Radioactive Waste: The Size of the Problem

In 50 years of producing electrical power and weapons from nuclear fuel, the US has accumulated millions of cubic meters and tens of billions of curies of radioactive waste.

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Exposure to radioactivity is not a new phenomenon: Jewelry workers painting luminous dials on watches in the 1920s were exposed to radium, a naturally occurring radioactive element, until its dangers were identified; in recent decades, homeowners have worried about radon, a naturally occurring radioactive gas that can adversely affect both their health and the resale value of their homes. Today, we must worry as well about the enormous quantities of anthropogenic radioactive materials that have accumulated as waste in this country and worldwide since the 1940s, largely because of the nuclear weapons programs and nuclear power plants. Some of the anthropogenic material is still valuable—for example, as fuel for nuclear reactors. Vast amounts of it, however, consist of waste. Radioactive waste, by definition, is unneeded material that contains unstable elements that decay by emitting alpha, beta or gamma emissions. This article describes the sources of this waste, the types of waste that are of concern and the amounts estimated to be present in the US.

Sources of the waste

Figure 1 shows schematically the processes that have led to the accumulation of most of the radioactive waste in this country. They begin with the mining of uranium. The ore is taken to a mill, where a small amount of uranium is extracted from tons of ore, leaving behind mill tailings about equal in weight to the mined ore. The natural uranium metal consists of a mixture of isotopes; about 0.7% of it is uranium-235, which fissions, and over 99% of it is ^{238}U, which does not. After milling, the metal is enriched to different levels of ^{235}U, depending on its intended purpose, with the slightly enriched uranium going to utility power reactors (see the branch labeled "commercial" in figure 1) and the more highly enriched uranium destined for defense-related purposes (see the "government" branch in figure 1). Left at the enrichment plant are piles of depleted uranium, which is stored for other uses.

The enriched uranium is fabricated into fuel of different types, depending on its enrichment level and intended use. In the commercial branch, the fuel is burned in a nuclear reactor and, after its removal from the reactor, is stored. Currently, all utilities store their spent fuel on-site.

In the government branch, enriched uranium is used to fabricate fuel for reactors that power the US Navy's nuclear submarines. Until 1988, enriched uranium also was sent to the military reactors that produced plutonium for weapons (no such production reactors operate today). Unlike in the commercial fuel cycle, the fuel from the government-run reactors was reprocessed—that is, it went through a chemical process that separated the uranium and plutonium from the fission products in the spent fuel. The uranium was sent back to the fuel fabrication plant, the plutonium to the weapons plant and the remaining liquids and solids stayed behind as high-level waste. The reprocessing plant also generated material contaminated by the transuranic elements (those elements beyond uranium in the periodic table). Such waste is called transuranic, or TRU, waste.

Categories of waste

The US categories of wastes are high-level, transuranic and low-level waste. Mill tailings may be added as a fourth category. Figure 1 shows that high-level waste (HLW), including spent nuclear fuel, is generated by nuclear reactors and reprocessing plants. Low-level waste (LLW), or waste contaminated with comparatively small quantities of radioactive materials, is generated in most of the processes shown in figure 1, as well as in medical and industrial activities. The TRU waste stems from both reprocessing plants and from weapons plants.

We can characterize the quantities of radioactive waste in two ways. One is to determine the volume or mass of the waste. HLW, for example, is usually measured in cubic meters. Spent fuel rods, however, are often characterized by the mass of actinides (largely uranium) that they contain, measured as metric tons of heavy metals (MTHM). The second way used to quantify the amount of radioactive waste is to measure its radioactivity, in curies. (See the glossary of terms on page 23.)

Figure 2 gives both the total volume and the decayed radioactivity of nuclear waste of all types[1] as of the end of 1995. (The "decayed radioactivity," plotted on the horizontal axis, is the calculated radioactivity as of the end

