CESIUM AND STRONTIUM CAPSULES

OVERVIEW

The cesium and strontium sealed within double-walled, metal alloy capsules and stored at the Hanford Site were extracted from high-level radioactive waste (HLW) stored in underground tanks at the site in order to reduce the heat load of the HLW. Cesium (mostly in the form of cesium-137, along with minor amounts of the much longer-lived cesium-135) and strontium (in the form of strontium-90) were separated from the HLW and solidified at the Hanford B Plant from 1967 to 1985. These materials were processed and encapsulated at the Hanford Waste Encapsulation and Storage Facility (WESF) from 1974 to 1985, and then placed in underwater storage at the facility. There are 1,335 cesium capsules and 601 strontium capsules stored at the WESF.

STORAGE AND LOCATION

At the WESF, cesium and strontium are stored in double-walled, stainless steel capsules under 13 ft (4.0 m) of water in pools lined with stainless steel. The water aids both in cooling the capsules and in providing shielding to protect plant operators and equipment from radiation. The U.S. Department of Energy (DOE) is considering moving the cesium and strontium capsules to dry storage to both reduce storage costs and increase storage safety (Sedillo, 2014). Wet storage at the WESF has a higher operating cost than dry storage and the WESF, which is more than10 years past its 30-year design life, has experienced some degradation of key structures and systems relied on for safety (Sedillo, 2014). Figure 1 shows one of the WESF water pools in which cesium and strontium capsules are stored.

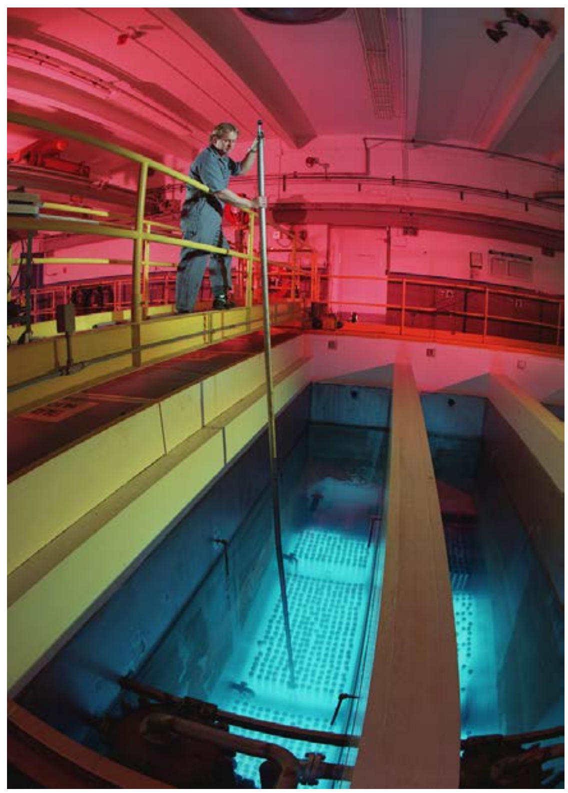

Figure 1. Pool Storage of Cesium and Strontium Capsules at the Waste Encapsulation and Storage Facility at Hanford (Gephart, 2003)

Some cesium and strontium capsules were sent to the Oak Ridge National Laboratory Office of Isotope Sales for dismantling and sale of the cesium and strontium for commercial use. Hundreds of cesium capsules also were leased for commercial use and government research purposes in the 1980s. However, in 1988, after a cesium leak from a leased capsule was detected at Radiation Sterilizers, Inc. in Decatur, Georgia, all the capsules were recalled and returned to the WESF. In 1983, four strontium capsules were permanently disposed of in a shallow, 36-m (120-ft) test borehole at the Nevada Test Site (Cochran *et al*., 2001).

COMPOSITION

Cesium Capsules

The cesium capsules were created by melting cesium chloride at 730 to 750°C (1,350 to 1,380°F) and pouring the molten cesium chloride into stainless steel inner capsules that then were welded shut (DOE, 1990). These capsules subsequently were placed into stainless steel outer capsules that also were welded shut. The inner and outer capsules together are referred to as a "standard capsule." Figure 2 shows a schematic diagram of a standard cesium capsule. The outer capsule has an outside diameter of 2.63 in (6.68 cm) and a total length of 20.78 in (52.78 cm) (SNL, 2014). The cesium chloride content of the capsules ranges from approximately 2.9 to 7.1 lb (1.3 to 3.2 kg) (Tingey *et al.*, 1984).

