TECHNOLOGY DEVELOPMENT TO SUPPORT HANFORD SITE TANK WASTE REMEDIATION SYSTEM OBJECTIVES

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ABSTRACT

Approximately 227,000 m^3 (60 Mgal) of highly radioactive, transuranic, and hazardous waste exist at the U.S. Department of Energy's Hanford Site in Washington State. These waste in the form of caustic liquids, slurries, saltcakes, and sludges are stored in 177 underground tanks (149 single-shell and 28 double-shell). Also, significant quantities of ^{90}Sr and ^{137}Cs are present in metal capsules stored in water basins. The total activity in the waste is estimated to be 420 Mci. Many of the single-shell tanks have leaked and the condition of others is suspect. In addition, recent identification of potentially flammable gases and explosive solid mixtures have caused concern for the safety of the tanks.

The 1988 Record of Decision stated that the double-shell tank waste would be retrieved, treated, and disposed and the Sr/Cs capsules would be overpacked and sent to the geologic repository. Low-level waste would be grouted and stored onsite and high-level waste would be vitrified (i.e., borosilicate glass) and also sent to the repository. Disposition of single-shell tank wastes would be addressed through a separate and future Environmental Impact Statement. In an effort to determine a strategy to safely and cost-effectively mitigate the safety concerns and ultimately dispose of the Hanford Site Tank Wastes, the U.S. Department of Energy, Environmental Protection Agency, and Washington State Department of Ecology created a pact to address and resolve the issues.

During 1991, it was decided to combine the single-shell tank and double-shell tank efforts into one program. The Tank Waste Remediation System program was established to store, treat and dispose of tank and some ancillary wastes. Major efforts have been expended to convene teams of national technological experts to identify and prioritize technologies that could be used to perform the Tank Waste Remediation System process functions. A decision-making methodology was developed that allowed the experts to evaluate technologies against a set of common criteria.

The TWRS Program is currently developing a new technical strategy to address the new scope. A systems-engineering and analysis approach will be used to judge the merit of each technology by assessing its affect on the total system. A technology plan will be created to describe technology development, testing, and implementation. The technology program will increase the probability of success of the Tank Waste Remediation System program by identifying and testing essential technologies, identifying vulnerabilities and testing contingent technologies, identifying and testing technologies common to a variety of processes, and by supplying data necessary to perform valid system engineering and analysis studies. This paper describes the decision process and initial technology prioritization results.

INTRODUCTION

Technology development for the Tank Waste Remediation System program is largely driven by the unique situation regarding the Hanford Site radioactive wastes. An "idealized strategy" for the ultimate disposal of tank waste at the Site is essentially the same as at nuclear fuel reprocessing sites elsewhere in the United States, as well as in Europe and Japan. These are (1) the encapsulation of the highly radioactive fission product and transuranic (TRU) wastes in a minimum volume of a very stable solid form, (i.e., one that will retain its integrity and insolubility for many millennia, and storage of this waste in a geologic repository), (2) the immobilization of remaining process waste solids with essentially no radioactivity (albeit probably a "hazardous" waste because of the chemical constituents) and the disposal in a Resource Conservation and Recovery Act-approved manner (1), and (3) the reduction of the activity in the residual water and gas streams to a level that can be released to the environment.

Immobilization of high-level radioactive waste is underway in two European countries (i.e., Britain and France). These foreign reprocessing sites segregate the waste from the first cycle of extraction of uranium/plutonium products from the fission products and thus have a waste stream that contains perhaps 99 percent of the fission products together with very small quantities of well-characterized process chemicals.

For several reasons, Hanford Site operations usually did not segregate the first cycle stream. Early separation processes resulted in very large volumes of waste and some of these were subsequently further processed to recover uranium. In addition, select fission products were also recovered. These operations further increased the volume of waste and this, together with shortages of tank volume, has resulted in extensive mixing of the highly concentrated fission product waste with a great variety of process chemical wastes. The concomitant neutralization and concentration of the waste have produced saturated slurries, solid salt cakes, and sludge. The result is an inventory of waste with a very high content of solids and great complexity.