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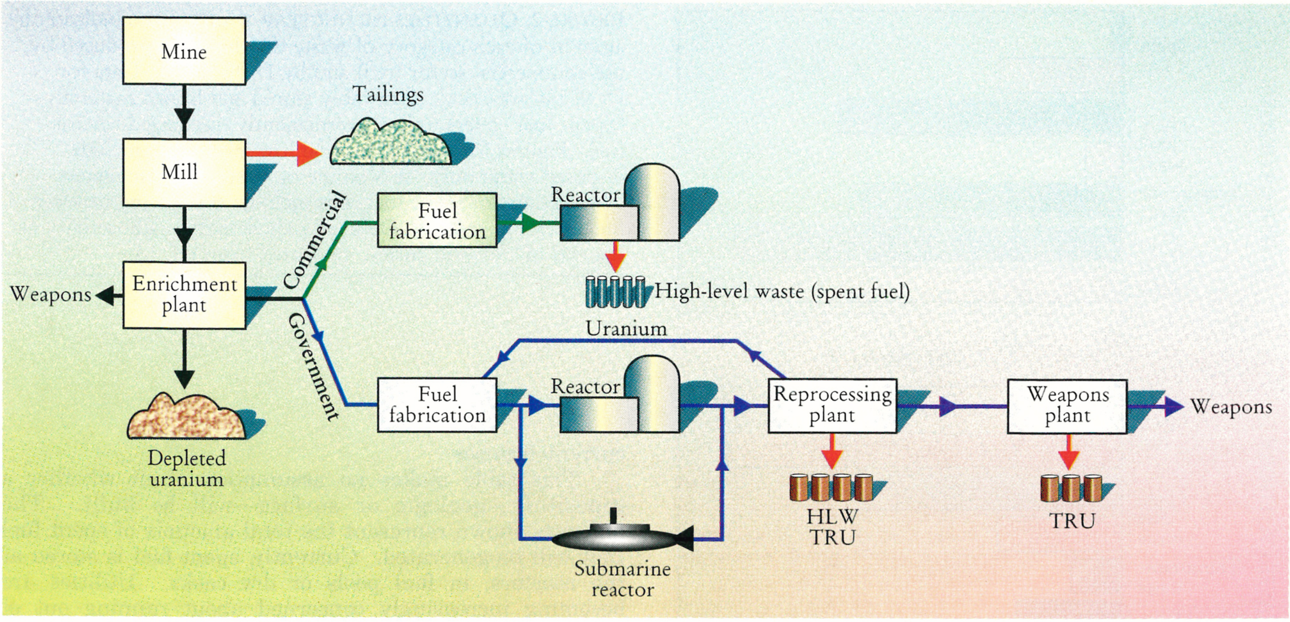

FIGURE 1. PROCESSES THAT GENERATE NUCLEAR WASTE. After uranium is mined, it goes through a series of steps by which it is converted to fuel for a utility-run power plant, naval submarine or plutonium-production reactor. Both the uranium and plutonium are used to make nuclear weapons. The processes shown produce various types of waste (red arrows): spent fuel, other high-level waste (HLW), transuranic (TRU) waste and mill tailings. The processes are conducted by the commercial sector (green branch) and by the Federal government (purple branch).

of the year indicated, so that decay has been taken into account.) The volume is dominated by LLW, which is nearly 90% of the total volume; the radioactivity stems primarily from the spent nuclear fuel. The combined volume of all categories of nuclear waste (excluding what lies in the soil and water) from both the government and commercial cycles is about 5.5 million cubic meters. The radioactivity from all anthropogenic sources is about 31 billion curies.

Whereas the metrics for volume and radioactivity are used internationally, the distinctions among types of waste are not. This article uses the terminology employed in the US regulatory system, which is naturally then the terminology used in discussions of many of the institutional issues.

High-level waste

Formally, the Nuclear Regulatory Commission (NRC), which is responsible for licensing any final disposal of HLW, defines it as follows: "High-level radioactive waste or HLW means (1) irradiated reactor fuel (usually called spent fuel), (2) liquid wastes resulting from the operation of the first cycle solvent extraction system, or equivalent, in a facility for reprocessing irradiated reactor fuel, and (3) solids into which such liquid wastes have been converted."[2]

The Department of Energy (DOE), a major generator of HLW, has a slightly different definition of HLW: "The highly radioactive waste material that results from the reprocessing of spent nuclear fuel, including liquid waste produced directly from reprocessing and any solid waste derived from the liquid that contains a combination of transuranic and fission product nuclides in quantities that require permanent isolation. High-level waste may include other highly radioactive material that the US NRC, consistent with existing law, determines by rule requires permanent isolation."[3]

The difference between the NRC and DOE definitions is whether spent nuclear fuel is characterized as a waste. Many in the nuclear power industry have argued that spent fuel is an energy source, not a waste. They would have the fuel reprocessed, separating out the uranium and plutonium, and letting the remainder, with the other actinides (elements whose atomic number is greater than 88) and all the fission products, be characterized as a waste. Accordingly, although there is a substantial amount of spent fuel at some of the national laboratories operated by DOE, the DOE definition does not include spent fuel as HLW. The DOE definition, as it must, does note that NRC may have a different definition. However, because US policy is not to reprocess commercial spent fuel, but to dispose of the fuel rods directly, NRC does include spent nuclear fuel in its definition of HLW. Neither the NRC nor the DOE definition specifies the amount of radioactivity that must be present to categorize a waste as HLW.

