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IDENTIFICATION AND SUMMARY CHARACTERIZATION OF MATERIALS POTENTIALLY REQUIRING VITRIFICATION: BACKGROUND INFORMATION

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PREFACE

What follows constitutes background information for the Workshop in general and the presentation entitled *Identification and Summary Characterization of Materials Potentially Requiring Vitrification* that was given during the first morning of the workshop. Summary characteristics of 9 categories of U.S. materials having some potential (interpreted liberally) to be vitrified are given in Table 1. This is followed by 1-2 page elaborations for each of the 9 categories. References to even more detailed information are included.

TABLE 1. SUMMARY OF U.S. MATERIALS HAVING SOME POTENTIAL TO BE VITRIFIED

Material Type	Volume, m^3 or m^3/yr[a]	Radioactivity Density, Ci/m^3	Power Density, W/m^3	Material Description	Vitrification Possibilities
1. Spent civilian nuclear fuel	12000	10,000,000	50000	Light-water reactor spent fuel	Unlikely unless required by repository
2. DOE spent fuel	1200	Not quantifiable; Moderate-to-high	Not quantifiable; Moderate-to-high	Variety of spent fuels	Likely for Al-clad fuels, possible for others
3. DOE "tank" wastes	375000	1,000 – 10,000	5 – 50	Alkaline liquid, saltcake, sludge; calcine	Highly likely for essentially all retrieved tank waste
4. Capsules: Cs	3.5	23,000,000	115,000	Capsules of CsCl	Likely if overpack is unacceptable
Sr	1.1	21,000,000	140,000	Capsules of SrF_2	
5. Transuranic wastes				Wide variety of materials with TRU >100 nCi/g	Likely for only a small fraction unless WIPP-WAC change substantially
Remotely handled	2,500 + 14/yr	1,000	1 – 2		
Contact handled	70,000 + 1500/yr	25 – 50	0.5 – 1.5		
6. Low-level radioactive waste				Extremely wide variety of materials with <100 nCi/g	Likely for LLW from tank waste processing. Unlikely for most other LLW.
DOE	38,000/yr	9 – 27	0.01 – 0.05		
Commercial: Class A		0.6	0.03 – 0.1		
Commercial: Class B	24,000/yr[b]	60	15		
Commercial: Class C		0.1 – 7,000	0.003 – 115		
Commercial: > Class C	63 + 20/yr	>0.1 – high	>0.003 – high		
7. Low-level mixed waste		Not quantifiable; low	Not quantifiable; low	Extremely wide variety of materials with <100 nCi/g and hazardous chemicals	Likely in selected applications, but extent is unpredictable
Commercial	2,100				
DOE	138,000				
8. Surplus plutonium	2	11,000,000	44,000	Plutonium in a variety of forms, mostly metal	Either vitrification or irradiation will be used
9. Environmental restoration	78,000,000	Not quantifiable; low with small-volume exceptions	Not quantifiable Low with small-volume exceptions	Extremely wide variety of materials and contamination	High-toxicity wastes and some in-situ are likely. Unlikely for the bulk of the waste.

[a] Fixed values are existing volumes which are given where production has essentially ceased or where disposal rates are approximately equal to production rates. Rates are given where volumes continue to increase significantly.
[b] Sum of annual production rates for Classes A, B, and C.

1. LIGHT-WATER-REACTOR SPENT FUEL

GENESIS:

Uranium dioxide fuel that have been irradiated for 3-5 years in the approximately 100 civilian pressurized-water reactors (PWRs; 2/3 of capacity) and boiling water reactors (BWRs; 1/3 of capacity) to produce electric power.

DESCRIPTION:

- Basic component is a *fuel rod* or *fuel element*, which is a stack of right-circular cylindrical uranium dioxide fuel pellets in a welded Zircaloy tube. Zircaloy is a metal alloy composed primarily of zirconium with small amounts of tin and iron.