BACKGROUND

The radioactive waste stored in tanks has come from various sources: (1) three different plutonium and uranium recovery processes involving approximately 100,000 MTU of irradiated fuel, (2) three different radionuclide recovery processes, and (3) miscellaneous sources (e.g., laboratories and reactor decontamination solutions). The neutralized wastes include sodium nitrate/nitrite, sodium hydroxide, sodium aluminate, sodium phosphate, and large amount of organics.

The single-shell tanks (SST) consist of reinforced concrete tanks with carbon-steel liners and have capacities ranging from 208 m^3 (55,000 gal) to 3,785 m^3 (1 Mgal). The double-shell tanks (DST) consist of carbon-steel tanks within steel-lined concrete tanks, each have a nominal capacity of 3,785 m^3 (1 Mgal). Sixty-seven the SSTs have leaked or are suspected to have leaked approximately 3,785 m^3 (1 Mgal). No waste has been added to the SSTs since 1980, and the pumpable liquids are being removed. The remaining waste will be primarily sludge and saltcake. None of the DSTs has leaked since first placed into service in 1971.

Miscellaneous waste covered within the scope of Tank Waste Remediation System (TWRS) include approximately 1900 doubly encapsulated metal containers, containing ^{90}Sr and ^{137}Cs salts, and possibly 2100 tonnes of irradiated N-reactor fuel. These materials are currently stored in water basins.

Tank safety issues center around the following: 1) hydrogen generation and potentially explosive mixtures, 2) high-heat tanks containing fission products, and 3) tanks containing ferrocyanide, and nitrate salts which could potentially create an explosion at elevated temperatures.

TANK WASTE REMEDIATION SYSTEM SCOPE

The TWRS program encompasses projects and activities for receiving, safely storing, maintaining, treating, and disposing of onsite or packaging for offsite disposal all tank wastes, capsules, and irradiated fuel. The scope includes existing facilities, such as waste storage tanks, evaporators, pipelines, planned pilot plant facilities, and the Grout low-level waste (LLW) treatment and disposal facilities. In addition, upgrades to existing facilities and equipment and new facilities are being considered. Major facility additions will include the Hanford Waste Vitrification Plant (HWVP), Multi-Purpose Storage Facility, Multi-waste tank facility, and the Initial Pretreatment Facility.

Closure (i.e., final disposal) of the SST and DST sites is not included in the program. Also, development of a geologic repository for disposal of high-level waste (HLW) is the responsibility of the U.S. Department of Energy's (DOE) Office of Civilian Radioactive Waste Management.

The TWRS is shown in Fig. 1. All tank waste will be retrieved. The strontium and cesium capsules will be overpacked and shipped to the geologic repository for disposal. The waste will be separated into two fractions, with the HLW fraction vitrified and disposed in the geologic repository and the LLW fraction grouted for disposal onsite. Both the irradiated fuel and the HLW glass canisters will be stored onsite temporarily in the Multi-Purpose Storage Complex. Environmental impact statements will be prepared for TWRS and the irradiated fuel.

TECHNOLOGY IDENTIFICATION AND EVALUATION PROCESS

The TWRS program was divided into logical elements: tank safety and operations, characterization, retrieval, pretreatment, LLW, and HLW. Teams of national experts (technology working groups), for each program element, convened