Much of the public debate over radioactive waste concerns high-level waste, including spent fuel, from both commercial and government operations. This waste has more radioactivity associated with it and also has material with much longer decay times than most of that in LLW, the other waste category that the public reads about. The bulk of the volume of HLW in the US was generated

Table 1. Commercial spent nuclear fuel: projected amounts

Year	Mass (MTHM)	Radioactivity (MCi)
1995	32 200	30 200
2000	42 300	32 600
2010	61 800	39 800
2020	77 100	34 700

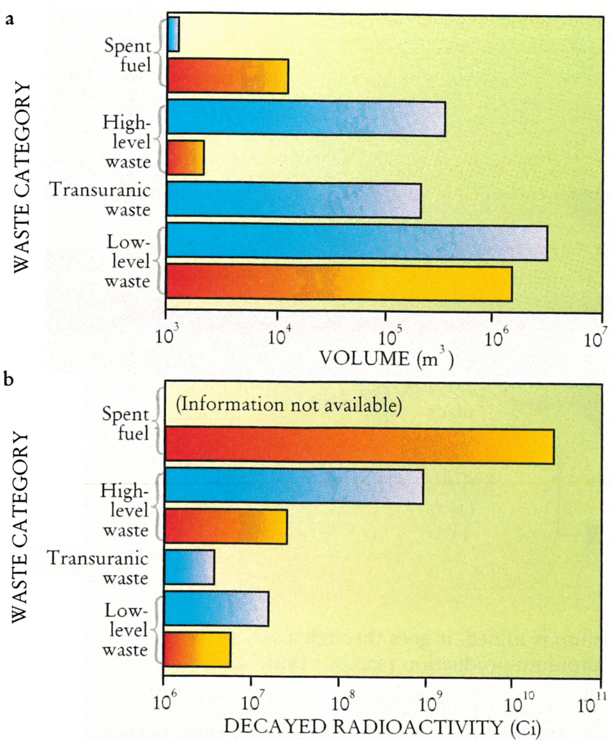

FIGURE 2. QUANTITIES OF NUCLEAR WASTE. Bars indicate the amount of each category of waste that has been produced by the commercial sector (red) and by DOE (blue). Data for DOE includes both retrievably stored and buried materials. "Spent fuel" refers only to permanently discharged reactor fuel. Figures for the commercial low-level waste include disposed waste only. **a:** Volume of waste, in cubic meters. The volume for spent fuel rods includes the spacing between fuel assembly rods. **b:** Decayed radioactivity—radioactivity at the end of 1995—in curies. (Adapted from ref. 1.)

during the production of nuclear weapons. Even though the government is not producing new HLW, large quantities exist from the nearly 50 years of production. Spent nuclear fuel constitutes the other large volume of HLW.

Spent nuclear fuel

Spent nuclear fuel is irradiated fuel discharged from a reactor—that is, the fuel assembly when it is removed from the reactor. Before insertion into a reactor, the fresh fuel, consisting of uranium and (for some fuel) plutonium, is only weakly radioactive. After the fuel has been in an operating reactor, producing energy and neutrons, fissioning has produced highly radioactive fission products as well as actinides.

Table 1 gives the projected amounts of commercial spent fuel, as of the end of 1995, in terms of both mass and radioactivity.[4] The amounts given for 1995 are actual values. Several assumptions have been made to arrive at the projections given for 2000, 2010 and 2020; in particular, it is assumed that no new nuclear plants will be built, no plants will obtain extensions of their operating licenses and all plants will run until the end of their current licenses.

This table makes no assumption about whether a repository—geologic or surface—will be built. The amounts shown represent the total amounts of spent fuel that will be generated. Currently, spent fuel is stored at the reactors, in fuel pools or dry casks. Utilities are becoming increasingly concerned about running out of space to store additional fuel removed from their reactors. DOE is required to take title to the commercial spent fuel in 1998 (this requirement was reinforced by a recent court order). However, DOE has no facility to use to accept such fuel. It has been attempting to develop a permanent repository at Yucca Mountain, Nevada, but the department has been slowed by state challenges and the difficulty of suitably characterizing and analyzing the site. An alternative—an interim storage facility at the Yucca Mountain site—has been proposed by some in Congress but President Clinton has vowed to veto any such legislation.

