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TRANSURANIC WASTE FORM CHARACTERIZATION AND DATA BASE

EXECUTIVE SUMMARY

Submitted to:

Dr. Henry Cheung
Lawrence Livermore Laboratory
P.O. Box 808, L-390
Livermore, California 94550

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TERA ADVANCED SERVICES CORPORATION

2150 Shattuck Avenue
Berkeley, California 94704
415-845-5200

Berkeley, California
Dallas, Texas
Bethesda, Maryland
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EXECUTIVE SUMMARY

The Transuranic Waste Form Characterization and Data Base (Volume 1) provides a wide range of information from which a comprehensive data base can be established and from which standards and criteria can be developed for the present NRC waste management program.

Supplementary information on each of the areas discussed in Volume 1 is presented in Appendices A through K (Volumes 2 and 3). The following outline establishes the structure of the study (Volume 1) and identifies which appendices of Volumes 2 and 3 correlate with each main section of the report.

<u>Section (Volume 1)</u>	<u>Appendices (Volumes 2 & 3)</u>
1 Introduction	A
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3 Characteristics of Wastes Generated by the Nuclear Industry	I & J
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The Executive Summary reviews the sources, quantities, characteristics and treatment of transuranic wastes in the United States. Due to the variety of potential treatment processes for transuranic wastes, the end products for long-term storage may have corresponding variations in quantities and characteristics.

1.0 SOURCES AND QUANTITY OF TRANSURANIC WASTES

The largest potential radiological hazard associated with wastes from the nuclear fuel cycle is the high-level waste stream from fuel reprocessing or the spent fuel assemblies in the absence of reprocessing. The second source of solid transuranic wastes, including materials such as cladding hulls, represent a problem comparable to that posed by isolating the actinide component of the high-level waste stream. Most of these wastes comes from the Department of Energy's reactors and facilities, commercial nuclear power plants, and federal (military) and commercial nuclear fuel cycle activities--mainly fuel fabrication and reprocessing facilities. No commercial fuel reprocessing facilities are currently operating; however, the Department of Energy has four fuel reprocessing facilities either in operation or on standby. The current regulatory position, as well as the definition of transuranic waste sources and inventory, is based upon a requirement for retrievable storage of all wastes with a transuranium content of 10 nanocuries per gram or higher.

To adequately define the sources of transuranic wastes in the U.S., it is necessary to identify all present inventories of such wastes, as well as the facilities which generate or may generate such wastes in the future. At present only eight Department of Energy facilities are burying or storing transuranic wastes. These are the Hanford Reservation, Idaho National Engineering Laboratory, Los Alamos Scientific Laboratory, Nevada Test Site, Oak Ridge National Laboratory, Pantex, Sandia Laboratory, and Savannah River Laboratories.

The list of the Department of Energy's facilities generating transuranic wastes is much larger. These wastes include almost anything which is used in the reprocessing of spent fuel, fabrication of nuclear weapons, development of advanced reactor designs, and a myriad of R&D activities.

Up to the present time, solid wastes containing transuranic nuclides have been disposed of at five of the six commercial burial sites.* These wastes were generated by industry and, to a limited extent, by the Department of Energy and its predecessors AEC and ERDA. These sites have accepted wastes generated in the fuel cycle, primarily fuel fabrication and spent fuel reprocessing, as well as from numerous R&D activities and small commercial uses of transuranic isotopes. The present commercial inventory of transuranic wastes is limited to DOE-generated wastes (shipments now terminated) and R&D fuel fabrication activities involving plutonium. The NRC does not have records on the volume of commercial transuranic wastes buried at commercial shallow land burial sites, but estimates that about 123.4 kilograms of plutonium were buried at five of the six sites through 1977. The best available information indicates that the cumulative DOE-generated wastes (through 1976) shipped to these commercial burial grounds contained about 7.6 kilograms of the 123.4 kilograms transuranic isotopes.

The inventory (as of October 1, 1978) of transuranic wastes, and contaminated soils and sediment is as follows:

	<u>Transuranic Mass</u>	<u>Volume</u>
Retrievably stored - DOE	494 kg	$4.9 \times 10^4 \text{ m}^3$
Previously buried - DOE	759 kg	$2.7 \times 10^5 \text{ m}^3$
Contaminated soils and sediments - DOE	190 kg	$1.2 \times 10^6 \text{ m}^3$
Previously buried - Commercial	123 kg	unknown

* Beatty, Nev.; West Valley, N.Y.; Maxey Flats, Ky.; Richland, Wash.; Sheffield, Ill.; Barnwell, S.C. (no transuranic waste except small quantities of Am-241 at Barnwell).

