Conceptual Design Considerations for the Storage of Solidified High-Level Waste in Canisters at a Commercial Fuel Reprocessing Plant

J. R. LaRiviere
E. L. Moore

November 1975

Prepared for the U.S. Energy Research and Development Administration
Under Contract E(45-1)-2130

Atlantic Richfield Hanford Company
Richland, Washington 99352
DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.
DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.
NOTICE

THIS REPORT WAS PREPARED AS AN ACCOUNT OF WORK SPONSORED BY THE UNITED STATES GOVERNMENT. NEITHER THE UNITED STATES NOR THE UNITED STATES ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION, NOR ANY OF THEIR EMPLOYEES, NOR ANY OF THEIR CONTRACTORS, SUBCONTRACTORS, OR THEIR EMPLOYEES, MAKES ANY WARRANTY, EXPRESS OR IMPLIED, OR ASSUMES ANY LEGAL LIABILITY OR RESPONSIBILITY FOR THE ACCURACY, COMPLETENESS, OR USEFULNESS OF ANY INFORMATION, APPARATUS, PRODUCT OR PROCESS DISCLOSED, OR REPRESENTS THAT ITS USE WOULD NOT INFRINGE PRIVATELY OWNED RIGHTS.
CONCEPTUAL DESIGN CONSIDERATIONS FOR THE STORAGE OF SOLIDIFIED HIGH-LEVEL WASTE IN CANISTERS AT A COMMERCIAL FUEL REPROCESSING PLANT

J. R. LaRiviere
Advanced Waste Engineering Department
Research and Engineering Division

E. L. Moore
Development Engineering Department
Research and Engineering Division

November 1975

To be presented at:
68th Annual Meeting of the American Institute of Chemical Engineers
Los Angeles, California
November 16 - 20, 1975

Atlantic Richfield Hanford Company
Richland, Washington 99352

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED
ABSTRACT

Onsite storage of canisters of solidified high-level waste generated by the commercial fuel reprocessing plants (FRP) may be required prior to shipping these canisters to a Federal repository. The most likely storage concept is to hold the waste-filled canisters in water storage basins. In the Retrievable Surface Storage Facility conceptual design studies, air-cooled and water-cooled storage of solidified high-level wastes has been considered. The studies of water-cooled storage included design considerations, as part of the conceptual design of the canisters and water storage basins, that would apply also to the conceptual design of similar facilities at an FRP. These similar considerations include: types of corrosion likely to develop on canisters stored in water, conditions which promote stress corrosion cracking (SCC), prevention of SCC, routine basin water cleanup, cleanup of a grossly contaminated water basin, effluent discharge discussions, storage basin integrity, and designing for decommissioning.
CONCEPTUAL DESIGN CONSIDERATIONS
FOR THE STORAGE OF SOLIDIFIED HIGH-LEVEL WASTE
IN CANISTERS AT A COMMERCIAL FUEL REPROCESSING PLANT

INTRODUCTION

The current regulations governing the disposition of high-level waste at fuel reprocessing plants (FRP) call for conversion of the liquid radioactive wastes to a dry solid within five years after reprocessing, and shipment to a Federal repository within 10 years (Figure 1). The most attractive method of storing the dry waste at the FRP is to package the solid waste in stainless steel canisters and store it under water in a water storage basin.

Atlantic Richfield Hanford Company (ARHCO) has been studying the problem of storing high-level waste canisters since early 1972. These studies have been conducted for the Atomic Energy Commission (AEC), and now the Energy Research and Development Administration (ERDA), as part of the overall waste management program of the Division of Waste Management and Transportation (WMT) (now the Division of Nuclear Fuel Cycle and Production).

Plans are to have a Retrievable Surface Storage Facility (RSSF) provide storage of the high-level waste canisters in the interim period pending demonstration of terminal storage. Various concepts for storage of the high-level waste at the RSSF have been developed. These include the:

- Water Basin Concept (WBC)
- Air-Cooled Vault Concept (ACVC)
- Sealed Storage Cask Concept (SSCC)

In the WBC (Figure 2), canisters of high-level waste are stored in racks under water in a basin. The basin water provides shielding and absorbs the decay heat. The basin water is cooled by circulation through a heat exchanger that transfers heat to a secondary water loop. The heat from the secondary loop is dissipated to the atmosphere via a cooling tower.
In the ACVC (Figure 3), canisters of high-level waste are overpacked and then stored in a concrete vault. Decay heat is removed by natural air convection through the bank of overpacks.

