VITRIFIED HIGH-LEVEL RADIOACTIVE WASTE

OVERVIEW

High-level radioactive waste (HLW) is the highly radioactive material generated from reprocessing spent nuclear fuel (SNF). Starting in the 1940s, HLW resulted from reprocessing SNF to recover fissile material for use in nuclear weapons production. More recently, HLW has been generated during processing of irradiated materials to recover special radioisotopes for medical and industrial purposes. A small amount of HLW (less than 5% by radioactivity contribution) was generated several decades ago as a by-product of commercial SNF reprocessing.

HLW was generated and is currently stored at four sites: the Hanford Site near Hanford, Washington; the Idaho National Laboratory near Idaho Falls, Idaho; the Savannah River Site near Aiken, South Carolina; and the West Valley Demonstration Project near West Valley, New York. Table 1 lists the sites and the years during which HLW was generated at the respective sites. The site locations are shown on the map in Figure 1.

Table 1. Locations and Years of High-Level Waste Generation in the United States

Location	Years
Hanford Site	1940s–1980s
Idaho National Laboratory	1963–2000
Savannah River Site	1950s–present
West Valley Demonstration Project	1966–1972

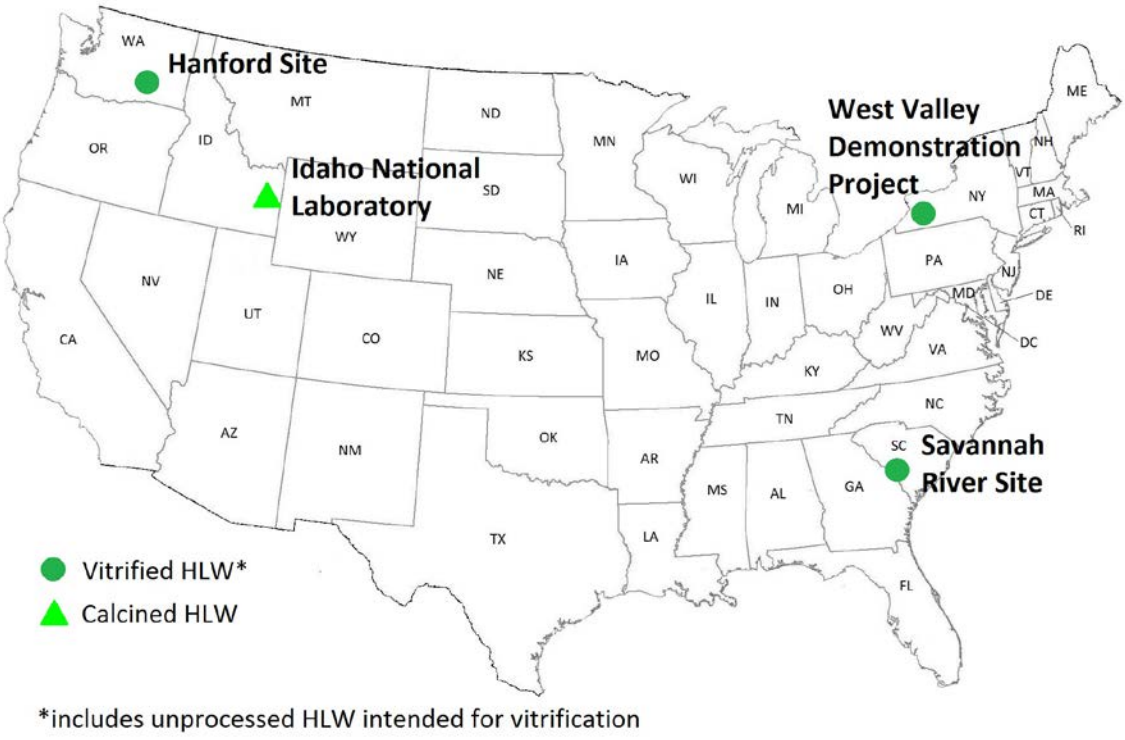

Figure 1. Locations of High-Level Waste Generation in the United States

Note: Circles indicate the sites where high-level wastes that have been or are planned to be vitrified are stored. Calcined high-level waste is stored at the Idaho National Laboratory, indicated by the triangle.