- The rods are held in a square array with a metal lattice *grid spacer* typical composed of Zircaloy but with some made of nickel alloys.
- The array of rods is held together in the axial direction with *tie rods* (typically made of Zircaloy) attached to metal *end pieces* (typically made of stainless steel) to constitute a *fuel assembly*.
- BWR fuel assemblies are enclosed by a solid sheet of Zircaloy called a *fuel channel* along the length of the fuel assembly.

Attribute			PWR	BWR
Diameter/width, cm		Fuel pellet	0.82	1.06
		Fuel rod	0.95	1.25
		Assembly	21.4	13.9
Fuel rods per assembly		Array	17 x 17	8 x 8
		Number	264	63
Height, m		Fuel stack	3.66	3.76
		Rod	3.85	4.06
		Assembly	4.06	4.47
Assembly weight, kg			658	320
Fuel per assembly, kg		Uranium metal	461	183
		Uranium dioxide	523	208
Metal hardware per assembly, kg			135	112
Assembly volume, m^3			0.186	0.086
Avg. specific power, MW/Mg U			37.5	25.9
Burnup, GWd/Mg U		Historical	33	27.5
		Future	60	46
Composition (Historical burnup – Future burnup)				
	Initial	^{235}U enrichment, %	3.30 - 4.73	2.77 - 3.64
	Final	Uranium, kg/Mg Initial U	955.4 - 922.2	962.5 - 937.1
		Uranium enrichment, % ^{235}U	0.84 - 0.54	0.79 - 0.57
		Plutonium, kg/Mg Initial U	9.47 - 14.38	8.26 - 12.3
		Fissile Pu, % $^{239,241}Pu$	71 - 62	72 - 65
		Other actinides, kg/Mg Initial U	0.71 - 1.8	0.59 - 1.50
		Fission products, kg/Mg Initial U	34.4 - 61.6	28.6 - 49.1
Inventory (Annual Addition – Cumulative), Mg Initial U		1994	1207 - 19,024	675 - 10,788
		2000	1300 - 27,400	600 - 14,900
		2010	1400 - 39,000	700 - 21,400
		2020	700 - 50,200	400 - 26,900

REFERENCES

CROFF 1980a, 1980b, 1982; DOE 1992, 1995a; LUDWIG 1989; RODDY 1986

2. DOE-RESPONSIBILITY SPENT FUELS

GENESIS:

Irradiated fuel produced in a diverse array of DOE-owned facilities or for which DOE has assumed responsibility. Principal sources of these spent fuels are as follows:

- Nuclear weapons production complex such as Hanford N-reactor fuel and unreprocessed production reactor fuel at Savannah River Site.
- Naval nuclear reactors.
- A diverse assortment of research, test, and demonstration reactors.

DESCRIPTION:

- DOE-responsibility spent nuclear fuels have an extremely wide-ranging assortment of shapes, forms, and characteristics. A categorization system for these fuels has been developed along 7 dimensions:
  - Enrichment: High, low, natural, depleted
  - Fuel Type: hydride, oxide, alloy, carbide, etc.
  - Fuel Matrix: Zr, Al, stainless steel, graphite, etc.
  - Cladding: Zircaloy, Al, stainless steel, etc.
  - Actinide Content: Minor actinides, Pu
  - Other Materials Present: graphite, Na, Ca, B, etc.
  - Burnup: High, medium, low

  More detailed characterization of fuels comprising the majority of the inventory is contained in some of the references.
- Understanding the DOE spent nuclear fuel inventory is further complicated by the fact that these materials are stored at a variety of sites and facilities. It is likely that not all of these materials have yet been identified, although what remains to be included is likely to add little to the existing inventory.
  - The largest amount of this material is unreprocessed production reactor fuel stored in basins at Hanford, and contains about 2100 MgU.
  - A substantial amount of Al-clad fuels are stored at Savannah River Site.
  - The Idaho site has a substantial amount of a wide variety of fuel stored ranging from Naval reactor to the core that was destroyed in the Three Mile Island Accident to HTGR fuel from Fort St. Vrain.
- The production of DOE-responsibility spent fuels has largely ceased with the following major exceptions:
  - Naval reactor fuels
  - Research reactor fuels: U.S. and other countries
  - Potential new fuels from resumption of tritium production