As indicated above, a total of 0.32 million cubic meters of transuranic solid wastes is in storage at Department of Energy sites. By October 1984, it is anticipated that the retrievable inventory will increase by a factor of nearly two, while the wastes in burial sites will remain unchanged. Consequently, the change in total inventory by 1984 represents an increase of about 15 percent. The commercial transuranic waste inventory is not expected to increase significantly, unless reprocessing of spent fuel becomes a national policy. According to an analysis by the Department of Energy, wastes generated by commercial facilities are estimated to increase approximately 280 cubic meters a year.

2.0 TRANSURANIC WASTES GENERATED BY THE NUCLEAR INDUSTRY AND COMMERCIAL OPERATIONS

The commercial nuclear industry generates wastes resulting from many nuclear fuel cycle activities. In addition, this report reviews nonfuel cycle waste sources and other potential sources. Most of the transuranic wastes are currently generated by the "back end" of the nuclear fuel cycle, while in a plutonium-recycle fuel cycle the transuranic wastes are generated throughout the fuel cycle: the fuel fabrication industry, the spent fuel reprocessing industry, and fuel enrichment facilities.

FUEL FABRICATION INDUSTRY

The United States commercial nuclear power industry is presently supported by seven major uranium fuel fabricators who process low enriched uranium hexafluoride (UF_6) into finished fuel assemblies of uranium oxide pellets clad in zircaloy tubing. Their total annual production capacity has been estimated to be 50-75 MT/yr. One additional MOX fuel rod plant of commercial production capacity is in the planning stage.

In general, solid wastes associated with fuel fabrication arise from the processing and contamination control efforts consist of paper (wipes and cartons) and plastic (bags).

SPENT FUEL REPROCESSING INDUSTRY

Following a cycle of exposure in the light-water reactor, the spent fuel can be chemically processed to recover the unburned uranium and plutonium and to separate the fission products for storage or ultimate disposal.

Solid wastes from reprocessing plants consist of cladding hulls, intermediate- and low-level liquid wastes, general trash, equipment and dry chemicals.

FUEL ENRICHMENT FACILITIES

While laser enrichment and gas centrifugation offer alternatives for future enrichment, the present discussion is limited to gaseous diffusion plants. Gaseous diffusion operating experience has been limited in terms of large throughputs of commercial power reactor recycled material. Although there has been considerable production reactor material returned to the cascade, irradiation exposure of that material has been ten- to twenty-fold less than that for power reactors.

The bulk of the radioactivity will be released as solids, either entrained on adsorbate or equipment removed from service for disposal. Liquid waste will be generated by rinsing (decontamination) of recycled equipment.

LIGHT-WATER REACTORS (LWRs)

A logical source of transuranic waste would appear to be the radwaste generated by LWRs. All studies and evaluations have shown the transuranic content below the 10 nCi/gram limit and substantially below the anticipated limit, which should be an order of magnitude or more higher concentration of transuranic isotopes.

MEDICAL AND COMMERCIAL SOURCES

Transuranic wastes arise also from the use of radioisotopes by the medical profession and well-logging industry. For example, the present model of

pacemakers uses plutonium as the source of energy. Consequently, a source of transuranic waste. Also, well-logging firms use radioisotopes in down-hole measurements to provide information on the underground strata and to assess a well's capability for secondary and tertiary recovery. Typically, an americium-beryllium neutron source of 5 to 20 curies and a Cs-137 gamma-ray source of several curies are used.

3.0 RADIOACTIVE WASTES FROM GOVERNMENT FACILITIES

Wastes resulting from Department of Energy (DOE) facilities fall into four categories: currently produced waste, retrievably stored waste, buried waste, and decontaminated and decommissioned wastes. The currently produced and the retrievably stored wastes are, by far, the best characterized and documented. Much less is known about buried radioactive wastes; although there are estimates of volumes involved, they may be off by as much as a factor of two. Decontaminated and decommissioned wastes are the most poorly characterized. If the 740 DOE facilities which are currently listed as surplus, or are likely to become surplus in the next few years, were decommissioned, the volume of radioactive waste generated might range from 1.4×10^5 to 2.7×10^6 cubic meters.

KINDS OF DEFENSE-RELATED WASTES

Plutonium

A large portion of all transuranic waste is generated in various processes associated with the fabrication of nuclear weapons. The weight fraction is dominated by Pu-239. Pu-241 dominates the curie content which determines if waste qualifies as transuranic waste. Finally, 60 kev gamma rays from Am-241 account for a large portion of the surface dose. The weight fraction of Pu-239 will not vary more than a few percentage points from sample to sample. However, the Pu-241 and Am-241 fractions are not known as well and vary significantly with time.

Heat source plutonium is used to fabricate radioisotope thermoelectric generators. Unlike weapon plutonium, the Pu-238 dominates both the weight fraction and the curie content.