The HLW generated from processing SNF using conventional aqueous technology is initially in liquid form and is stored in underground steel tanks. Vitrification is a process by which HLW is solidified in preparation for disposal in a geologic repository. HLW is vitrified by mixing it with a combination of silica sand and other glass-forming chemicals, heating the mixture to very high temperatures [approximately 1,150°C (2,100°F)] until it melts, and pouring the molten material into stainless steel canisters where it cools to form a glass. Vitrification is used in several countries to immobilize HLW because it has advantages over other modes of treatment. It is a well-demonstrated technology resulting from more than 40 years of industrial experience, it can be used for a wide range of HLW compositions, it is a continuous process that can be applied to large volumes of HLW, and the resulting glass product is chemically durable in many geologic disposal environments. In addition, it exhibits good thermal and mechanical stability properties (NRC, 2011).

The HLW generated at the Idaho National Laboratory was not vitrified; it was calcined, converted from liquid to granular solids by evaporation and thermal decomposition at high temperatures (see fact sheet on Calcined HLW).

STORAGE AND LOCATION

At the West Valley Demonstration Project, vitrification of the complete inventory [2.3 million liters (600,000 gallons)] of HLW, which resulted from processing commercial SNF blended with some defense SNF, was completed in 2002 (Petkus *et al*., 2003). At the Savannah River Site, a vitrification facility initiated radioactive operations in 1996 and over 15 million liters (~4 million gallons) of liquid HLW generated at the site have been vitrified (SNL, 2014). About 140 million liters (37 million gallons) of HLW stored in underground carbon steel tanks at the Savannah River Site remain to be vitrified (SRS, 2015). At the Hanford Site, a vitrification facility is under construction. Approximately 210 million liters (55 million gallons) of HLW that eventually will be vitrified are stored in 177 underground carbon steel tanks (Certa *et al*., 2011). Figure 2 shows the volumes of liquid HLW that have been vitrified or will be vitrified in the future.

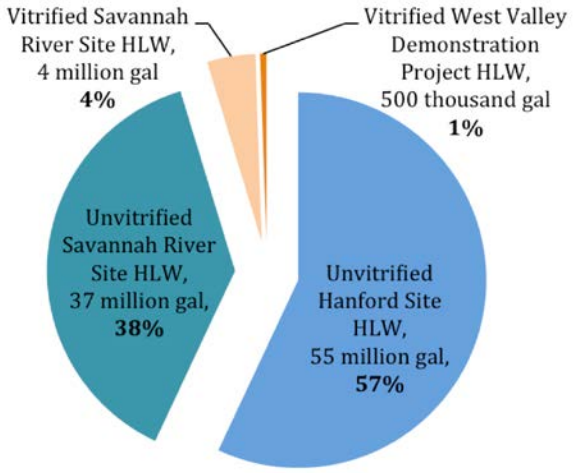

Figure 2. Relative Volumes of High-Level Waste That Have Been or Will Be Vitrified as of 2014
Note: Data from SNL (2014) and SRS (2015)

The vitrified HLW at West Valley is in 275 stainless steel canisters that are in dry storage on-site (SNL, 2014). In the future, these canisters will be moved to dry storage similar to the independent spent fuel storage installations used for commercial SNF until the canisters can be moved to a geologic repository for permanent disposal. At the Savannah River Site, almost 4,000 canisters of vitrified HLW (as of March 2015) are stored in underground reinforced concrete vaults at two Glass Waste Storage Buildings (SRS, 2015). A schematic diagram of a glass storage building is shown in Figure 3. To provide storage for the liquid HLW that remains to be vitrified at the Savannah River Site, the Glass Waste Storage Project is being developed to accommodate additional canisters in above-ground storage containers similar to those used for commercial SNF dry storage. Also, there are plans to double stack the canisters in one of the glass-waste storage buildings, which will effectively double the interim storage capacity of that building.

At the Hanford Site, 34 canisters of radioactive borosilicate glass containing cesium-137 and strontium-90 that were produced in 1986 and 1987 for the Federal Republic of Germany are in dry storage inside casks provided by the German government. The radioactive glass was intended for use as isotopic and radiation sources in Germany's geologic repository testing program. The German testing program was discontinued before the canisters could be shipped to Germany and the canisters remained at the Hanford site.

