crust samples and one core sample were taken in May 1991, and an additional core sample was obtained in December 1991. Data obtained from the analyses of these samples, together with the results from laboratory studies with synthetic waste, are providing us with the technical basis for formulating and selecting mitigation options for near-term implementation. These data will also allow us to consider the integration of remediation options with pretreatment required prior to vitrification of the high-level fraction for future repository disposal.

Mitigation options focus on minimizing gas accumulation in the waste, thus preventing periodic venting of flammable gases. Some of the mitigation options under consideration include: (1) stirring and/or mixing to release gas bubbles; (2) transferring a portion of the waste to other double-shell tanks and then diluting and circulating it in the affected tanks; and (3) heating or using ultrasonic devices to release trapped gas bubbles. It is critically important that the selected mitigation option does not preclude options for the waste retrieval and pretreatment that will be necessary to prepare the waste for vitrification.

Remediation measures would be aimed at interrupting the flammable gas generating mechanism in the waste. It is expected to take one to two years to complete and correlate data from analyses of core samples and laboratory synthetic waste studies to identify feasible remediation measures. This would also be the timeframe that DOE would be considering for possible pretreatment options for waste in Tank 101-SY for vitrification.

To date, the initial activity is focused on Tank 101-SY because it exhibits the largest level fluctuation. We expect knowledge gained from Tank 101-SY would be directly applicable to the 22 other flammable gas tanks. The sampling and characterization program for the other tanks are in the planning stage prior to implementation.

Potential Explosive Mixtures of Ferrocyanide

Twenty-four single-shell tanks may contain appreciable amounts of ferrocyanide precipitates. The estimated ferrocyanide content of the 24 tanks ranges from 1,000 g-moles (465 pounds) to approximately 200,000 g-moles (93,000 pounds) in Tank 241-BY-104 (referred to as Tank 104-BY). To quickly obtain additional waste storage volume during the early 1950s, a process was developed to reduce radionuclide concentration in the freestanding liquid tank wastes to levels low enough to permit disposal of the liquid to the soil. This process involved the addition of ferrocyanide-containing compounds to the tanks to co-precipitate radioactive cesium out of the liquid. Ferrocyanide salts in the presence of nitrate and/or nitrite constituents (which these tanks contain) can react and detonate under certain conditions that include dryness, proper chemical concentration and mixture, and elevated temperatures or a high-energy spark. Such reactions can occur at 285°C.

A 1990 General Accounting Office (GAO)-sponsored study (2) postulated dose consequences that were up to two orders of magnitude greater than stated in the 1987 "Final Environmental Impact Statement - Disposal of Hanford Defense High-Level, Transuranic and Tank Wastes"(3) for a postulated accident. Such a release is believed to have a low probability of occurrence because the maximum temperatures measured inside the ferrocyanide tanks at the Hanford Site are at or below 60°C, thus providing a significant margin of safety. Tank temperatures have also been declining at a little less than 2°C per year as the tank fission products continue to decay.

An aggressive program has been implemented to collect in-tank data, conduct laboratory studies with synthetic waste, and perform model analysis. Preparation is ongoing to collect crust and core samples from selected ferrocyanide tanks. These core sample data will be used to validate data from synthetic waste studies, which include small-scale thermal properties studies and large-scale explosive testing.

Ongoing model studies include thermal (hot spot) analysis, source term evaluation, and particle size generation. Improved methods for in-tank data collection are also being developed, such as gamma spectral measurements as a function of depth in the waste sludge, and infra-red scanning to determine the possible presence of a hot spot. Also, work is planned to ensure that measured temperatures are representative of the entire tank contents. This will further ensure that the tank contents are maintained well below temperatures that can cause exothermic reactions.

If the probability for a ferrocyanide explosion is found to be low, it might be acceptable to continue the current mode of storage. However, 66 of the 149 single-shell tanks at Hanford have either leaked or are suspected to be leaking (13 of the 24 ferrocyanide tanks are leaking or assumed leakers). Hanford has an ongoing program to pump all drainable liquid from all single-shell tanks to double-shell tanks to minimize the potential for environmental contamination; pumping of ferrocyanide tanks was stopped in September 1990 since water content is a significant factor in minimizing the possibility of a ferrocyanide reaction. Safety studies are ongoing to assist contingency planning in the event that one of the ferrocyanide tanks is determined to be leaking, and to support reconsideration of the DOE decision not to stabilize the remaining ferrocyanide tanks.

Options are being considered to remediate the ferrocyanide tanks, including removal of the waste and separating the reactive waste constituents (nitrates and/or nitrites and ferrocyanide) from each other. Planning for remediation will also be integrated with planning for retrieval/pretreatment of the waste to prepare for vitrification.

Potential Organic-Nitrate Reactions

Eight single-shell tanks may contain unacceptable concentrations of organic chemicals (e.g., more than 3 percent dry weight sodium acetate equivalent). The actual concentrations of organic materials in these eight single-shell tanks and their chemical identities are not known at present. However, very concentrated organics could support an exothermic reaction at temperatures greater than 180°C.

Under a scenario involving significant overheating (to greater than 180°C) of waste in the tanks, it may be postulated that a mixture of organic-nitrate/nitrite solids might react rapidly, possibly damaging the tank and leading to the release of radioactive materials to the environment. This is believed to have a very low probability of occurring, since the margin between minimum ignition temperature and measured tank temperatures is very large.

DOE is in the process of developing a program to study the organic safety issue. Two of the options being considered to address this safety issue include: (1) demonstrating that the probability of an explosion is acceptably small and that no action is required; and (2) removing the waste and separating the oxidizer material (nitrates and/or nitrites) from the organic constituents.

High-Heat Generation in Tank 106-C

Tank 241-C-106 (referred to as Tank 106-C) is a 2.0-million-liter- (530,000-gallon-) capacity single-shell tank. The tank has been used for HLW storage since mid-1947 and currently is approximately half-full of waste. In the late 1960s, during the implementation of a program to recover heat-generating strontium and cesium from HLW in some of the single-shell tanks, heat-generating sludge was inadvertently transferred to Tank 106-C. Since mid-1971, water has been added periodically to Tank 106-C to keep the sludge wet and to promote heat removal by evaporative cooling to the vapor space. The cooling water is in addition to the other drainable liquids in the tank.

Although Tank 106-C is currently not a declared leaker, if it should leak, the addition of cooling water could increase the amount of leakage to the ground. If cooling water additions to the tank are stopped, the sludge will heat to temperatures greater than established limits and may cause tank structural damage, possibly leading to an unacceptable radioactive release to the environment. A Tri-Party Agreement milestone has been established to remove all the drainable liquid (which would eliminate the use of cooling water) and interim isolate Tank 106-C by September 1996.

Two options are being considered to address this high-heat generation safety issue in Tank 106-C: (1) partially remove, to double-shell tanks, sufficient contents to reduce heat generation in Tank 106-C to an acceptable limit; and (2) provide a mechanical means of cooling the radioactive waste. In addition, DOE is aggressively investigating the option of using waste from Tank 106-C as feed for processing in the proposed Hanford Waste Vitrification Project. The waste retrieval study is at the conceptual stage as the proven jet pumping removal process demonstrated at the Savannah River Site and the West Valley Demonstration Project would not be applicable to Tank 106-C, given concern with tank wall integrity.

SUMMARY

The above material provides a glimpse into the complex nature of the DOE high-level radioactive waste problems facing the managers of these aging waste management facilities. A system is in place and progress is being made in understanding the nature of some of the most vexing problems for resolution.

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