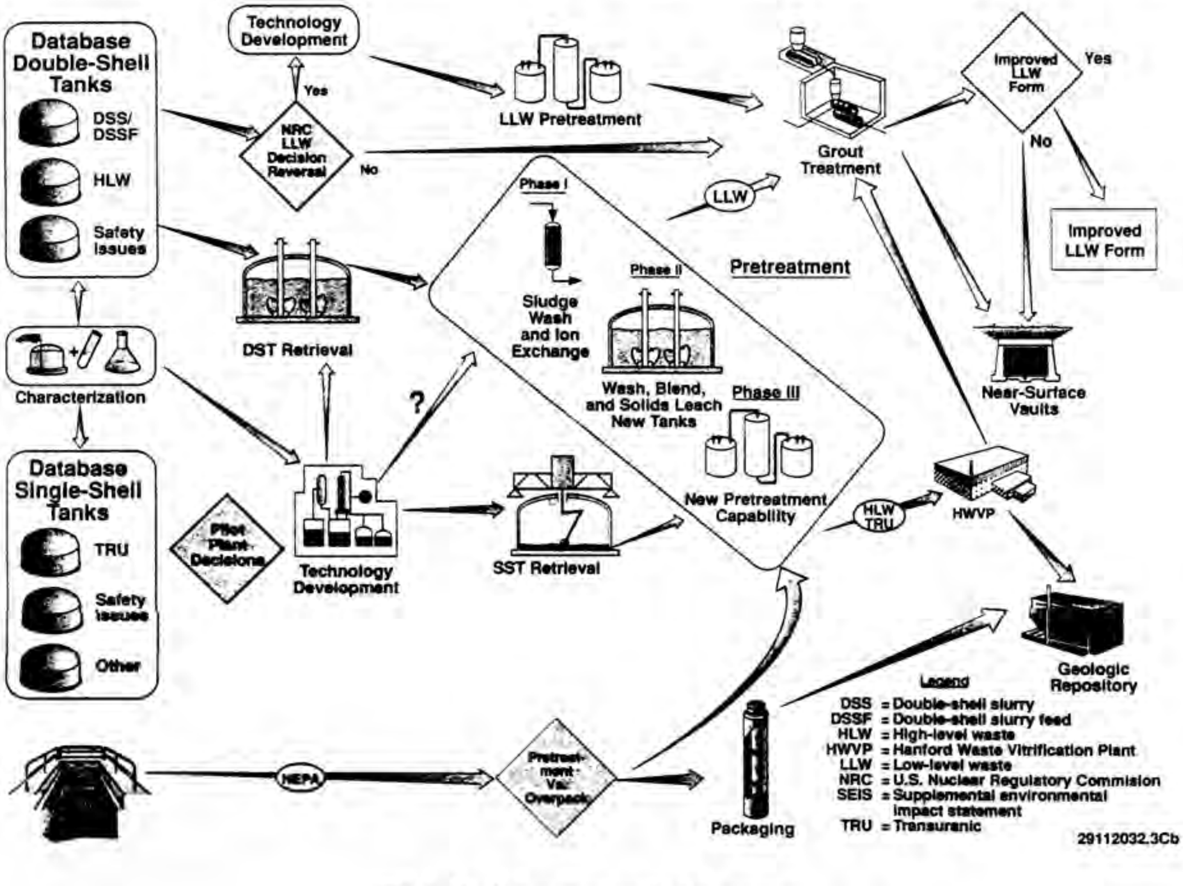

Fig. 1. Tank waste remediation system.

to identify process functions and requirements and prioritize technologies to perform these functions. A systems engineering and analysis approach and a set of standard criteria were used to evaluate the affect of each technology on the total system. Where data was lacking expert judgement and consensus was substituted. The general process is shown in Fig. 2.

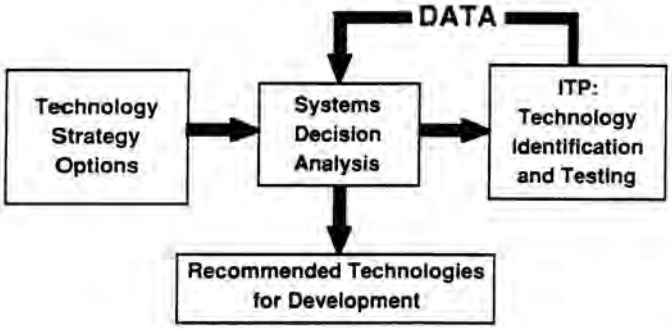

Fig. 2. Systems analysis approach.

Process Function Criteria

Because the approach to resolving the waste problem is not well defined, various processing scenarios were formulated to accomplish the overall objectives. Three criteria, which were used to judge process functions, were as follows:

- Centrality--Is the process function necessary to achieve overall objectives?
- Uncertainty reduction--Will the process function reduce uncertainties associated with achieving TWRS objectives?
- Urgency--Is the process function necessary to achieve program milestones?

