U.S. NUCLEAR WASTE TECHNICAL REVIEW BOARD

CALCINED RADIOACTIVE WASTE

OVERVIEW[1]

From 1952 to 1991, the U.S. Department of Energy (DOE) and its predecessor agencies reprocessed spent nuclear fuel (SNF) at the Idaho Nuclear Technology and Engineering Center (INTEC), known prior to 1998 as the Idaho Chemical Processing Plant, to recover the highly enriched uranium that remained in the fuel and other materials. INTEC is located on the Idaho National Laboratory (INL) site near Idaho Falls, Idaho. The reprocessing operations generated liquid radioactive waste that was stored in underground tanks at the INTEC Tank Farm Facility. The liquid waste was converted into a solid, granular form, referred to as calcine, which has a much smaller volume than the liquid form, is less corrosive, and is less likely to be released in the event of degradation of the storage system. In the calcination process, liquid waste is sprayed onto a fluidized bed of spherical particles, which are most often composed of dolomite [$CaMg(CO_3)_2$], heated to 400–600°C (750–1,100°F) (Staiger and Swenson 2018). At calcination temperatures, water and nitric acid in the waste are evaporated, metallic nitrates are decomposed to metal oxides, and the calcine is formed in layers on the fluidized bed particles (Freeby 1975; Newby and O'Brien 2000). The resulting fine, granular material has particle diameters typically in the 0.3–0.7 mm (0.01–0.03 in) range (Staiger and Swenson 2018), similar to medium to coarse sand, and comprises mainly metal oxides, but it also contains fluorides, chlorides, phosphates, and sulfates. The liquid waste was calcined at the Waste Calcining Facility (WCF) from 1963 to 1981 and subsequently at the New Waste Calcining Facility (NWCF) from 1982 to 2000. In 2000, DOE decided to use a different process to treat the remaining liquid waste in the tank farm and shut down the NWCF. A 1995 settlement agreement between the State of Idaho, the Department of Energy (DOE), and the U.S. Navy (Idaho *et al*. 1995) requires DOE to treat the calcine waste and put it into a form suitable for transport to a permanent repository or interim storage outside of Idaho by December 31, 2035.

STORAGE AND LOCATION

The calcined wastes are stored in Calcined Solids Storage Facilities (CSSFs). Seven CSSFs were constructed, but only six had been used when DOE decided to shut down the NWCF (Staiger and Swenson 2018). Each CSSF consists of three to twelve stainless steel storage bins (43 total bins) housed within a reinforced concrete vault. The design of each CSSF is slightly different—they were designed and constructed at different times and by different contractors and each incorporates lessons learned from the construction and operation of the prior CSSFs. Figure 1 is an historical aerial photograph showing the locations of the calcine processing and storage facilities. The WCF calcine is stored in CSSFs I, II, and III. The NWCF calcine is stored in CSSFs IV, V, and VI. CSSF VII is empty. Table 1 summarizes the calcining operations at the WCF and NWCF, as well as the calcined waste storage locations. Figure 2 is a photograph of the storage bins in CSSF III.

[1] Unless explicitly stated, this fact sheet does not present U.S. Nuclear Waste Technical Review Board findings, conclusions, or recommendations and none should be inferred from its content.