All elements with atomic numbers from 1 to 65 and from 81 to 96 are found in spent fuel, in many cases in more than one isotopic form. Table 2 lists those elements in commercial spent fuel[4] that contributed the largest amount of radioactivity in 1995. The table includes the total mass of each isotope and its half-life.

In addition to the spent fuel at commercial reactors, there is spent fuel in storage at several of DOE's sites. This spent fuel includes fuel from the former production reactors, naval reactors and foreign and university research reactors. The foreign research reactors are those whose spent fuel has been returned to the US under the agreement by which the original fuel, highly enriched in uranium, was supplied. DOE does not provide data on the radioactivity of its spent fuel, but has revealed that, as of July 1995, there were 2640 MTHM of such fuel,[1] about 8% of the mass of the commercial spent fuel.

Table 2. Isotopes in commercial spent fuel

Isotope	Mass (g)	Radioactivity (10^9 Ci)	Half-life
Cerium-144	8.22×10^5	2.62	285 days
Praseodymium-144	3.47×10^1	2.62	17.3 min.
Plutonium-241	2.42×10^7	2.49	14.4 yrs.
Cesium-137	2.66×10^7	2.31	30.2 yrs.
Ruthenium-106	5.06×10^5	1.69	1.02 yrs.
Strontium-90	1.17×10^7	1.60	28.5 yrs.
Niobium-95	2.86×10^4	1.12	35.0 days

Other high-level waste

Besides spent nuclear fuel, the US accumulation of HLW is almost entirely that generated by DOE and its predecessor agencies in the production of nuclear weapons. Although some such waste may be collected in the decontamination and decommissioning of DOE facilities, there is no expectation that more HLW will be generated. The existing stores of HLW resulted from the reprocessing of spent research and production reactor fuel, irradiated targets and naval propulsion fuel. In reprocessing, plutonium and uranium are recovered, leaving the fission products in a highly radioactive,

FIGURE 3. HIGH-LEVEL WASTE TANK at the Hanford site in Richland, Washington. Workers stand atop one of the 28 buried double-walled tanks, each of which is 75 feet in diameter. The manholes give access to risers coming out of the top of the tank. There are also 149 older, single-walled underground tanks at the Hanford site. (Photo courtesy of Fluor Daniel Hanford.)

acidic liquid.

Several approaches were used to store this liquid. The HLW now is in several forms (liquid, sludge and solid), which are stored primarily in tanks at the Savannah River site near Barnwell, South Carolina, (SR), the Hanford site in Richland, Washington, and the Idaho National Engineering Laboratory (INEL) in Idaho Falls. A small amount is stored at West Valley, New York, where a reprocessing plant once operated.

Table 3 lists the volume and radioactivity of HLW stored at these four locations[1] at the end of 1995. According to DOE, "The department currently stores about 100 million gallons of high-level waste—enough to fill about 10 000 tanker trucks . . . Most of this waste has been stored in 243 underground tanks in Washington, South Carolina, Idaho and New York."[5] (Figure 3 shows access ports to tanks of high-level waste at the Hanford site.)

Transuranic waste

Only the US, or more specifically, DOE, has defined a special waste category for transuranic (TRU) waste, although some other nations have generated this type of waste. NRC does not regulate TRU waste as a separate category. DOE defines it as "Waste containing more than 100 nanocuries of alpha-emitting transuranic isotopes, with half-lives greater than 20 years, per gram of waste, except for (a) high-level waste, (b) waste that DOE has determined, with the concurrence of the Administrator of the Environmental Protection Agency, does not need the degree of isolation required by 40 CFR 191, or (c) waste that the US [NRC] has approved for disposal on a case-by-case basis in accordance with 10 CFR 61."[3]

Other countries, such as France and the UK, have a third category, such as "intermediate-level waste," but do not share a common definition.[6]

Transuranic waste consists of material with atomic numbers greater than 92. It is generated in the reprocessing of fuel used in the production of nuclear weapons material. Although some TRU is in sludge or liquid form, most of it consists of items that have become contaminated, such as protective clothing, rags and equipment. TRU is classified as either contact handled or remote handled, depending on the radiation level at the surface of the package: If the level is greater than 200 millirems per hour, the waste is considered to be remote handled TRU.