Technology Criteria

The list of potential criteria was shortened to the following five: technological feasibility, life cycle and capital costs, environmental/safety/health impacts, waste minimization, and schedule performance.

- Technical feasibility--Will the technology perform the function?
- Life cycle costs--What are the relative total system costs for the life of the program to include disposal?
- Capital costs--What are the relative capital construction costs?
- Environmental/Safety/Health--What are the acute and chronic impacts?
- Waste minimization--What is the magnitude of total waste disposed to include primary and secondary waste streams?
- Schedule performance-When can the technology be implemented?

Judgments of high, medium, and low were assigned values of 100, 50, and 0 to obtain a numerical ranking. The criteria were weighted equally for this exercise.

Areas of uncertainty or vulnerability were also identified and contingent strategies and technologies recommended.

Because of numerous program uncertainties and constraints, the technology development program was structured to ensure the greatest probability of successfully satisfying the TWRS objectives. Essentially, there are three levels of technology development.

- Reference--Technology required to implement the baseline processes within the program strategy
- Enhanced--Technologies (within the program strategy) to the baseline that: (1) provide backup technology where baseline technology has high uncertainty, and (2) could achieve major improvements and potentially become the baseline.
- Alternative--Technologies in support of significantly different program strategies, which may make very significant improvement in the Program (i.e., significant life cycle cost reduction), but have major impacts to the current reference strategy.

RESULTS

A summary description of the functions and technologies is presented in Fig. 3.

Tank Safety and Operations

The process functions include the following: receipt, storage, and limited treatment of the waste, identification and resolution of safety issues, response to emergency conditions, disposition of safety issues, and limited waste characterization. The current safety issues include the following: flammable gases (hydrogen), potential explosive mixtures (ferrocyanide and organics), noxious vapors, high-heat (radiolytic decay), and criticality. Technologies associated with performing these functions as well as monitoring the wastes (i.e., gaseous, liquid, and solid) and evaluating the integrity of the tanks need to be developed and implemented.

The reference process for mitigating hydrogen will be to use a mixer pump to agitate the tank contents and thereby exhaust gases continuously below the explosive limits. Alternative options include diluting and pumping the contents to other tanks. The reference strategies for resolving ferrocyanide, organics, and high-heat conditions involve retrieving the waste for processing.

In situations where the conditions may be mitigated in-tank, limited technology development will be required. Technology development will be necessary primarily in instances where safety resolution occurs subsequent to removing the tank contents for processing and in monitoring and characterizing waste.

If leaking tanks are identified, then the means of removing liquid waste from the tanks need to be implemented. Technologies associated with limiting the potential impact to the environment will have the greatest merit. Subsurface barrier technologies also must be considered in this regard.

Technology vulnerabilities include the following: inability to predict, monitor, and control leaks, inability of the mixer pump to dissipate hydrogen in a 1 Mgal tank, inadequacy of analytical equipment to characterize waste in a timely manner, and inability to perform non-destructive testing on tanks to evaluate structural integrity.

Characterization

Characterization includes the quantitative physical, chemical, and radiological description of underground