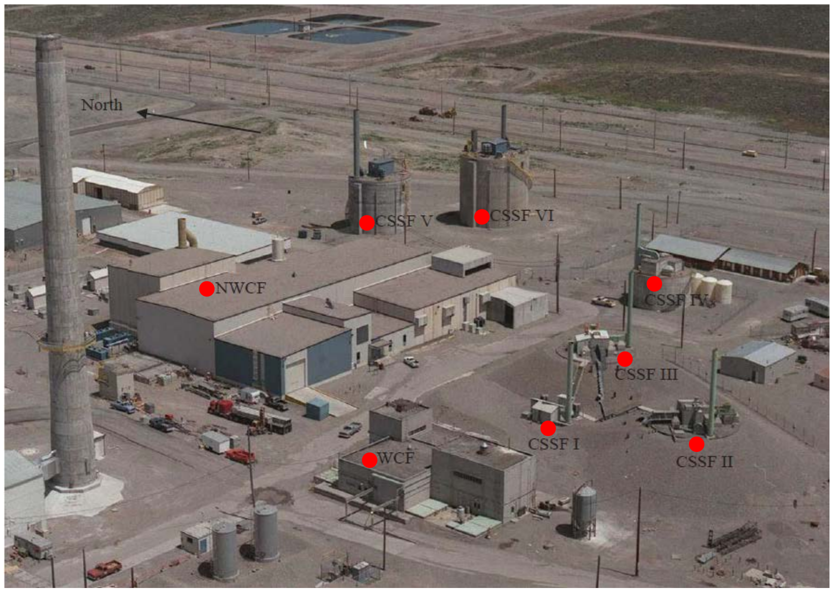

Figure 1. Historical Aerial Photograph Showing the Waste Calcining Facility (WCF) (Since Decommissioned and Demolished), the New Waste Calcining Facility (NWCF), and the Calcined Solids Storage Facilities (CSSFs) (Modified from Staiger and Swenson 2018).
Note: The photo was taken in the mid-1980s, prior to the construction of CSSF VII, which now is located east of CSSF IV in the figure.

Table 1. Summary of High-Level Waste Calcination and Storage at the Idaho National Laboratory (Staiger and Swenson 2018)

Calcined Waste Production Facility	Operating Years	Volume of Liquid Waste Processed	Volume of Calcined Waste Produced	Storage Facilities
Waste Calcining Facility	1963–1981	4,090,000 gal (15,500,000 L)	77,300 ft^3 (2,190 m^3)	CSSFs I, II, III
New Waste Calcining Facility	1982–2000	3,640,000 gal (13,800,000 L)	77,900 ft^3 (2,210 m^3)	CSSFs IV, V, VI

COMPOSITION

The composition of the calcine varies significantly because diverse types of SNF from numerous reactors were reprocessed. The different types of SNF generated chemically different liquid waste and, consequently, calcination produced chemically different calcine. The liquid waste and the resulting calcine often were named for the cladding on the SNF from which they were derived. For example, “aluminum waste” and “zirconium waste” were names applied to liquid waste generated from reprocessing aluminum- and zirconium-clad SNF, respectively. These names also were used to refer to the calcine, such as “aluminum calcine” or “zirconium calcine.” Liquid waste that contained relatively high concentrations of sodium (1 to 2 moles per liter) was called sodium-bearing waste (SBW). SBW included most of the waste generated from equipment decontamination and support systems (*e.g.*, ion exchangers, off-gas systems, scrubbers, and laboratory analyses). Some SBW was blended with other waste types prior to calcination to prevent clumping of solids in the calciner vessel. After the calcination of liquid radioactive waste was terminated, 900,000 gallons (3.4 million liters) of SBW remain in the tank farm awaiting treatment at INL’s Integrated Waste Treatment Unit.

Figure 3 shows the typical major chemical composition of four of the common types of calcine: aluminum, zirconium, Fluorinel/SBW blend, and aluminum nitrate/SBW blend. Fluorinel calcine is similar to zirconium calcine, except that Fluorinel calcine contains cadmium and sulfate from the cadmium sulfate and cadmium nitrate that were used as neutron absorbers in the Fluorinel fuel-dissolution process. Radioisotopes constitute only a small fraction (less than 1% by mass) of the calcined waste. The calcined waste also contains small amounts of chromium, mercury, nickel, lead, barium, silver, arsenic, and selenium.

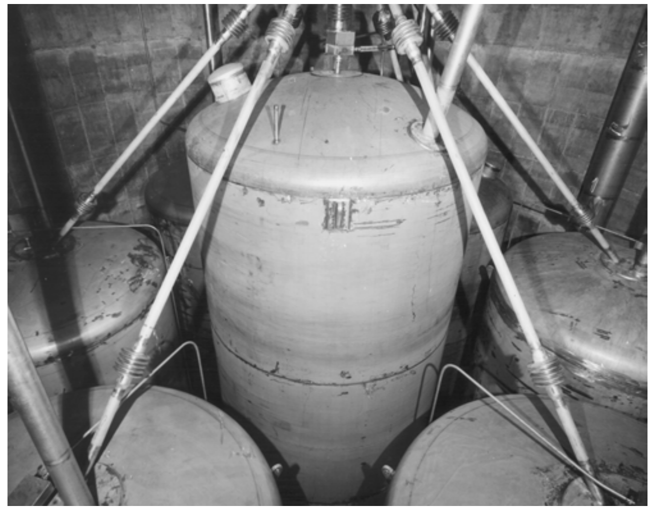

Figure 2. Storage Bins in CSSF III. The Center Bin is Surrounded by Six Shorter Bins (Staiger and Swenson 2018).

