

# Evaporation and Storage of Liquid Radioactive Waste

Claude B. Goodlett

## Abstract

The liquid wastes produced during the processing of radioactive materials at the Savannah River Site were initially stored in large underground tanks constructed of carbon steel. These liquid wastes were generated from the Purex process (for producing plutonium) and the HM process (for producing tritium). The liquid wastes were designated as high-level wastes and low-level wastes. As the production requirements increased with the resulting increase in liquid wastes, additional storage tanks and associated facilities were constructed. Since the waste volume was large and waste storage facilities construction was expensive, methods to reduce the cost of storing these wastes and increase the safety of storage were implemented. Storage tanks of differing designs and facilities to concentrate and to handle waste in the tank farms were constructed. These modifications resulted in significant cost savings and increases in liquid waste storage safety.

---

Liquid wastes produced during radioactive materials processing at the Savannah River Site were discharged to large underground tanks for interim storage. Processing these stored wastes is underway to convert them to a solid form for permanent disposal. A vitrified waste form will contain most of the radioactivity, and a concrete waste form (saltstone) will contain most of the chemicals. This paper addresses the interim storage of these liquid radioactive wastes from the initial processing of radioactive material, which began in 1954. During storage, these wastes were concentrated by evaporation to reduce their volume resulting in major economic savings and increase the safety of storage (Goodlett 1976a, 1976b).

## Types of Waste

The liquid radioactive wastes from the separations plants (221-F and 221-H) are alkaline, with a dissolved solids content of 30-35 wt %. Two different processes were used to produce plutonium and tritium. Although there were variations in each process, blending these wastes in the large storage tanks and using similar chemicals in the two main processes resulted in using related processes to handle the waste.

## Plutonium Production

Plutonium production used uranium metal in a rod or tubular form clad in aluminum. The aluminum cladding was dissolved from the uranium metal core in a solution of sodium hydroxide and sodium nitrate. The uranium metal core was then dissolved in nitric acid and processed through a solvent extraction/purification process (Purex process) to recover the plutonium and uranium. The wastes produced in the Purex process are of two general types; high-level waste, which contains sufficient radioactive fission products to produce decay heat at 0.5 to 5 Btu/(hr) (gal), and low-level waste, with fission product content 1/1000 to 1/100,000 that of the high-level waste, but still too high to discard to the environment. The low-level waste contains principally sodium aluminate and sodium nitrate from the caustic dissolution of the aluminum cladding on the irradiated fuel elements. The high-level waste, principally sodium nitrate with some sodium sulfate and sodium carbonate, contains nearly all of the radioactive fission products from processing the irradiated fuel elements. These liquid wastes are stored separately in underground storage tanks in the tank farms. During storage, both wastes separate into a layer of sludge and a layer of relatively clear supernatant liquid.

## Tritium Production

Tritium production uses fuel tubes of enriched uranium-aluminum alloy with an aluminum cladding. After removal from the reactor, these spent fuel elements were processed to recover the enriched uranium. The entire fuel tube, uranium plus aluminum, was dissolved in nitric acid with mercury as a catalyst. The resulting solution was processed through a modified Purex solvent extraction process called the HM process. The waste produced in the HM process was similar to a mixture of the two types of waste from plutonium production.

## Original Design Bases

The major liquid radioactive waste producers were the separations facilities located in the 200-F and 200-H Areas; specifically, the canyon Buildings 221-F and 221-H. Small quantities of liquid wastes from the Savannah River Laboratory and from the production reactors were sent to the 200-F Area.

### 200-F Area

The 200-F Area was originally provided with eight 750,000-gallon underground tanks constructed of carbon steel and designated as Type-I tanks. These tanks were of a cup-and-saucer design with the storage tank totally enclosed inside a 5-foot-high short steel tank, which served as a saucer. This entire unit was contained in a massive concrete tank. These tanks were coil cooled to remove heat from radioactive decay. The tank-in-tank arrangement would contain any radioactive material that might be spilled or leak from the primary tank. The concrete tank provided radiation shielding from the tank contents, was a vault to prevent water from contacting the carbon steel primary tank and saucer, and added seismic protection. An annular space between the concrete vault and primary tank was supplied with warm air to remove any moisture that might cause primary tank corrosion and facilitated visual inspection of the primary tank. As demonstrated during

operation, the dry warm air in the annulus dried waste that seeped through small cracks in some of the primary tanks.