To store the canisters of vitrified HLW to be produced from the Hanford tank waste, DOE plans to build an Interim Hanford Storage Facility that includes below-grade storage vaults similar to those of the Glass Waste Storage Buildings at the Savannah River Site (DOE, 2014).

Figure 3. Schematic Diagram of a Glass Waste Storage Building at the Savannah River Site (Ridley, 2012)

COMPOSITION

Because most radionuclides are incorporated into the structure of the glass, vitrified HLW is relatively homogeneous. Typical HLW borosilicate glasses are composed of 33 to 65% silica (SiO_2), 3 to 20% boria (B_2O_3), 4 to 22% soda (Na_2O), 3 to 20% alumina (Al_2O_3), and 0 to 50% other metal oxides (SNL, 2014). Figure 4 shows the average composition of borosilicate glass produced at the Savannah River Site. The radionuclide content of HLW includes dozens of different nuclear fission products, activation products, minor actinides, and residual uranium and plutonium not recovered during processing. The relatively short-lived cesium-137 and strontium-90 (with half-lives of about 30 years) are initially the largest contributors to overall radioactivity, whereas plutonium isotopes are the major contributors to long-term radioactivity. Much of the radionuclide inventory is long-lived, so the HLW must remain isolated over long time scales, up to several hundred thousand years (ANDRA, 2005). In contrast, the German borosilicate glass canisters only contain relatively short-lived cesium-137, strontium-90, and their daughter products. After only a few hundred years, the radionuclides will have decayed, resulting in very low levels of radioactivity in the German glass (Stewart and Stewart, 2011).

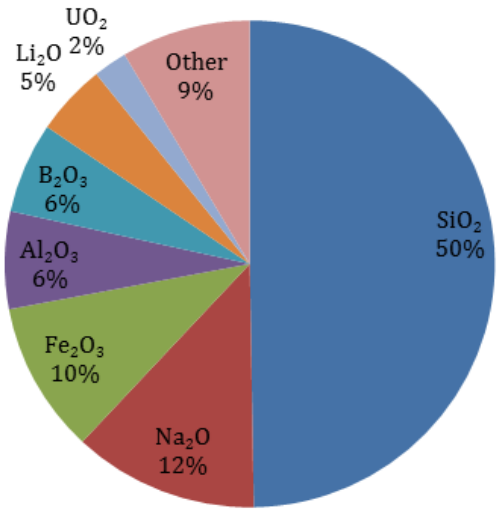

Figure 4. Average Composition of Savannah River Site Vitrified High-Level Waste
Note: Based on Macrobatches 1 to 8 (SNL, 2014)

MASS AND RADIOACTIVITY

There is a total of approximately 489 million curies (Ci) of radioactivity (estimated for 2017) in the almost 7 million kilograms (15 million pounds) of vitrified HLW currently stored at the Hanford Site, Savannah River Site, and West Valley (SNL, 2014), and in the 350 million liters (92 million gallons) of liquid HLW at the Hanford Site and Savannah River Site (Certa *et al.*, 2011; SRS, 2015). Table 2 lists the total radioactivity and the amount of existing and projected vitrified HLW at each site. In 2048, which is the year DOE has set as its target for having a geologic repository for SNF and HLW constructed

Table 2. Estimated Radioactivity, Mass, and Volume of Existing and Projected Vitrified High-Level Waste by Location

Storage Location	Radioactivity*	Existing Vitrified Waste		Projected Total Vitrified Waste	
West Valley Demonstration Project	15 million Ci†	573,802 kg‡ (1,265,000 lb)	245 m^3‡ (8,650 ft^3]	573,802 kg‡ (1,265,000 lb)	245 m^3‡ (8,650 ft^3)
Savannah River Site	332 million Ci#	>5,900,000 kg‡ (>13,000,000 lb)	>2,969 m^3‡ (>104,800 ft^3)	14,000,000 kg§ (31,000,000 lb)	6,957 m^3‡ (245,700 ft^3)
Hanford Site (tank waste)	134 million Ci†	0	0	32,000,000 kg¶ (70,500,000 lb)	14,089 m^3‡ (497,500 ft^3)
Hanford Site (German glass)	8.2 million Ci‡	5,380 kg¤ (11,900 lb)	2.1 m^3¤ (74 ft^3)	5,380 kg¤ (11,900 lb)	2.1 m^3¤ (74 ft^3)