MASS AND RADIOACTIVITY

Approximately 4,400 m^3 (about 160,000 ft^3) of calcined waste, with a total mass of 5,600 metric tons (6,200 short tons) are currently stored in the CSSFs (Staiger and Swenson 2018). The total radioactivity of the calcined waste is estimated to be 27 million curies (on January 1, 2022) (Staiger and Swenson, 2018, Appendix A14). More than 99% of the radioactivity comes from the relatively short-lived cesium-137 and strontium-90, which have half-lives of 30.2 and 28.8 years, respectively, and their respective daughter products, barium-137m and yttrium-90 (Staiger and Swenson, 2018).

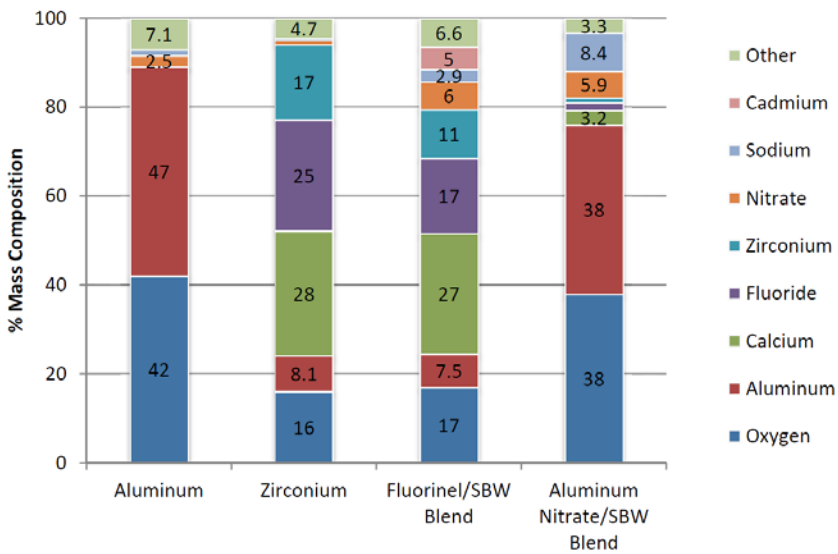

Figure 3. Typical Chemical Compositions of Four Calcine Types.
Data from Staiger and Swenson (2018).

PATH FORWARD FOR MANAGEMENT OF CALCINED RADIOACTIVE WASTE

DOE is planning to further process the calcined waste prior to disposal. In 2010, DOE published an Amended Record of Decision (DOE 2010) documenting the selection of hot isostatic pressing (HIP) technology to "treat calcine to provide a volume reduced monolithic waste form that is suitable for transport outside Idaho, with completion of treatment by a target date of December 31, 2035." In the HIP process, calcine and chemical additives would be mixed and then loaded into thin-walled canisters that would be welded shut. The canisters would be placed in a pressure vessel that would be heated and pressurized isostatically (uniformly on all sides) with argon gas. The temperature range for HIP processing of calcine is 1,050–1,200°C (1,920–2,190°F) and the pressure range is 7,200–15,000 psi (Case 2012). The net effect would be to produce a homogeneous, but multiphase, glass–ceramic waste and reduce the volume by approximately 40 to 60% compared with the original volume of calcine (Case 2012), without generating a secondary waste stream. According to DOE, the HIP technology has been demonstrated to generate a waste form with performance characteristics consistent with that of the single-phase borosilicate glass being produced from high-level radioactive liquid waste at DOE's Defense Waste Processing Facility at the Savannah River Site (DOE 2010).

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