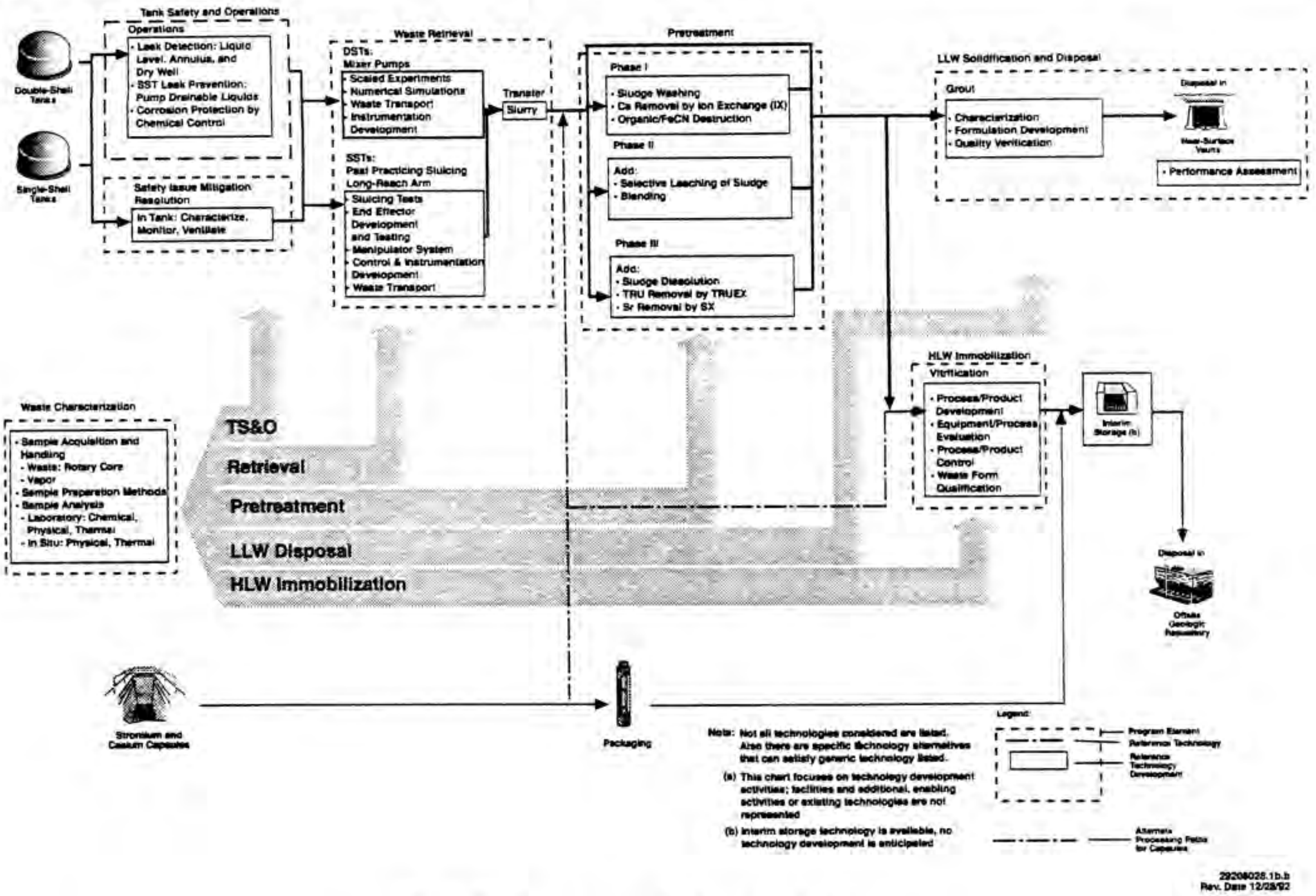

Fig. 3. Tank waste remediation technical strategy.

storage tank waste and waste products, process streams, and disposal forms. Characterization satisfies predetermined data quality objectives for making process, disposal and safety risk based decisions.

Included in the reference case are the functions of (1) sample acquisition and handling, (2) sample preparation, (3) laboratory analysis (chemical, radiochemical, physical, and thermal), (4) in-situ analysis (physical and thermal), and (5) data management. Enhancements to the reference case include improved throughput of analyses through the analytical laboratory by automation and improved analytical quality by obtaining more representative samples, preserving sampling integrity after removal from tank, and developing field deployable laboratory units. Alternatives to the reference case require an entirely different approach. The alternative case is broadly classified into technologies that employ rapid scanning of samples in a hot cell and in-situ chemical and radiochemical measurement systems.

Specific analyses include the following: metal ions using Inductively Coupled Plasma Mass/Atomic Emission Spectroscopy (ICP-MS, ICP-AES) and atomic adsorption, anions using spectrophotometric methods, radionuclides, total organic carbon, volatile organic analytes (VOA), ferrocyanide, noble metals (using ICP-MS), neutrons, alpha, beta, weak beta, and x-ray.