Curium, Thorium and Other Transuranics

Savannah River Laboratories has conducted several campaigns to create Cm-244 for use as heat sources in radioisotope thermoelectric generators. No isotopic breakdown is available. Unlike plutonium discussed above, curium wastes tend to contain a considerable amount of fission products and are, therefore, placed in Savannah River concrete boxes which provide considerable shielding.

Bettis has a program to develop a breeder reactor based on the Thorium-Uranium-233 fuel cycle. Although U-233 is not a transuranic, it is included in the definition of transuranic waste. No isotopic breakdown is available.

Some wastes are difficult to categorize. Much of this miscellaneous waste is probably associated with the creation of specific transuranic isotopes such as Cf-252. Also, there are activities which can alter the isotopic content of one of the above processes. For instance, the process at Rocky Flats that creates first stage sludge greatly increases the Am-241 content of the waste. Over 10 kg of the Am-241 at Idaho is in the first stage sludge generated by Rocky Flats.

CHARACTERISTICS OF DEFENSE-RELATED WASTES

The form of the waste is of considerable importance in assessing operational and long-term risks. The combustibility and dispersibility of the waste are important factors in operational accidents. The tendency of waste to produce gas and its susceptibility to leaching are important factors in long-term risk assessments.

Many of the DOE transuranic waste generators have developed fairly detailed descriptive codes to describe the various waste forms in their containers. For example, in order of contribution, the top ten contributors to the volume and the

plutonium content for waste stored at Idaho between September 1, 1971, and December 31, 1976 are as follows:

- Metal scrap
- Paper and rags - dry
- Paper and rags - wet
- Filters - CWS
- Second stage sludge
- LSA metal, glass, etc.
- First stage sludge
- Organic setups (solidified oils)
- Unknown
- Concrete, asphalt, etc.

These ten waste forms account for about 80 percent of the totals. In developing this ranking, no effort was made to combine waste forms with similar descriptions.

4.0 HANDLING AND TREATMENT OF TRANSURANIC WASTES

A qualitative distinction between contact-handled and remotely handled transuranic waste is made by the Department of Energy. Contact-handled waste emits so little gamma or beta radiation that workers can handle it without extensive shielding; remotely handled waste requires shielding and/or remote handling to protect operating personnel. Therefore, contact-handled transuranic waste is distinguished from remotely handled transuranic waste on the basis of the surface-dose rate. A criterion endorsed by the Department of Energy designates waste packages with surface-dose rates no higher than 200 millirem per hour (mrem/hr) as contact-handled transuranic waste and those with surface-dose rates higher than 200 mrem/hr as remotely handled transuranic waste.

Transuranic wastes can be treated and packaged to accomplish several purposes:

- To reduce the volume of waste required for storage and disposal
- To increase the margin of safety associated with handling and storing of wastes
- To recover transuranic elements and
- To reduce the mobility of the wastes during prolonged storage and final geologic disposal.

In the following paragraphs, waste treatment alternatives are outlined according to types of waste independently of waste content.

The quantities and characteristics of these wastes vary appreciably from facility to facility, even within comparable sections of the fuel cycle, depending largely on the maintenance and operational philosophy used at each site. Each category of waste can be processed according to a selected treatment to produce a product which can be disposed.

TREATMENT OF GENERAL TRASH

General trash consists of many materials, both combustible and noncombustible, discarded from a facility. Combustible solids consist of a large variety of items such as paper, rags, plastic sheeting, protective clothing, gloves, rubber shoes, wood, filter cartridges, etc., as well as some partially combustible items such as HEPA (high-efficiency particulate air) filters encased in wooden frames.

General trash can be treated for several purposes:

- To segregate combustibles from noncombustibles
- To reduce the chemical reactivity of the wastes
- To reduce the volume
- To decontaminate it by removing radioactivity

TREATMENT OF DISCARDED EQUIPMENT

Processing equipment used in radioactive service fails or becomes obsolete during the operating life of the nuclear facility and becomes a waste requiring disposal. Discarded equipment normally is flushed and decontaminated prior to its removal from service. Fuel cycle noncombustible waste also includes large components that require size reduction before removal from the facility. This waste may be highly radioactive, requiring personnel shielding equivalent to several feet of concrete. The treatment options for noncombustible wastes

include size reduction, compaction, melting-casting, dissolution and decontamination.

TREATMENT OF WET WASTES

Wet transuranic wastes consist mainly of aqueous solutions and slurries, evaporator concentrates, spent demineralizer resins, filters and filter sludges, and organic oil and solvents. Treatment options for radioactive liquids and other wet wastes include evaporation, drying and calcination, filtration, ion exchanges, membrane processes.

Evaporative processes are generally used on aqueous wastes with high total solids contents. Wastes with low total solids are generally treated by nonevaporative processes to remove contaminants from liquids in order to recycle or discharge the liquid as an effluent. Organic wastes are sent to a combustion process similar to incinerators previously described. The treatments for aqueous solutions and slurries with high total solids contents (both dissolved and undissolved) include evaporators, drying, and calcination.