Liquid radioactive waste from Building 221-F flowed by gravity to a diversion box in the tank farm, where the waste was routed to one of the eight storage tanks. Since the waste contained only dissolved solids, the velocity of waste in the stainless steel waste transfer lines did not have to be controlled; however, the waste transfer lines were built without low points that would allow solids to settle and plug the transfer line. The Head End precipitation step in the separations process produced a solid manganese dioxide ( $MnO_2$ ) cake. To prevent the transfer of solids to the tank farm, this cake was dissolved with gluconic acid. Adding of this organic acid was later discontinued when it was determined to be unnecessary. Radioactive waste from SRL and any other locations was trucked to an unloading station in 200-F before transfer to Building 221-F, where it was combined with waste from the 200-Area Laboratory (Building 772-F) prior to transfer to the tank farm.

### 200-H Area

Since the original concept was for Building 221-H to back up Building 221-F, only four 750,000-gallon underground tanks were provided. These tanks were identical to the Type-I tanks constructed in 200-F. To accommodate increased plutonium production, Building 221-H was placed in operation, and four additional liquid waste storage tanks were constructed. These carbon-steel storage tanks (Type II) were similar to the Type-I tanks, but held one million gallons. Because of water table considerations, this group of four tanks was constructed at a higher elevation than the original Type-I tanks. Consequently, the liquid waste had to be transferred by gravity feed and pumping.

The liquid radioactive waste flowed by gravity from Building 221-H to a diversion box in the tank farm, where it was routed to one of the

four Type-I storage tanks. Since the elevation difference required that the waste had to be pumped to the four Type-II storage tanks, a second diversion box and three pump tanks were added to the original set of four Type-I tanks. All waste transfer lines and the pump tanks were constructed of stainless steel.

## Second-Generation Waste Storage Tanks and Evaporators

To reduce the costs of storing these ever-increasing waste volumes, facilities were constructed in the tank farms to concentrate the stored waste and reduce its volume. The storage tank design was modified to reduce storage costs. Four uncooled waste tanks with a centrally located evaporator were constructed in the F-Area Tank Farm (Taber 1959). This facility became operational in 1960. These were the first facilities to concentrate the liquid radioactive waste in the waste tank farms. A similar facility containing four uncooled waste tanks with evaporator were constructed in the H-Tank Farm (Taber 1960); this facility became operational in 1963.

### Tank Farm Evaporator

In the mid to late 1950s, laboratory work at Brookhaven National Laboratory with nonradioactive wastes from the Purex process showed that the low-level waste, principally a mixture of sodium aluminate, sodium hydroxide, and sodium nitrate, could be evaporated to reduce volume. Experimental work on the design of an evaporator suitable to evaporate this waste was done by the Griscom-Russell Company in conjunction with Brookhaven National Laboratory. This work showed that a chemical scale would form on the surfaces of the steam coils but could be removed by the design of a unique evaporator.

- It featured a steam chest with the heating tubes installed in a bent condition. This allowed the tubes to flex when supplied by

alternating water and steam, thereby causing the scale to flake off of the heating surface.

- The lower portion of the evaporator was conically shaped to allow insoluble solids and the scale to settle.
- Since the contents of the evaporator were at the boiling point and design criteria did not allow bottom openings on vessels containing radioactive materials, a steam lift was used to remove the concentrated waste from the evaporator. A steam lift is a simple device consisting of an open pipe into which steam is injected at the lower end. The steam reduces the density of the concentrated waste in the pipe, allowing atmospheric pressure to carry the waste up the tube and out of the vessel (Goodlett 1963).

### Uncooled Waste Tanks

The heat load in the low-level waste was sufficiently low that cooling coils would not be required in waste storage tanks that only contained low-level waste. Eliminating the cooling coils resulted in significant cost savings. Since these uncooled waste tanks would only be used to store low-level waste, the low radioactivity level in this waste did not require the tank-in-tank containment that was necessary for the high-level waste. These Type-IV storage tanks were 1.3-million-gallon underground tanks constructed of carbon steel. These single-wall carbon steel tanks were encased in a blown-on concrete shell. Prestressed steel reinforcing bands were used to support and prevent cracking of the blown-on concrete. This type construction eliminated the annulus between the steel tank and the concrete tank, which allowed for moisture removal and visual inspection of the primary tank.

Although this type of construction was justified when these tanks were built, later experience with cracking and waste leaking through the walls of some of the double containment tanks indicated that a single wall tank would not provide the protection to prevent waste loss to the environment that was present in the tank-

in-tank steel tank. However, no single wall tanks have leaked waste to the environment. There was no annulus to allow inspection of the waste tank.

### **Waste Transfer Facilities**

Installing evaporators in the tank farm was the first step in changing the waste storage areas into processing facilities. This change has continued to date. The evaporators are fed soluble liquid waste from one of the waste storage tanks using one of two systems: (1) a steam jet (this method of transferring waste was used in the separations buildings), or (2) a feed pump (an adaptation of a standard deep-well jet-pump system). Because the use of steam jets added water to the waste while the objective of evaporation was to remove water, later feed systems utilized a feed pump (Goodlett 1968d, 1972).