The highest priority functional needs can be classified as follows: (1) in-situ measurement of thermal, rheological, and physical properties using acoustic, remote relative viscometer, and thermoelectric AC modulation, (2) hot cell segment scanning using laser scanning fluorescence, laser ablation ICP-MS/ICP-AES, laser Raman and infrared spectroscopy, gamma energy analysis (GEA), and dedicated testbed, and (3) obtaining representative samples using robotic mechanical manipulators, gas tight sampler, fluidized bed sampling, Summa canisters, adsorbent tubes, and permeable membrane with ion trap.

The principal vulnerability is defining the Data Quality Objective needs for TWRS.

Retrieval

Waste retrieval activities are associated with removing and transporting wastes and in-tank hardware from both the DST and SST tanks. The functions associated with this endeavor can be categorized as follows:

- Confine the waste
- Control the system
- Monitor the process
- Deploy the system
- Access the tanks
- Mobilize waste
- Convey waste
- Process waste
- Transport waste
- Characterize waste and environment
- Dispose of in-tank hardware
- Protect the tanks

- Control the tank environment.

Most of the drainable liquid has been pumped from the SSTs, leaving hard salt cake, thick sludge, and combinations of these forms. The DST waste is stored in dilute form with sludge deposits in the bottom of the tanks. Access to most of the SSTs is limited to single 106.7-cm (42-in.) diameter ports. Access to the DSTs includes two 106.7-cm (42-in.) risers. Both DSTs and SSTs have in-tank hardware that could obstruct access to the waste and inhibit slurry efforts. Other miscellaneous materials in the SSTs include experimental fuel elements, irradiation sources, and failed in-tank equipment.

Retrieval technologies for SST waste include the following: methods to mobilize the waste (i.e., water jetting, air jetting, water jet bore mining, sluicing, localized melting, rheology modifications and pumping, and localized chemical adding), deployable retrieval systems (e.g., long-reach manipulator arm with end-effectors, tethered systems, and tool change-out system), control of the retrieval process (e.g., dynamic control, fault detection, position control, and retrieval process control), confinement of waste during retrieval operations (e.g., freeze barriers, grout barriers, soil liquefaction-polymer injection, and nonleaking fluids), waste conveyance out of the tank (e.g., air conveyance, pumping, screws, and buckets), installation of new tank penetrations, [e.g., diamond saws, core drill, abrasive water jets, welding, grouting (all remote operations)], and monitoring the retrieval operations (e.g., video, ultrasonics, acoustics, infrared vision systems, laser position, gamma spectroscopy, near infrared absorbance and backscatter, and gamma emission scanning).

Retrieval technologies for DST waste include the same basic functions as SST waste. Waste slurry mixing and pumping is the preferred retrieval method. Instrumentation systems to monitor the retrieval process and evaluate slurry mixing uniformity and cleaning radii are required. High shear-strength sludges may require alternative solutions, if the current mixer pump concept proves inadequate.

Reference technologies encompass mixer pumps, past-practice sluicing, and long-reach manipulators. Enhancements and alternatives include using stop leak gel, large tank openings, open pit and/or hydraulic mining, confined sluicing; and melting or vaporizing the waste.

Technical uncertainties include the following: control of the manipulator arm, feasibility of tank access, imposition of dome and vibration loads, level of heat generation, percent recovery, amount of waste generated, amount of tank waste leakage, and use of organic fluids.

Pretreatment

Pretreatment functions are associated with preparing the wastes for subsequent immobilization and disposal. Pretreating the wastes to resolve safety issues such as organic and ferrocyanide destruction is also considered. The functions for which technologies will be developed and implemented, include the following: waste blending, sludge wash and acid dissolution, solid/liquid separation, organic/ferrocyanide/nitrate/nitrite destruction, selective and bulk leaching, and separation of Cs, Sr, transuranics, Tc, and I. Technologies need to be investigated to perform these functions on both basic and acidic solutions.