In the SSCC (Figure 4), canisters of high-level waste are sealed inside thick-walled, carbon steel storage casks. The storage casks are placed inside concrete shields, and the complete assemblies are stored in an outdoor area. Decay heat is removed by natural air convection through an annulus between the storage cask and the inner face of the concrete shield.

Since the WBC is similar to the type of storage that will most likely be used at an FRP, we will present some of the pertinent design considerations that we have encountered in our conceptual design work on the WBC. These design considerations, we feel, are worth studying for possible application to an FRP high-level waste storage basin.

**DISCUSSION**

Based on the current design plans and the draft acceptance criteria for the waste canisters, as developed to date for the RSSF, the solidified waste would be packaged in 300 series stainless steel containers called canisters that would be up to 2 feet in diameter and up to 15 feet long (Figure 5). It is possible that canisters with smaller diameters, such as 8 inches and 15 inches, and shorter lengths, such as 5 feet and 10 feet, would be used by the reprocessors.

Use of a water basin permits storage of waste with higher heat content per unit volume than does air-cooled storage. The waste and the canister wall can be maintained at lower temperatures in water than in air because of the superior heat transfer characteristics of water.

In designing a waste storage basin, the main technical concerns are to provide proper shielding for personnel protection, to remove decay heat, and to confine waste. The basin water provides several useful functions in meeting the needs of each of these concerns. By using the proper amount of water cover, the water effectively shields the operators from the emanating radiation of the high-level waste and, at the same time, provides for direct viewing of the waste canisters. The water also absorbs the decay heat from the waste and, through the use of a water-to-water heat
exchanger cooling system, its temperature can be controlled to furnish favorable conditions for maintaining waste confinement. The water also serves as a confinement barrier for solid waste in those instances where the solid waste has escaped from the canister.

The design considerations for heat transfer and shielding are briefly mentioned herein, but the application of existing heat transfer and shielding technology to water basin design is well known. The principal design considerations which we shall discuss relate to confinement of the waste within the canister and confinement of contamination found within and emanating from basin water.

Since the waste canisters would always be handled under water, all radiation confinement problems would be associated with contaminated basin water. Contamination would occur through canister failure or through a slow buildup process in which contamination would move from the surfaces of previously contaminated canisters, racks, or basin liners to the water. We shall, therefore, discuss water environment factors and their relationship to canister failures, routine basin water cleanup systems, systems needed for the cleanup of a grossly contaminated basin, control of effluents, and storage basin integrity.

In light of these topics, we shall also comment upon planning for ultimate decommissioning of the facility.

WASTE CANISTER FAILURE

During conceptual design of the RSSF, the construction material for the reference waste canister was specified as AISI, 304L stainless steel. As one of the austenitic stainless steels, this alloy exhibits low corrosion rates in high purity water at ambient temperatures. Consequently, failure of a 304L stainless steel waste canister in water storage due to general corrosion is not considered a possibility. However, there are other types of corrosion, such as pitting corrosion and chloride stress corrosion cracking, which could result in premature canister failures.

Pitting corrosion occurs in crevices or under deposits found on the surface of the stainless steel. This form of attack results from the depletion of oxygen at the metal surface under the deposit or in the crevice. These areas become anodic to the more oxygen-rich surrounding areas and, since the anodic areas are much smaller in comparison with the surrounding cathodic areas, corrosion under the deposit or within the crevice is extremely rapid.
Szaklarska and Smialowski have published a review of the literature on pitting corrosion of steels which discusses the variables that affect this form of localized attack.\(^{(1)}\) In particular, they review the role of the chloride ion. To prevent loss of integrity of a waste canister because of pitting or crevice corrosion, care must be exercised in design to eliminate crevices and in storage basin operation to prevent deposits from forming on the canister surface.

Chloride stress corrosion cracking (SCC) is of greatest concern as a potential corrosion mechanism that could cause loss of waste canister integrity in the water storage basin. This form of localized attack occurs as a result of the combined action of tensile stresses and corrosion at elevated temperatures. The chloride ion is mainly responsible for the cracking of austenitic stainless steels in water, which occurs more readily at higher stress levels. A great deal of study has been devoted to this mode of localized attack upon stainless steels and reported in the literature by Ashbaugh, Bryant and Le Surf, and Berry, to name a few.\(^{(2, 3, 4)}\) A review by Latanision and Staehle also gives an excellent coverage of the literature on stress corrosion cracking of iron-nickel-chromium alloys.\(^{(5)}\) The work cited was performed under conditions generally more aggressive than those which would be experienced in water basin storage. However, stress corrosion cracking of the 300 (austenitic) series stainless steels has occurred under conditions which may appear quite innocuous. For example, SCC of stressed 304 stainless steel has been observed in water containing 10 ppm chloride at 165° F.\(^{(6)}\) Failures by SCC have also occurred in environments containing 1 ppm chloride where a mechanism exists for concentrating chloride, such as localized boiling. Stress levels for initiating cracking can be quite low. Staehle, et al., found that in 400° F water containing 50 ppm chloride ion, threshold stress levels for cracking 347 stainless steel tubing were below 2000 psi.\(^{(7)}\)