*The estimated radioactivity is for the year 2017.
†Source: Carter *et al.* (2013)
#Source: Carter *et al.* (2013), decay corrected to 2017
‡Source: SNL (2014), decay corrected to 2017
‡Source: SNL (2014)
§Based on an average glass mass of ~1,800 kg (~3,970 lb) per canister reported in SNL (2014; Table A-14) and a projected 7,824 canisters (SNL, 2014, p. 18)
¶Based on an average glass mass of 3,020 kg (6,660 lb) per canister of Hanford HLW reported in SNL (2014, Table A-24) and a projected 10,586 canisters (SNL, 2014, p. A-60)
¤Based on an average glass mass of 158.3 kg (349.0 lb) or volume of 60.6 L (0.0606 m^3) per canister reported in SNL (2014; Table A-20) and a total of 34 canisters

and operating (DOE, 2013), the total radioactivity of vitrified HLW at all three sites is estimated to decay by approximately 50% to 232 million Ci (SNL, 2014).

STABILITY AND RADIONUCLIDE RELEASE IN A GEOLOGIC REPOSITORY

The ability of vitrified HLW to sequester and retain its radioactive constituents is an important factor in evaluating the performance of geologic repositories for HLW. The stainless steel canister containing the glass and any overpack or other engineered barrier system initially can isolate the glass from groundwater. If the canister and engineered barrier system are breached, then the glass waste form can corrode. Three stages of glass degradation are recognized (illustrated in Figure 5) based on the results of glass corrosion studies (Vienna *et al.*, 2013). During Stage I, when groundwater initially contacts the glass, alkali and alkaline earth ions (including cesium and strontium ions) and boron are replaced by hydrogen ions. The corrosion rate during this stage is controlled by the rate of detachment of silicic acid from the glass. In Stage II, as the concentration of silica and other glass components in solution increases, the dissolution rate slows due to greater equilibrium between the glass and solution, as well as the formation of a protective gel layer on the glass surface. The reaction rate during this stage is very low and stable, but glass may enter Stage III, where the reaction rate increases rapidly. The exact cause and timing of Stage III initiation are unknown, but it often

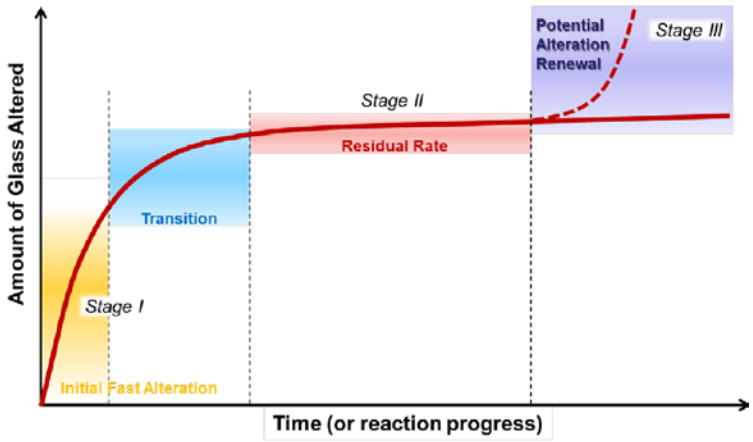

Figure 5. The Three Stages of High-Level Waste Borosilicate Glass Degradation as a Function of Time (Ryan *et al.*, 2014)

occurs during sudden precipitation of silica-containing compounds such as zeolite minerals. If the Stage II degradation rate is maintained, vitrified HLW could take hundreds of thousands of years to completely degrade (ANDRA, 2005). However, because it is unknown how or when Stage III will occur in a geologic repository, assuming a Stage II degradation rate may overestimate the actual glass lifetime.

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The U.S. Nuclear Waste Technical Review Board is an independent federal agency established in the 1987 amendments to the Nuclear Waste Policy Act (NWPA). The Board evaluates the technical and scientific validity of U.S. Department of Energy activities related to implementing the NWPA and provides objective expert advice on nuclear waste issues to Congress and the Secretary of Energy. The eleven Board members are nominated by the National Academy of Sciences and are appointed by the President.