The reference technology for transuranic separation is the "TRUEX" solvent extraction process on the acid side. However, extraction chromatography is also being studied. Cs removal in alkaline solutions using CS-100 or resorcinol resins is preferred, but recent developments in the use of silico-titanates are promising. Organic destruction by means of ozonation, calcination, steam reforming, and hydro-thermal processing is being evaluated. However, those technologies which can be used to simultaneously destroy a hazardous compounds may be preferred. Magnetic separation may also have merit in the initial stages of separation. Leaching of certain non-radioactive metal ions, from the high-level waste, may also be implemented to reduce the number of glass canisters. Enhancements and alternatives to the reference technologies will result in a reduced number of high-level waste canisters being produced. A "Clean" option is also being considered, which could result in the minimal production of both high-level and low-level waste.

Various implementation strategies also are being considered. These range from centralized facilities, incorporating either predefined or modular design concepts, to distributed processing units located at the tanks and operated remotely.

Major technological vulnerabilities are associated with the degree of dissolution achievable, the impact of certain chemical species on the efficiencies of both solvent extraction and ion-exchange technologies, and the implementation of cost-effective technologies in a timely manner.

Low-Level Waste Immobilization

LLW functions include the following: storage and transfer, characterization, waste form processing, transportation, and disposal. The current reference final waste form is grout. Waste is transferred to the grout feed tank, where it is blended to satisfy processing requirements, and characterized. Dry raw materials are preblended and then mixed with the liquid waste to form a slurry. The slurry is pumped to underground vaults for final disposal on-site.

Technology development is required in the following areas: in-tank sampling and analysis, product quality control, grout performance assessment, predicting and measuring the quantity of hydrogen gas generated by radiolytic and chemical decomposition, barrier improvement and testing, process control, modeling long-term durability, increased processing capacity and product uniformity. Alternative waste forms are also being considered, such as glass, mineral grout, ceramic, polymer and combinations of these.

Technical uncertainties are associated with formulation, performance assessment, process control, feed variability, facility decontamination and maintenance, and rework of the grout process.

High-Level Waste Immobilization

The HLW immobilization processes HLW into a form suitable for interim storage and ultimate disposal in a geologic repository. The reference process is vitrification and the waste form is borosilicate glass. Seven basic functions were identified: storage and characterization, receipt and feed preparation, immobilization, process control/analytical support, waste effluent treatment, packaging, and storage.

Technology development is necessary in the areas of waste form qualification, influence of noble metals on melter performance, impact of waste feed materials compositional variability, offgas system performance, and product/process control systems. Potential enhancements and alternatives include greater production rate melters, improved waste form (i.e., ceramic, phosphate/sodium glass), and increased waste loading.

Technological uncertainties include the following: the ability to meet waste acceptance specifications, ability to process feed, plant capacity, ability to achieve waste loading goals, melter life expectancy, hydrogen/ammonia generation and control, rheology/redox control, composition and properties of organic feed, solids accumulation in offgas system, iodine removal from offgas, and ability to rework off-standard products.

CONCLUSIONS

The TWRS program is extremely large and complex with a multitude of uncertainties. A systematic approach in identifying technologies to be developed and implemented is essential. However, a scarcity of necessary and sufficient data requires a comprehensive technology testing program to obtain the information to be used in the systems engineering studies.

Because the TWRS program is currently being rebaselined, the final selected processes and technologies remain to be determined. Many multifunction and common technologies were identified that could be developed with minimal risk that they would be superseded by a change in technical strategy. These include in-situ analyses, subsurface barriers; manipulator arms; sluicing, pumping, or blending wastes; actinide/cesium/technetium/strontium separation; organic/nitrate/ferrocyanide destruction; grout performance improvement; increased waste loading; and improved HLW melter performance.

The use of technology working groups comprised of national experts is invaluable. The working groups should be convened annually to review the TWRS technology status and potential benefit of new technologies to the program.

The uncertainties present in the TWRS program require that a limited number of technology options be developed in parallel, until such time as the program is well defined and the competing technologies understood. In the early stages, this could be accomplished through comprehensive laboratory testing, pilot-plants, and demonstration programs. Pilot-plants testing should be reserved for those technologies with the highest probability of being implemented in production.

Technology development should not be limited to Hanford Site assets. National assets throughout the DOE complex, private industry, and universities should be used to the maximum extent possible; and foreign technology exchanges also should be continued.

REFERENCES

1. Resource Conservation and Recovery Act of 1976, 42 USC 6901 et seq.