The major sources of stress in the waste canister would be residual stresses resulting from the welding process during fabrication. Stress levels would approach the yield strength of the stainless steel and, in the presence of a few parts per million chloride ion at temperatures above 150° F, could cause cracking. It seems obvious, then, that if one were able to reduce the stresses below a dangerous level by a procedure such as thermal stress relief, the susceptibility to cracking would be eliminated. However, it is not possible to specify this "dangerous stress level" below which cracking will not occur because of the complex interaction of environmental factors and metal composition.
While thermal stress relief does seem to be an obvious means of reducing susceptibility to SCC, this treatment may introduce some problems of a metallurgical nature. The stress relief treatment would require that the canister be heated to 1800°F and slow-cooled. Slow cooling through the range of 1400°F to 1000°F will sensitize (form carbide in the grain boundaries) the stainless steel and possibly increase its susceptibility to SCC at much lower chloride concentrations and temperatures than if the metal has not been sensitized. Of course, tensile stresses must subsequently be reintroduced in the canister by a means such as denting, since the presence of tensile stress is one of the conditions necessary for SCC to occur. The extra low carbon grade stainless steel, 304L, is not as susceptible to sensitization as are the normal carbon grades; nevertheless, some sensitization of the 304L can be expected and may or may not increase its susceptibility to SCC. This would have to be established.

The use of post-fabrication stress relief may not be practical. In addition to the possibility of increased susceptibility of the 304L canister to SCC as the result of sensitization, major problems could occur with the control of dimensional tolerances during heat treating. Additionally, should the waste form be a glass, the act of filling the previously stress-relieved waste canister with the molten glass may very well reintroduce severe stresses because of differences in the thermal expansion of metal and glass.

For these reasons, certain problems could result from using thermal stress relief as a means of reducing susceptibility of stainless steel type 304L to SCC in a water environment. The advantages and disadvantages of its use need to be carefully evaluated during design.

Additional methods of reducing or perhaps eliminating SCC include: controlling chloride content of the water, controlling water temperature, designing to eliminate crevices where chlorides can concentrate, and substituting alternate materials. Considering the last measure first, substituting a higher alloyed material such as Inconel Alloy 600 or Incoloy Alloy 800, which are more immune to chloride SCC, may be the best approach to solving the problem. Some feel, however, that if a steel such as 304L will crack in service, a higher alloy will probably also fail after longer exposure. The higher alloys are more costly; consequently, the benefit to be gained by their use would require careful evaluation.
Control of chloride concentration and basin water temperature would be imperative to the prevention of SCC regardless of container material. A realistic control level would be a maximum of 10 ppm chloride and 150°F.

When considering the consequences of canister failure in the water basin, thought must be given to waste form, because calcine waste is much more dispersible and leachable than glass-form waste.

**ROUTINE BASIN WATER CLEANUP SYSTEMS**

The waste storage basins would most likely operate at a maximum water temperature of about 120°F. One method of removing the decay heat from the basins consists of using a water-to-water heat exchanger. The basin water would be pumped through the heat exchanger and would be considered the primary cooling stream. It would be cooled by a second water stream called the secondary cooling stream.

The primary cooling stream would be pumped from the top of the basin, through the heat exchanger, and returned to the bottom of the basin. A fraction of the primary cooling stream would be continuously routed through a water filtration system for routine cleanup of particulate matter. Filtration would provide water clarification and help keep the chloride content of the water low, because particulate matter can be high in chloride content. When the radioactivity or the chloride content of the basin water approaches the maximum allowable level, this fraction of the primary cooling stream would also be routed through ion exchange systems to reduce radionuclide and/or chloride concentration levels. All wetted parts of the primary cooling system should be stainless steel, type 304L or better, for ease of decontamination.