The concentrated waste from the evaporator is different from the waste discharged to the waste tank farm from the separations buildings in that some or all of the waste would solidify on cooling and could plug the transfer lines handling the concentrated waste. To prevent this pluggage, it was necessary to keep the transfer line from the evaporator to the receiving tank as short as possible, well-sloped, and insulated.

### **Third-Generation Waste Storage Tanks and Evaporators**

As the production of nuclear materials continued, additional waste storage tanks and support facilities were needed. These needs resulted in the construction of additional waste storage tanks with a modified tank-in-tank design (Type III). The limitations of the single-wall tanks (Type IV) were recognized, and no more tanks of this design were constructed. Laboratory studies continued to improve the methodology to concentrate the high-level wastes from the Purex and HM processes to reduce volume and increase the safety of storage. Twenty-nine of these Type-III tanks (10 in 200-F Area and 19

in 200-H Area) and two additional bent-tube evaporators (one in 200-F Area and one in 200-H Area) were constructed. Also, facilities were installed to feed the waste to the evaporators and then transfer the waste from the evaporators to the waste tanks. In addition, a pipe line to transfer soluble waste between 200-F and 200-H Areas was installed, a distance of 2.5 miles (Goodlett 1968c).

### **Stress-Relieved Waste Tanks**

Experience with small stress-corrosion cracks occurring in the primary waste tank in some of the Type-I and Type-II tanks resulted in the design of a new waste storage tank, Type III. These 1.3-million-gallon tanks were constructed of carbon steel and were similar to the tank-in-tank design of the Type I and II tanks except that the outer tank was a full-height tank providing two barriers of steel. They were annealed by heating the primary tank after construction to relieve any stresses present. Some tanks have insertable coils that were added through tank risers after construction. However, most of these tanks have installed cooling coils like the Type-I and -II tanks. These tanks had an annulus and a concrete outer tank. Since they were constructed over a number of years, the designs were modified as new knowledge was acquired.

### **Evaporation of High-Level Wastes**

Experimental work carried out in the Savannah River Laboratory showed that all the wastes in the tank farms could be concentrated by evaporation to result in a solid when stored at room temperature (Goodlett 1968a). This process has been utilized in the tank farms.

The de-cladding waste from the Purex process is ideal for concentration. This aqueous waste contains no solid phase at its boiling point. A large difference exists between the boiling point of the solution and the temperature at which a solid phase is precipitated. When this solution is concentrated by a factor of 3.4, the hot concentrate is fluid, and a solid phase appears only

when the solution is cooled 60°C below its boiling point. The concentrated waste solidifies completely when cooled to room temperature.

Purex waste is more difficult to concentrate than decladding waste. When concentrated by more than a factor of 2.5, this solution contains a solid phase at the boiling point. The solid phase is initially a sandy, white precipitate of sodium carbonate and sulfate. Sodium nitrate is also crystallized as the solution is concentrated further or is cooled. When concentrated by more than a factor of about 3, the solution contains a large amount of solid phase at the boiling point. However, the concentrate does not solidify completely while cooled to 23°C; about one-third remains liquid. Although further concentration can produce a material that will solidify completely on cooling, the amount of solids present near the boiling point is too high to be handled in the bent-tube evaporator without causing pluggage. Purex waste cannot be concentrated sufficiently by evaporation in one stage. However, this waste can be eventually reduced to a solid by four stages of evaporation, each stage followed by cooling and partial crystallization in a waste storage tank. The volume reduction obtained is 3.4.

HM waste behaves similarly to Purex waste with the exception that only three evaporation passes and successive coolings are required.

### **Transfer of Concentrated Wastes**

Results from the SRL experimental data showed that the Purex and HM wastes could be evaporated sufficiently to generate a waste that solidified completely after cooling. However, this concentrated waste must be transferred over distances of hundreds of feet to waste tanks at elevations equal to or higher than the evaporator. All this must be done while maintaining a temperature close to its boiling point to prevent the settling of the undissolved solids present in the concentrated waste. Tests showed

that simulated concentrated waste slurries containing at least 20 vol % solids could be pumped through a 2-inch pipeline if the bulk velocity of the waste was maintained at 1.6 ft/sec or higher (Goodlett 1968b). Based on these data, several transfer loops were installed to transfer aged alkaline waste (after concentration in a tank farm evaporator) to distant underground waste storage tanks. The circulation rate in these tank farm transfer loops is 3 to 5 ft/sec; two or three times the velocity corresponding to the onset of pluggage. These systems have operated successfully since initial startup in 1967.