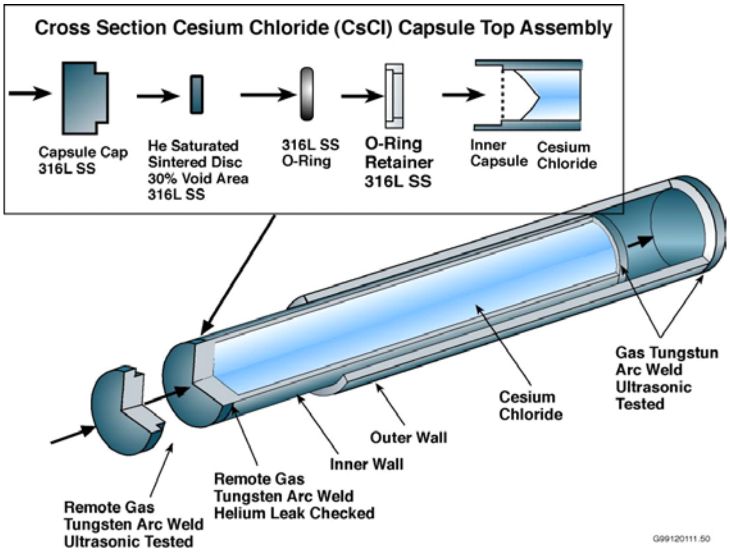

Figure 2. Schematic Diagram of a Standard Cesium Capsule (Plys and Miller, 2003)

The 1,335 cesium capsules stored at the WESF include 1,312 standard capsules and 23 Type W overpack capsules. The Type W overpack capsules, which also are made of stainless steel, have an outside diameter of 3.25 in (8.26 cm) and a total length of 21.83 in (55.45 cm) (SNL, 2014). Most of the Type W overpacks contain cesium capsules that have swollen due to expansion of the cesium chloride salt in phase transition. The phase change is explained further in the Stability and Radionuclide Release section. Due to this swelling, the cesium capsules do not meet the acceptance criteria for the WESF storage pools without an overpack.

The radioisotope content of the cesium capsules is mostly short-lived cesium-137 (with a half-life of 30.2 years), while the rest is long-lived cesium-135 (with a half-life of 2.3 million years). Cesium-137 initially is the main contributor to radioactivity, but after several hundred years, cesium-135 will become the dominant source of radioactivity. Eventually, all of the cesium will decay to barium-137 and barium-135, which are both stable isotopes. The cesium capsules also contain chemical impurities, including alkali and alkaline earth metals, silicon, aluminum, iron, and some heavy metals such as chromium, lead, cadmium, and silver.

Strontium Capsules

In preparation for loading into capsules, strontium nitrate from the Hanford B-Plant was converted into strontium fluoride at the WESF and then dried at temperatures in the range 800 to 950°C (1,470 to 1,740°F) in a drying pan (SNL, 2014). Capsules fabricated from Hastelloy™ (a metal alloy composed mainly of nickel, chromium, and molybdenum) were loaded with dried strontium fluoride that was chipped from the drying pan and compacted. Capsules were welded shut and placed into outer capsules made of stainless steel or Hastelloy™, which also were welded shut. The strontium capsules are approximately the same size, shape, and mass as the cesium capsules. Strontium-90 has a half-life of 28.8 years and decays to yttrium-90, which has a half-life of 64 hours. Eventually, all of the yttrium-90 will decay to stable zirconium-90.

Of the 601 strontium capsules stored at the WESF, 411 were filled using the method described in the preceding paragraph. These capsules are referred to as "standard capsules." An additional 189, referred to as "waste capsules," were filled with strontium fluoride that had fallen onto surfaces such as floors and tables during the strontium chipping process. One strontium capsule, called the tracer capsule, is filled with non-radioactive strontium.