The secondary cooling stream would be pumped from a cooling tower sump, through the heat exchanger, and to the top of the cooling tower for rejection of the heat to the atmosphere. The pumping pressure of the secondary cooling stream in the heat exchanger would always be higher than that of the primary cooling stream, so that in case of a leak in the heat exchanger at a time when the primary cooling stream is contaminated, the leakage in the heat exchanger would be from the secondary to the primary system and would not contaminate the secondary system.
All of the equipment in the primary cooling system, and this includes the pump, the heat exchanger, the water filter, and the ion exchange systems, should be located in shielded areas to protect personnel from radiation should the basin water become contaminated. Designers should also consider the need to maintain or replace this equipment when the basin water is contaminated. Remote or semiremote types of maintenance and replacement would be required.

**CLEANUP OF A GROSSLY CONTAMINATED BASIN**

Equipment provisions are needed for the cleanup of a grossly contaminated basin that might result from failure of a canister stored in the basin. We shall discuss an approach to cleanup that would maintain normal basin water temperatures by continuing the circulation of primary cooling water through the heat exchanger, where it is cooled by the secondary cooling water stream.

Routine cleanup of the basin water would divert only a fraction of the primary cooling water through filters or ion exchangers for chloride or radionuclide removal. Gross cleanup, however, would require an ion exchange system of relatively high capacity, because the flow diverted from the primary cooling water stream would be much larger than that normally diverted for routine cleanup. Upon completion of gross radionuclide removal from the basin water, the ion exchange column would be remotely removed, packaged, and transferred to the contaminated failed equipment storage area. The basin water would still be contaminated, but at a much lower level. It would then be routed to the liquid waste treatment system for concentration and disposal. At the same time, fresh demineralized water would be added to the basin to maintain an adequate level of water for shielding and cooling during this time. This addition of makeup water to the contaminated basin water would add to the processing load of the liquid waste treatment system, and these increased liquid waste volumes need to be considered during design of the waste treatment system.

When contamination of the basin water had been reduced to a reasonable level, the contaminated canisters would then be transferred to a spare basin. Provisions would be needed to rinse off the canisters to minimize the spread of contamination to the spare basin. During rinsing, it would be desirable to keep the canisters immersed to minimize temperature fluctuations, so high-pressure under water flushing could be used to clean off the canisters. Remote operation of the
water basin crane and adequate viewing from behind a shielded barrier would be needed to transfer either the bare canister or a cask containing the canister or canisters. After the removal of all the canisters from the basin, the remaining water in the basin would be discharged to the liquid waste treatment system for concentration and disposal.

The basin and the racks would then be decontaminated with appropriate chemicals and decontamination techniques, and as much of the work as possible would be done remotely in order to minimize the total man-rem dosage of the operation. For ease of decontamination, it would be highly desirable to have the basin liners and canister storage racks fabricated of stainless steel, type 304L. During design and fabrication of the liners, and especially the racks, care needs to be exercised to provide these items with smooth surfaces in order to minimize crevice formation. Special decontamination equipment requirements need to be considered in planning for canister rack cleanup. The decontamination chemicals would also be routed to the liquid waste treatment system for concentration and disposal.

During the period of gross radionuclide removal by ion exchange, shielding blocks covering the basin would greatly reduce the amount of radiation exposure at the top of the pools. The space between the water level and the cover blocks would need to be positively vented to prevent the accumulation of hydrogen above the water surface. The cover blocks could be designed for remote installation, and viewing could be provided through the use of shielding windows and television.

CONTROL OF EFFLUENT DISCHARGES

Radioactive liquid effluents should not be discharged to the natural area drainage system. If this is not feasible, the waste treatment system for liquid effluents discharged to unrestricted areas should ensure that the radioactivity in such effluents is as low as practicable and well within the limits of 10 CFR 20. All liquid discharges from the water basin storage area should be routed to the liquid waste treatment system. There they would be collected and sampled to determine the radionuclide content. Treatment and disposal of the wastes would be decided based on the radionuclide content.
Should the water basin become contaminated, for whatever reason, some fraction of the contained volatile radionuclides could escape. In addition, decontamination and other routine operations could result in airborne radioactive materials. The ventilation system should therefore be designed to keep the amount of radioactive material as low as possible in the ventilation exhaust stream and the personnel occupancy areas and well within the limits set by 10 CFR 20. It could be designed as an emergency system that would activate upon receiving a signal that the concentration of radionuclides in the air had approached or reached the maximum allowable level. The use of cover blocks over a contaminated basin would aid in minimizing the problems encountered during cleanup of a grossly contaminated basin.

STORAGE BASIN INTEGRITY

It is important to maintain an adequate water level in the storage basin. A supply of makeup water is needed to maintain the water level should leakage occur. An emergency water supply is also needed.