REFERENCES: DOE 1992, 1993, 1994b, 1995a

3. DOE "TANK" WASTES

GENESIS:

Initially-acid wastes from the reprocessing of spent fuels or processing of irradiated targets to recover valuable constituents by applying a variety of chemical technologies. Most of this is classified as high-level waste, although some is transuranic waste and some low-level waste. Most of the material was neutralized by adding an excess of sodium hydroxide, resulting in the precipitation of many chemicals. Additionally, some of the tank wastes have been further separated and concentrated. There is now about 380,000 m^3 of radioactive mixed waste stored in 332 tanks at Savannah River Site, Hanford Site, Idaho Chemical Processing Plant, West Valley Demonstration Project, and Oak Ridge National Laboratory.

DESCRIPTION:

Alkaline wastes comprise the largest volume of DOE tank wastes and have roughly similar characteristics. These wastes are composed of one or more of the following constituents:

- Liquid: Supernatant and drainable interstitial liquid in the tanks. Alkaline liquids contain substantial amounts of dissolved chemicals, especially sodium salts such as the hydroxide and nitrate/nitrite, often near or at their respective solubility limit. Acidic liquids typically contain only process chemicals, including much lower sodium concentrations, because they have not been neutralized.
- Salt Cake: A crystalline mixture of chemical salts that were precipitated when neutralized liquids were concentrated to reduce storage volume or potential waste mobility. Composed of the same mix of chemicals that are dissolved in the liquid.
- Sludge: A generally thick, amorphous mixture of relatively insoluble chemicals that precipitated as a result of neutralization. Iron and aluminum compounds are typically important, but sludges are usually heterogeneous and contain a wide variety of cations and anions as well as interstitial salt cake or liquid.
- Slurry: Tank waste comprised of solid particles suspended in a liquid. Most of the solids are alkaline nitrate salts that crystallized when liquid wastes were concentrated, but some solids similar to sludges are also present. Only found in double-shell tanks at Hanford.
- Calcine: A granular, flowable solid (similar to powdered detergent) resulting from heating liquid wastes to the point where all of the water is evaporated but where the more stable oxygen-bearing anions (nitrate, sulfate) are not decomposed to oxides. Only found at ICPP.
- Zeolite: An inorganic ion exchange material that has been used to sorb and precipitate radioactive cesium from liquids at West Valley.
- Precipitate: Radioactive cesium that has been precipitated from liquid waste at the Savannah River Site using potassium tetraphenyl borate.

REFERENCES:

DOE 1994a; GEPHART 1995; KUPFER 1993; LEE 1991; SEARS 1990

	Waste Location and Type							
	Savannah River Site				Hanford Site			
Characteristic	Liquid	Sludge	Salt Cake	Precipitate	Liquid	Sludge	Salt Cake	Slurry
Volume, 10^3 m^3	59.3	14.3	53.1	0.2	25.1	46	93	94.7
Radioactivity, MCi	86.4	400.9	145.0	0.1	19.9	110.3	11.5	62.1
Water, Wt %	71.0	55.0	6.4	88.5	40.2	33.6	10.5	56.2
Density	1.1	1.4	1.9	1.05	1.6	1.7	1.4	1.3

Characteristic	Waste Location and Type							
	West Valley Development Project				Idaho Chemical Processing Plant		Oak Ridge National Laboratory	
	Alkaline Liquid	Sludge	Acidic Liquid[a]	Zeolite	Liquids	Calcines	Liquid	Sludge
Volume, 10^3 m^3	1.39	0.05	0.05	0.06	7.7	3.5	0.98	0.41
Radioactivity, Ci	1.9	11.6	1.8	10.6	4.5	40.4	0.02	0.04
Water, Wt %	60.5		40.0		60 – 77	0	68.5	52.2
Density, g/cm^3					1.1 – 1.3	1.1 – 1.8	1.23	1.35

[a]This waste was recently combined with the neutralized waste at WVDP.