### **Overall Effect of Waste Concentration in Waste Tank Farms**

The evaporation of the liquid radioactive waste in the tank farms has significantly reduced the volume of stored waste. For example, in late 1986, the 77 million gallons of waste that was generated by operations in 200-F and 200-H Areas had been reduced to 32 million gallons, a reduction factor of 2.4 (Goodlett 1986). This volume reduction was less than achieved in the laboratory because all of the waste had not been concentrated to a solid. If this volume reduction had not occurred, additional waste storage tanks would have been required. Construction of these additional tanks would have increased the land area that contained radioactive facilities and would also have required costly subsequent decommissioning.

The waste transfer line between the 200-F and 200-H tank farms increased the safety and reduced the cost of waste storage by transferring waste between the areas.

Evaporation of the waste also increased the safety of waste storage because waste with a higher solids content was less prone to leak through any cracks that might develop in the steel wall of the storage tanks.

## Acknowledgments

The author gratefully acknowledges the assistance of the following persons in preparation of this paper: Faye J. Lewis in obtaining reference material, Al E. Symonds for editorial suggestions, and Susan S. Cathey, Harold L. Hull, and Mal C. Schroder for reviewing the contents.

## References

- Goodlett, C. B. Jr., 1963, *Concentration and Transfer of Stored Wastes, Steam-Lift Performance*, USDOE Report DPST-63-461 (RTA-255-S), Savannah River Laboratory, E. I. du Pont de Nemours & Co., Aiken, S.C. (October 11, 1963).
- Goodlett, C. B. Jr., 1968a, *Concentration of Radioactive Wastes*, USDOE Report DP-1135, Savannah River Laboratory, E. I. du Pont de Nemours & Co., Aiken, S.C..
- Goodlett, C. B. Jr., 1968b, *Transfer of Radioactive Slurries*, USDOE Report DP-1136, Savannah River Laboratory, E. I. du Pont de Nemours & Co., Aiken, S.C.
- Goodlett, C. B. Jr., 1968c, *Steam Jet for Interarea Transfer Line*, USDOE Report DPST-68-395, Savannah River Laboratory, E. I. du Pont de Nemours & Co., Aiken, S.C. (May 10, 1968).
- Goodlett, C. B. Jr., 1968d, *Completion of RTA-476-S, Steam Jet for Feeding Waste to Tank Farm Evaporator*, USDOE Report DPST-68-454, Savannah River Laboratory, E. I. du Pont de Nemours & Co., Aiken, S.C. (July 12, 1968).
- Goodlett, C. B. Jr., 1972, *Testing of a Higher Capacity Feed Pump for Building 242-F and Building 242-H Evaporators*, USDOE Report DPST-72-343, Savannah River Laboratory, E. I. du Pont de Nemours & Co., Aiken, S.C. (May 8, 1972).
- Goodlett, C. B. Jr., 1976a "Concentration of Aqueous Radioactive Waste with Wiped-Film Evaporators", *Radioactive Wastes from the Nuclear Fuel Cycle*, AIChE Symposium Series,

72, 154, American Institute of Chemical Engineers.

Goodlett, C. B. Jr., 1976b, "Putting Evaporators to Work: Concentration of Aqueous Radioactive Waste", *Chem. Eng. Prog.* 72(4), p. 63-64.

Goodlett, C. B. Jr., 1986, *Role of SRP*, Presentation to Representatives of the Nuclear Regulatory Commission, Washington, DC, (December 9, 1986).

Taber, B. L., 1959, *Design Data Report, 200 Area-Building 242-F, Waste Tank Farm Evaporator*, USDOE Report DPWA-59-202 (October 1, 1959).

Taber, B. L., 1960, *Design Data Report, 200-H Area-Buildings 241, 242, And 242-1H, Additional Waste Handling Facilities*, USDOE Report DPWA-60-207 (September 1, 1960).

## Biography

Claude Goodlett graduated from Clemson University with a BS Degree in chemical engineering in 1954. He worked for Du Pont at the Savannah River Site from graduation until he retired in 1989. His areas of work were primarily in the processes and equipment for the processing of irradiated nuclear fuel and handling and storing radioactive wastes. He worked in both research and production functions.

He served as Chairman of the Savannah River Section of the American Institute of Chemical Engineers and Chairman of the Joint Council of the Engineering and Scientific Societies of the Central Savannah River Area. He has authored or co-authored 22 papers.

After retiring from Du Pont, Mr. Goodlett has worked as a consultant for Los Alamos National Laboratory, Brookhaven National Laboratory, Battelle Pacific Northwest Laboratory, Ebasco Constructors, Stone and Webster Engineering Company, and Westinghouse Hanford Company.