Before 1970, TRU was put into shallow landfills at what were then Atomic Energy Commission sites. This material is labeled by DOE as buried TRU and is not mentioned in the normal tables, which list "retrievably stored" TRU. Table 4 lists[1] the volumes and the radioactivity of retrievably stored TRU at the major sites at the end of 1995. (Figure 4 shows TRU waste in retrievable storage at INEL.) In addition to the sites listed, an additional 17 locations store

Table 3. High-level waste

Site	Volume (10^3 m^3)	Radioactivity (MCi)
Hanford	233.5	339.9
Savannah River	126.5	502.2
Idaho National Engineering Laboratory	11.2	49.3
West Valley	2.2	24.1
Total	373.4	915.5

Table 4. Transuranic waste (retrievably stored)

	Contact Handled		Remote Handled	
Site	Volume (m^3)	Radioactivity (10^4 Ci)	Volume (m^3)	Radioactivity (10^4 Ci)
Hanford	1.15×10^4	15.5	273	3.15
Idaho	3.93×10^4	35.0	200	0.72
Los Alamos	1.12×10^4	20.2	94.1	0.05
Oak Ridge	1.33×10^3	5.5	1842	9.65
Savannah River	6.98×10^3	56.4	—	—
West Valley	4.32×10^1	<0.01	529	0.01
Rocky Flats	1.98×10^3	116.	—	—

FIGURE 4. BARRELS OF TRU WASTE being stacked in a building at the Idaho National Engineering Laboratory. DOE is studying the suitability of long-term disposal of such wastes at the Waste Isolation Pilot Plant in New Mexico. (Photo courtesy of DOE.)

smaller amounts.

DOE is studying the suitability of disposing of the TRU wastes at the Waste Isolation Pilot Plant in New Mexico. Plans for this facility and for Yucca Mountain are discussed in more detail in the accompanying articles by Kevin Crowley (page 32) and by William Kastenberg and Luca Gratton (page 41).

Low-level waste

Basically, all radioactive waste that is not HLW or TRU is classified as LLW. More precisely, LLW is defined under the Nuclear Waste Policy Act to be radioactive waste that (a) is not HLW, spent nuclear fuel, TRU or by-product material, and (b) is classified by NRC as LLW.[1] There are some further distinctions. For example, DOE categorizes waste with less than 100 nanocuries per gram of TRU as LLW[1] and NRC breaks such waste into three categories: A, B and C, with A being the least radioactive.[7]

The total volume of LLW is larger than that for either HLW or TRU, but its radioactivity is significantly less, and most of the LLW consists of material with half-lives much shorter than those of most of the isotopes in HLW and TRU waste.

DOE and its predecessors have generated about two-thirds of all the LLW in the US. Much of the DOE waste consists of "large amounts of waste contaminated with small amounts of radionuclides such as contaminated equipment (such as glove boxes, ventilation ducts, shielding, and laboratory equipment), protective clothing, paper, rags, packing material and solidified sludges."[3] The commercial uses of radioactive materials that lead to LLW are operations at nuclear power plants, more than 100 million annual medical procedures, the testing and development of about 80% of all new drugs, sterilization of consumer products and production of commercial products such as smoke detectors.[8]

Most DOE waste is buried at DOE sites. (See figure 5.) Commercial LLW is sent to sites operated by private firms at several locations: a site at Richland, Washington, which is open to only a few states; one at Barnwell, South Carolina, open to all states but North Carolina; and a new site run by Envirocare near Clive, Utah, which has no restrictions on the origin of the waste it can accept.

Table 5 summarizes the amount of LLW that had been disposed of at DOE and commercial sites by the end of 1995, showing the 2:1 ratio in volume between DOE and commercial LLW.[1] Table 6 shows[1] how much LLW was shipped to two of the privately operated facilities—Richland and Barnwell—in 1995. The utility industry was the main volume generator of LLW that year. In table 6, the academic category includes university-related hospitals and medical research facilities. The government category in the table consists of state and non-DOE federal agencies.