4. CAPSULES OF SEPARATED RADIOCESIUM AND RADIOSTRONTIUM

GENESIS:

During the late 1960s and 1970s the contents of many Hanford tanks were recovered and chemically processed to remove radiocesium and radiostrontium, after which the wastes were returned to the tanks. This was done to reduce the heat and radioactivity generated by the wastes in the tank, thus allowing its volume to be further reduced. The radiocesium and radiostrontium in the separate streams were processed into solids and encapsulated.

DESCRIPTION:

Description of Hanford Radioisotope Capsules		
Characteristic	Radiocesium Capsules	Radiostrontium Capsules
Number of Capsules[a]	1328	605
Capsule Construction	Double-encapsulated cylinders (SS 316L/SS 316L) with welded lids	Double-encapsulated cylinders (Hastelloy C-276/SS 316L) with welded lids
Capsule Dimensions Length, cm Diameter, cm	 53 6.67	 51 6.67
Capsule contents	Melt-cast CsCl 38,500 Ci (average)[b] 260 W (average)[b]	Compacted SrF_2 powder 40,100 Ci (average)[b] 193 W (average)[b]
Inventory Volume, m^3 Radioactivity, MCi	 2.4 55.5[c]	 1.1 23.0

[a]An additional 249 radiocesium capsules and 35 radiostrontium capsules have been dismantled. The contents are not expected to be returned to Hanford.
[b]As of January 1, 1995.
[c]Includes ~200 Ci of ^{135}Cs which has a half-life of 3 million years.

REFERENCES: DOE 1991, 1995a, 1996b; ERDA 1977

5. TRANSURANIC WASTES

GENESIS:

- Transuranic (TRU) wastes are materials (a) contaminated with alpha-emitting radionuclides that have an atomic number greater than 92 and half-lives greater than 20 years such that the total concentration of these radionuclides exceeds 100 nCi/g of waste at the time of assay. Before 1984 TRU wastes were defined as those containing 10 nCi/g of such radionuclides, and some TRU waste in storage has TRU radionuclide concentrations in the 10 – 100 nCi/g range.
- Wastes contaminated with other alpha-emitting radionuclides (e.g., ^{233}U, ^{244}Cm) or radionuclides that eventually decay to other alpha-emitting radionuclides (e.g., ^{241}Pu) may be managed as if they were TRU waste according to DOE orders this is not codified in law.
- TRU wastes are produced as secondary wastes during the processing (e.g., separation, fabrication) of materials (e.g., spent fuel, targets, recovered plutonium). Such wastes are produced only by the Department of Energy. Similar wastes produced by commercial operations are considered to be Greater-Than-Class-C low-level waste.

DESCRIPTION:

- TRU wastes exist as a wide range of materials that have been contaminated with sufficient amounts of TRU radionuclides as described above:
  - Assorted solid trash such as protective clothing, paper, rags, glass, tools, and equipment that have been stored awaiting further processing and/or disposal.
  - Liquids, sludges, and a variety of chemical compounds that are being stored awaiting further processing and disposal
  - Waste (> 10 nCi/g) that was managed by burial in near-surface trenches before 1970
  - Soil contaminated by leaking TRU waste containers or the use of soil columns as an ion exchange medium to retard radionuclides released in dilute liquid waste streams
- TRU wastes are further subclassified as "contact handled" or "remote handled", depending on whether the dose rate at the surface of the waste package is less or greater than 200 mrem/hr. Remotely-handled TRU (RH-TRU) wastes constitute about 3% of the total volume and 25% (0.2%) of the total (TRU) radioactivity. The higher radiation levels of RH-TRU wastes results from the presence of fission products, primarily ^{137}Cs.
- TRU wastes are also further subclassified as to whether they are "mixed" wastes by virtue of containing chemically hazardous constituents regulated under the Resource Conservation and Recovery Act (primarily), but also the Toxic Substances Control Act or various state regulations. About 55% of TRU wastes are mixed wastes.