Evaporation

Evaporation is the most widely used technique for reducing the volume of liquid wastes in fuel cycle facilities. Unlike filters and ion exchangers, which remove small amounts of impurities from large volumes of water, evaporators remove the water from the impurities. Evaporators remove soluble and insoluble materials (but not volatile materials), reduce volume, remove water from a solution, and distill relatively pure water from impure water. Through evaporation, a solution or slurry is concentrated by vaporizing or boiling away the solvent, normally water or mineral acid. Evaporators, coupled with efficient deentrainment devices, provide the capability for a high degree of separation for most radioactive materials. However, the inherently high operating cost of evaporation limits the applications to those liquids which have a high concentration of dissolved solids and require high decontamination factors.

Drying and Calcination

Drying involves removal of liquid through application of heat to leave a dry solid. This process has had little application thus far in the disposal of liquid wastes, but increased disposal costs are expected to encourage use of this technology in nuclear facilities. The dried product from treatment of aqueous solutions/slurries with high solids content is generally a dry or nearly dry salt cake or powder. In many situations, this material is considered suitable for storage but not disposal. The material would be immobilized further by one of the techniques for cement, bitumen, glass, etc.

Calcination, in contrast to drying, is a high-temperature process in which liquid wastes are dried and thermally decomposed to form stable, nonfused compounds such as oxides. Most of the calcination process development work with radioactive wastes has been directed toward treatment of high-level wastes, but the technology is also applicable to low- and intermediate-level wastes.

Filtration

Filtration is the most commonly used method for the removal of precipitates and particulate matter from liquid waste streams. Other methods used to separate solids from liquids include liquid cyclones (hydroclones), and centrifugation, settling and decantation. Centrifuges are mechanically driven devices of variable design; solids-bearing fluids are fed into a rotating section which imparts rotational motion to the suspension. Centrifuges have been used to concentrate and dewater waste slurries (including resins) at reactors.

Ion Exchange

The relatively common application of ion-exchange processes has proven to be effective for decontamination of low-level waste solutions that are relatively low in ionic contaminants. In these processes, the solid ion exchange treatment material preferentially bonds with the radioactive ions to be removed rather than with the similar but less hazardous ions present in the original ion-exchange

material. The loaded ion-exchange material can be either treated by solid waste treatment methods or chemically regenerated for further use, producing a more concentrated liquid regeneration waste for further treatment.

Membrane Processes

Reverse osmosis, ultrafiltration and electrodialysis are membrane processes which could potentially be used for purification of radwaste streams. Basically, these processes result in the separation of soluble constituents from solute-free liquids by selective passage through a membrane; this separation is induced by feed solution pressure for reverse osmosis and ultrafiltration and by passage of an electric current through the feed solution for electrodialysis.

IMMOBILIZATION AND WASTE SOLIDIFICATION TECHNOLOGIES

Immobilization describes a treatment designed to reduce the mobility of wastes by fixation of the "as generated" or final treated form of the low-level wastes. This is typically accomplished by incorporating the wastes into a monolithic solid of low leachability.

Immobilization technologies can be divided into those that are commercially available, and those under development to provide improved product integrity. The major immobilizing agents in use include urea-formaldehyde resins, hydraulic (portland) cement and cement plus additives, bitumen (asphalt), and modified vinyl ester resins.

Two other processes used to immobilize waste liquids involve the use of absorbents and evaporation to a salt cake. While the latter process is not used commercially, it is being used at Hanford to immobilize sodium salt solutions accumulated during DOE's plutonium-production operations. Atomic International also proposes to use the salt media of its molten-salt combustion process to encapsulate the ash product. Other immobilization technologies under development include clay fixation, glass formation, and pelletization.

MULTI-PROCESS TREATMENTS

There are several possible processes that combine volume reduction with solidification. The most widely applied commercial technique is bituminizing of radioactive wastes, which has been developed and used in Europe for several years and is now being marketed in the United States. Basically, bitumen and evaporator concentrates are mixed in either an extruder-evaporator or a thin film evaporator. By heating the extruder or evaporator, residual water of the concentrate is evaporated so that a bitumen-salt mixture results. This mixture is then discharged into containers for offsite disposal. Other solidification agents such as plastics can be used.

An alternative multi-process technology currently under development is the inert carrier radwaste process. This process evaporates all of the water (or other low-temperature volatiles) in the waste by mixing a small stream of cold waste into a large stream of hot inert fluid under highly turbulent flow. The water flash evaporates, is recovered as a condensate, and returned to the system or further processed for discharge. The dried residue (carried by the inert carrier) is coated in suspension with a small quantity of high-melting epoxy resin and separated as a stable solid for packaging and offsite disposal.