The states regulate the disposal of LLW, following the regulations es-

Table 5. Disposed low-level waste

Category	Volume (10^3 m^3)	Radioactivity (10^3 Ci)
DOE	3011	12 550
Commercial*	1538	5376
Envirocare	156	N/A

* excludes Envirocare

Table 6. Low-level waste sent in 1995

	Sent to Barnwell		Sent to Richland	
Source	Volume (m^3)	Radioactivity (Ci)	Volume (m^3)	Radioactivity (Ci)
Academic*	132	39	81	8
Industrial	3200	25 000	1600	13
Government**	3170	74 800	535	186
Medical	19	2	36	4
Utility	7210	69 100	3560	2630

*Includes university-related hospitals and medical research facilities
**States and non-DOE federal agencies

FIGURE 5. LOW-LEVEL WASTE being buried in an engineered trench at the Savannah River site early in 1994. The trench was designed to hold more than 30 000 carbon steel boxes of waste, each measuring 4 by 4 by 6 feet. When this trench was filled in March 1995, the trench burial of waste at the Savannah River Site was stopped. LLW waste there is now being stored in covered structures. (Photo courtesy of DOE.)

tablished by NRC. It is also NRC that defines different types of LLW by the concentration of specific radionuclides and the decay times for these nuclides. All three NRC classes (A, B, C) can be disposed of in underground burial sites, with category C requiring deeper burial. These burial sites are pits about 30 meters deep. Essentially, category A and B waste must be protected for 100 years; category C, for 500 years, according to Federal regulation.[9]

LLW falls under the jurisdiction of NRC if the concentrations of certain radionuclides exceed stated limits. In that case, NRC requires burial of the LLW in a deep repository, similar to the repositories designed for HLW. This class of LLW is known as "greater than class C" wastes. At the end of 1993, DOE estimated that the volume of greater-than-class-C LLW was slightly more than 200 m^3 and that it contained 4.2×10^6 curies, of which almost 92% was waste from nuclear utilities.[4]

Perhaps not as widely discussed as HLW at a national level, LLW has generated intense local interest as attempts to develop new LLW sites have led to drawn-out and heated disputes. Examples include the continuing battle over licensing a LLW site at Ward Valley, California, with Senator Boxer opposing it and Governor Wilson in favor of it; the abandoned attempts to develop sites in Illinois and, independently, in New York State, after an investment of eight years and at least \$55 million in each case; and the continuing effort to develop a site in North Carolina, on which \$90 million has already been spent.

Finally, the DOE waste problem also includes the cleanup of the contaminated sites used to make nuclear weapons, such as Hanford, Savannah River and Rocky Flats (near Denver). Not included in the data presented so far in this article are estimates for the contamination in the ground, soil and water, and in the debris, buildings, and so forth. DOE has only poor estimates of these amounts. But the department expects to have to deal with 73 million cubic meters of contaminated soil and 3.8 million cubic meters of contaminated debris, and it does not even have an estimate for groundwater contamination.[4]

Mill tailings and other wastes

There are other types of radioactive waste, which are mentioned here only for completeness, but are not described in detail. Mill tailings are the residues from milling uranium ore. The volumes are large (120×10^6 m^3),[1] but the radioactivity levels are small. The principal hazard is radon gas, leading to a requirement for the tailings piles to be covered. A separate law governs the method of covering and consolidating these tailings. Finally, radioactive materials occur naturally and include radium in old watches and radioactive material in coal ash. Operation of research accelerators produces some LLW, labeled as accelerator-produced radioactive material. These sources do not represent a major amount of waste and, in many cases, are not regulated.

In sum

Large volumes and significant amounts of radioactivity are represented by the radioactive waste in the US. Almost all is now being stored on the sites where it was generated. In many cases, particularly in the case of the HLW, the storage facilities were not built for long-term use. Progress toward resolving the radioactive waste problems has been painfully slow.

References

1. Department of Energy, *Integrated Data Base Report 1995: US Spent Nuclear Fuel and Radioactive Waste Inventories, Projections and Characteristics*, DOE/RW-0006, Revision 12, (1996).
2. *Disposal of High-Level Radioactive Wastes in Geologic Repositories*, Code of Federal Regulations 60, section 60.2.
3. Department of Energy, *Draft Waste Management Programmatic Environmental Impact Statement (PEIS) For Managing Treatment, Storage, and Disposal of Radioactive and Hazardous Waste*, DOE/EIS-0200-D (1995).
4. Department of Energy, *Integrated Data Base Report 1994: US Spent Nuclear Fuel and Radioactive Waste Inventories, Projections and Characteristics*, DOE/RW-0006, Revision 11, (1995).
5. Department of Energy, Office of Environmental Management, *Closing the Circle on the Splitting of the Atom* (1995).
6. See, for example, Pierre Saverot, J. Nucl. Mater. Management, April 1994, p. 18.
7. Code of Federal Regulations 10, section 61.55.
8. General Accounting Office, *Radioactive Waste: Status of Commercial Low-Level Waste Facilities*, GAO/RCED-95-67 (1995), p. 12.
9. Code of Federal Regulations, section 61.4(b)(4) and (5). ■