The stainless steel liner in the storage basin serves several purposes. It provides protection to the concrete, it provides a decontaminable surface, its use minimizes the amount of particulate matter flaking off the wall into the water, and it provides protection against normal leakage from the basin. The last function requires that: (1) the stainless steel liner be fabricated to form a tight pan with no leak paths, and (2) sumps be installed in the concrete beneath the liner in case liner leaks develop. Water collection troughs that lead to the sumps could be installed in the concrete walls and the floor of the basin. As the sumps collect water, a liquid-level alarm should sound to signal the operator to take remedial action.

The water basin would be designed to withstand the design basis earthquake, a maximum probable earthquake. The racks supporting the waste canisters would be designed to prevent earthquake damage to the canisters. Earthquake-induced canister failure or gross basin leakage is therefore not considered likely.

Total loss of water from the basin is a concern because of the potential for severe consequences. Without water for cooling, the canisters will ultimately heat up to a temperature at which a melt-down could occur, depending upon the type and age of the waste and the geometric arrangement of
the canisters. Reference 8 gives guidance on the design of fuels storage basins to prevent total loss of water. In applying this same guidance to a waste storage basin, one would probably specify that the basin be built either in impervious soils or with a secondary water containment envelope packed with fill material. The leak rate afforded by either design should be low enough that, in the event of a gross basin leak, makeup water could be supplied to the basin at a rate sufficient to keep the waste canisters adequately covered. A containment envelope could ensure a very safe, though very expensive, facility; but some designers feel these higher costs are justifiable because of a leak's severe potential consequences.

PLANNING FOR ULTIMATE DECOMMISSIONING

Decommissioning plans need to be considered during the design of the facility. Some of the design considerations applied to the problems of providing decontaminable surfaces are also applicable to decommissioning requirements. These include the avoidance of rough surfaces, cracks, and crevices in potential contamination areas. Providing adequate decontamination equipment for routine cleanup operations and having adequate operating procedures in effect will also help to minimize the work required in decommissioning the facilities.

Consideration should also be given to disposal of the potentially contaminated material beneath the basin, whether it is impervious soil or fill material from the secondary basin water containment envelope discussed above.
REFERENCES

(1) Szaklarska - Smialowski, Z., Corrosion, Vol. 27, No. 6, p 223 (June 1971)


(4) Berry, Warren E., Reactor Materials, Vol. 7, No. 1, p 1 (Spring 1964)


(6) Scharfstein, L. R., and Brindley, W. F., Corrosion, Vol. 14, No. 12, p 588t (December 1958)


COMMITMENT

10 CFR 50, APPENDIX F

"POLICY RELATING TO THE SITING OF FUEL REPROCESSING PLANTS AND RELATED WASTE MANAGEMENT FACILITIES"

REQUIRES THAT HIGH-LEVEL WASTES BE CONVERTED TO A CHEMICALLY, THERMALLY, AND RADIOLYTICALLY STABLE SOLID WITHIN FIVE YEARS OF SEPARATION AND SHIPPED TO A FEDERAL REPOSITORY NO LATER THAN 10 YEARS AFTER SEPARATION.

FIGURE 1
RETRIEVABLE SURFACE STORAGE FACILITY
WATER BASIN CONCEPT
HEAT REMOVAL SYSTEM

Figure 2
RETRIEVABLE SURFACE STORAGE FACILITY

AIR-COOLED VAULT CONCEPT

AIR INLET TEMPERATURE 110°F

DOSE RATE AT TOP OF DECK 2 MREM/HR

CONCRETE SURFACE TEMPERATURE 200°F

OVERPACKED WASTE CANISTER

EMPTY SLEEVE WITH TEMPORARY AIR SEAL

EXHAUST PORT

AIR OUTLET TEMPERATURE 210°F

FIGURE 3
RETRIEVABLE SURFACE STORAGE FACILITY

RECEIVING AND ASSEMBLY BUILDING

AIR OUT

GAMMA-NEUTRON SHIELD

WASTE CANISTER

SEALED STORAGE CASK

AIR IN

CONCRETE SUPPORT PAD

FIGURE 4
RETRIEVABLE SURFACE STORAGE FACILITY
HIGH LEVEL WASTE CANISTER

SIZE
6 TO 24 INCH DIAMETER
10 TO 15 FEET LENGTH
12 INCH DIAMETER × 10 FEET LENGTH TYPICAL

MATERIAL
300 SERIES STAINLESS STEEL

HEAT
< 1 TO 20 KW
5 KW TYPICAL

FIGURE 5