The strontium "standard capsules" contain chemical impurities including barium, lead, cadmium, chromium, and silver. The strontium "waste capsules" also could contain inorganic materials from the drying room surfaces and some of these capsules contain pieces of equipment, building materials, metals and metal fluorides, and other chemicals that were inadvertently mixed with the waste while collecting strontium fluoride from facility surfaces. Some strontium waste capsules may contain as much as 50% foreign materials (SNL, 2014).

Figure 3 shows the distribution of the different types of cesium and strontium capsules in inventory.

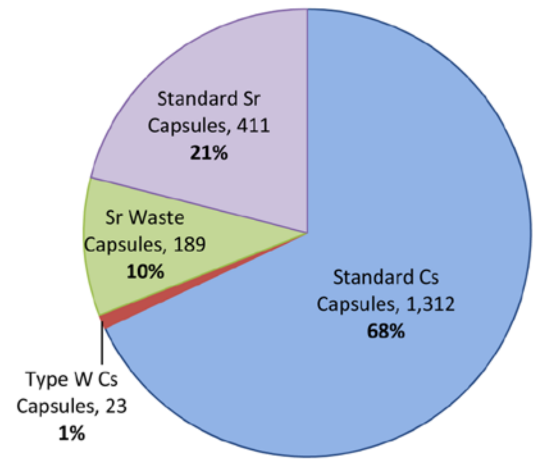

Figure 3. Distribution of Radioactive Cesium and Strontium Capsule Types Stored at Hanford
Note: Data from SNL (2014). The single non-radioactive strontium capsule is not represented in the figure.

MASS AND RADIOACTIVITY

The total mass of the cesium and strontium capsules and their contents is approximately 15 metric tons (NRC, 2003). Both the cesium and strontium capsules contain an average of approximately 5.96 lb (2.7 kg) of their respective materials (Randklev, 1994). The total radioactivity of the cesium, strontium, and daughter products in the capsules was 109 million Curies (Ci) as of August 6, 2006 (Carter *et al.*, 2013). The cesium and strontium capsules contain over a third of the total radioactivity at the Hanford Site, the remainder of which is stored mostly as HLW in underground tanks (Carter *et al.*, 2013; SNL, 2014). Due to the relatively short half-lives of cesium-137 and strontium-90, capsule radiation will diminish significantly after several hundred years, rather than tens of thousands of years, or longer, for longer-lived radionuclides (OTA, 1991). However, it would take more than 800 years for the radioactivity of strontium capsules to decrease sufficiently for these capsules to be classified as low-level waste (NRC, 2003). In 2048, which is the year DOE has set as its target for having a geologic repository for HLW constructed and operating (DOE, 2013), the total radioactivity of these capsules will decrease to approximately 44 million Ci (SNL, 2014).

STABILITY AND RADIONUCLIDE RELEASE

Cesium chloride can undergo a phase change as a result of the heat produced by radioactive decay. This phase change occurs at about 450°C (840°F) with pure cesium chloride, but can occur between 300 and 500°C (570 and 930°F) in the encapsulated cesium chloride, depending on the impurities present (DOE, 1990). When cesium chloride undergoes a phase change, the lattice structure of the salt alters, resulting in an increase in volume, which can cause the cesium capsule to bulge. This can cause failure of the capsule, increasing the possibility of radioactive material being released. Cesium chloride is very soluble in water, so any breach of the stainless steel capsule could release radioactive cesium to the environment, for example to the pool water at the WESF, or to groundwater in a mined geologic repository or in a deep borehole. In contrast to cesium chloride, strontium fluoride is highly insoluble in water, which would result in a low radionuclide release rate if a strontium capsule were breached. The WESF pool water temperature is maintained below 50°C (122°F) (DOE, 1990), which minimizes the potential for swelling and breach of the capsules during storage at the facility.

DOE is considering a number of options for geologic disposal of cesium and strontium, including direct disposal of the capsules in a repository or deep borehole, as well as extracting the cesium chloride and strontium fluoride from the capsules, blending them with other HLW waste streams, and then vitrifying them at an HLW vitrification facility (DOE, 2012; SNL, 2014).

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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.

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