REFERENCES: DOE 1991, 1994a, 1995a

6. LOW-LEVEL RADIOACTIVE WASTES

GENESIS:

- Low-level waste (LLW) is defined by exclusion: it is waste that is not spent fuel, high-level waste, transuranic waste, or byproduct material such as uranium and thorium mill tailings. As such, it must contain less than 100 nCi/g of TRU radionuclides and limits also exist on medium-to-long-lived fission and activation products as well as non-TRU actinides. LLW containing hazardous chemicals is considered separately in Sect. 7.
- Commercial LLW is governed by NRC regulations. It is managed in three classes (A, B, C) with increasing radionuclide concentrations and increasingly stringent disposal requirements. Commercial LLW having radionuclide concentrations greater than Class C is also produced. These wastes are produced by utilities generating electricity using nuclear power plants, commercial firms using radioactive materials to manufacture various items and substances, hospitals that use radionuclides for diagnosis and treatment, and research institutions that use radionuclides in R&D.
- DOE LLW is governed by DOE orders. Sub-classes of DOE LLW are defined on a site-by-site basis, as are waste acceptance criteria which may vary widely. These wastes result from a wide range of DOE activities related to production of nuclear weapons and R&D.

DESCRIPTION:

- Commercial LLW is composed of a collage of waste types as diverse as their sources:
  - Irradiated components, contaminated materials, and immobilized liquids and sludges from nuclear power plant operations
  - Contaminated trash from nuclear fuel cycle operations (e.g., fuel fabrication)
  - Industrial activities (e.g., radiopharmaceuticals, manufacture of sealed sources)
  - Medical wastes from radiopharmaceuticals administered to humans and radioactive sources used to treat diseases
  - Research activities, primarily tracers used in biological research but also in geological research
- In part, DOE LLW is composed of many of the same waste types as commercial LLW because it undertakes many similar activities. In addition, a large amount of DOE LLW has been produced by the processing of materials related to the production of nuclear weapons which has no parallels in the commercial sector. This not only includes general process wastes, but unusual waste forms such as grouted LLW resulting from the processing of high-level waste at Savannah River Site and grouted waste that was injected into the earth at Oak Ridge National Laboratory.
- The preponderance of commercial and DOE LLW is emplaced in near-surface disposal facilities relatively soon after it is generated. Thus, the amount of LLW in storage is small compared to what is already emplaced.
  - One exception to this is LLW that has radionuclide concentrations greater than Class C. By law, disposal of this waste is the responsibility of the Federal government (i.e., DOE). Its disposal destination and attendant waste acceptance criteria are yet to be determined.

REFERENCES: DOE 1995a; LOGHRY 1995

7. LOW-LEVEL MIXED WASTES

GENESIS:

- Mixed low-level waste (MLLW) contains both radionuclides and hazardous chemicals
- Low level radioactive waste is defined in Sect. 6
- Hazardous chemicals are those defined in the Resource Conservation and Recovery Act (RCRA), although chemicals defined by other acts (e.g., Toxic Substances Control Act, state regulations) are included in this category
- Although not specifically denominated as such, many wastes in earlier sections are actually mixed wastes. In particular, tank wastes and many transuranic wastes contain hazardous chemicals that result in their being considered to be mixed

DESCRIPTION:

- Commercial MLLW is composed of a variety of materials from diverse operations and institutional sources.
  - Annual production in 1990 was about 3500 m^3, of which the largest portion was liquid scintillation fluids.
  - Other materials comprising commercial MLLW include waste oils, chlorinated organic chemicals, chlorofluorocarbons, contaminated heavy metals (e.g., lead, mercury), and corrosive aqueous liquids.
  - A large portion of commercial MLLW (especially the organic chemicals) are treated soon after being generated.
  - A total of about 2,100 m^3 of commercial MLLW was in storage in 1990. Contaminated heavy metals constituted the largest volume with contaminated organic chemicals closely following. It is estimated that about 75% of this is waste being accumulated prior to treatment.
  - The most important generators (in decreasing order of importance) are industrial, academic, government, medical, and civilian nuclear power.
  - Details are included in tables that follow.
- DOE MLLW is composed of an extremely wide variety of materials from diverse operations and legacies
  - The inventory of DOE MLLW is about 140,000 m^3 of which 68% is contaminated inorganic solids and contaminated soils and gravels. Mostly-inorganic contaminated debris accounts for most of the remainder.
  - Projected generation of DOE MLLW for the next 5 years is estimated to be about 31,000 m^3 (ignoring final waste forms) which is composed of mostly contaminated inorganic solids although liquids are more significant than in the legacy material.
  - The vast majority of these wastes are being stored at DOE sites and the rate of treatment and disposal is far less than the generation rate

REFERENCES: DOE 1995a,b; KLEIN 1992

8. SURPLUS PLUTONIUM

GENESIS:

High-quality plutonium was produced and separated for military purposes for decades. Recent agreements to substantially reduce the size of nuclear arsenals will lead to some of the existing plutonium stockpile no longer being needed for national security purposes.

In the U.S. the primary permanent disposition alternatives being considered are to:

(a) Convert the Pu to the oxide, fabricate it into spent fuel, irradiate it in light-water reactors, and then dispose of it in a repository as spent fuel, or
(b) Incorporate the Pu directly into a waste form for subsequent disposal in a repository.

The other country with significant amounts of surplus military Pu is Russia. The Russians view the Pu as a valuable fuel resource and plan on using it as such. If the Russians were to sell the Pu to the U.S., its permanent disposition would presumably be the same as stated above. It should be noted that the Russians do not sharply distinguish military and civilian plutonium stocks as in the U.S., and most Pu has been and continues to be generated in power reactors.

DESCRIPTION:

The composition of military Pu has been stated to be approximately as follows:

^{239}Pu	93.0%
^{240}Pu	6.0%
^{241}Pu	0.5%

The amounts of Pu that will be declared surplus to national security needs are officially stated as follows:

United States	38 Mg
Russia	100 Mg

The Russians continue to produce military-grade Pu at a rate of about 1.5 Mg/y because of the need for electric power from three production reactors that remain in operation.

About 28 Mg of the U.S. surplus plutonium exists as the metal and the rest in a variety of forms (oxide, unirradiated fuel, irradiated fuel, and other forms).

REFERENCES:

DIAKOV 1995; DOE 1996a; ALBRIGHT 1993

9. DOE ENVIRONMENTAL RESTORATION WASTES

GENESIS: The has hundreds of unused legacy sites and facilities that are contaminated with radionuclides, hazardous chemicals, or combinations thereof. It has undertaken a long-term environmental restoration program to remediate the sites, and to decontaminate and decommission (D&D) the facilities.

DESCRIPTION:

- Environmental restoration wastes are not well characterized because:
  - In-situ legacy contents are often not well characterized concerning the nature of the materials and spread of contamination
  - The processes by which D&D of facilities will be accomplished is not yet known. Thus, the secondary waste streams have not yet been defined.
- Taken as a whole, environmental restoration wastes are projected to be less heavily contaminated and more heterogenous than other waste types.

  The wastes are segregated into two broad categories: contaminated soil (including sediment and sludge) and contaminated debris (metal, concrete, wood, asphalt, brick, plastic, rubble).

- A small fraction of this waste (~170,000 m^3) is residues from processing of highly concentrated uranium ores during WW II, and as a consequence contains very high concentrations of radium.

REFERENCES: DOE 1995a; NAS 1995

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