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WASTE MANAGEMENT IMPLICATIONS OF IRRADIATED NUCLEAR FUEL STORAGE

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INTRODUCTION

Irradiated nuclear fuel is temporarily stored underwater in spent fuel pools after reactor discharge. The fuel is then transferred to chemical separations plants where it is stored underwater in large basins prior to reprocessing. In the United States a critically short capacity to process spent reactor fuels is developing. This trend is forcing operators of U.S. commercial and development reactors to face increasingly longer-term storage of their spent fuel charges. Currently an estimated 68 percent of the fuel assemblies discharged from operating commercial reactors remain in storage at the sites where they were irradiated[1] and many existing fuel storage basins are rapidly approaching their design capacities. The current approach to the problem has been to modify existing basin storage arrangements to obtain maximum fuel storage capacity within basin criticallity limits. The long term solution is, of course, to increase U.S. fuel reprocessing capacity within the provisions of the Presidential Nuclear Policy Statement issued October 28, 1976.

As with other reactor operating sites, the Hanford N Reactor fuel storage basin is near capacity. Efforts are in progress to convert basins at two adjacent shutdown reactors for additional storage of N Reactor fuel. The N Reactor basin and these older basins have unlined, uncoated concrete walls and no recirculating purification system. As an initial step, a literature review and several contacts at other fuel storage basin sites were made to assess current operating practices and waste management implications as a result of these practices. This report discusses the sources and amounts of radioactivity in spent-fuel storage basins, current methods of controlling the radioactivity at manageable levels, and the amounts of waste generated.

DISCUSSION

A. Sources of Radioactive Wastes

The principal source of radioactivity in fuel storage basins originates from leaching of fission products from fuel with defective cladding. For aged fuel the predominant radionuclides affecting exposure and waste management control are ^{137}Cs, ^{3}H, ^{60}Co, ^{90}Sr, $^{144}CePr$, and transuranics. The radioisotope content of a composite reference LWR spent-fuel assembly has been calculated to be about 28 PBq (0.76 MCi) at 1-year after discharge.[2,3] Of this total 85.5 percent is calculated to be fission products, 1.4 percent activation products and 12.5 percent transuranics. The significance of leaching through defective fuel claddings can be illustrated by considering that only about 370 Bq/ml (0.01 μCi/ml) fission product concentration uniformly distributed in a typical basin contributes about 5 mrem/hour to the basin exposure rate.

Activated corrosion products leached or mechanically dislodged from deposits on fuel surfaces also contribute to the radionuclide inventory in spent-fuel pools. In the absence of fuel cladding failures this mechanism is the principal source of radioactivity in basin water. At reactor sites where the reactor vessel and transport canal are integrally flooded during transfer operations, additional quantities of fission and activation products from the primary coolant system can enter the basin. Unloading of fuel transport casks at reprocessing site basins is an additional source of radioactivity in these basins.

Typical concentrations of radionuclides in basin water are given in Table I. It should be noted that almost every fuel storage basin has a unique set of operating conditions that can cause some variation from the concentration ranges shown. Examples are differences in amounts of defectively clad fuel in storage, and variations in fuel leaching rates, basin volumes and purification system flow rates.

B. Routine Wastes Generated During Basin Operation

Most routine wastes generated during fuel storage basin operation originate from filtration and ion exchange purification of the water and filtration and absorption of airborne radioactivity. A typical treatment system is shown in Figure 1. The HEPA filters for removing airborne particulate materials and the iodine absorption columns of charcoal or silver zeolites have very long service lives and contribute little to total waste volumes; therefore, no further discussion of these units will be made. Many variations in the water purification flow diagram in Figure 1 are practiced at the various sites. At some sites the filtration operation is performed on a separate stream originating from the pool skimmer. Some sites employ additional filtration downstream of the ion exchange columns. Other sites combine the filtration and ion exchange operations in one unit. At some sites the full purification stream is processed through all treatment steps while at other sites the ion exchange process may be conducted on only a small fraction of the flow passing through the heat exchangers and filters. Some of the various filtration and ion exchange practices are discussed below.

Filtration

The results of a survey to determine the types of filtration units employed for various fuel storage basins are given in Table II. Precoat filters are most commonly employed at Boiling Water Reactor sites and spun fiber cartridge filters at Pressurized Water Reactor sites. Fuel storage basins at reprocessing sites and at older ERDA facilities have employed both filter types and in addition have had good success with sand filters.

The precoat filters employ various coatings such as Solka-floc, diatomaceous earth, and powdered ion exchange resin, either singly or in combination. The powdered ion exchange resin (Powdex) is most commonly used and performs a dual function of removing turbidity and dissolved ionic impurities. Several of the basins employ a conventional mixed bed ion exchange column downstream of the Powdex filters and in these cases the Powdex removes radioactivity to extend the mixed bed operating periods but can be operated past ionic breakthrough and replaced only upon high pressure drop. Loss of flow through precoat filters causes the filter cake to drop off requiring a new coat and generating extra waste. Several sites have incorporated a small auxilliary pump to maintain sufficient flow to prevent inadvertent loss of the precoat.

Powdex precoat filters normally operate for a few days to a few weeks. The thin layer of mixed anion and cation resin shards on the filter has little ion exchange capacity and high cesium leakage is common. A novel method of overcoming this feature is employed at the Midwest Fuel Recovery Plant (MFRP). They first place a layer of Solka-floc on the filter then a mixture of Powdex and Zeolon 100. The Zeolon 100 is specific for cesium removal and significantly extends their operating runs. Operating runs of 4-5 weeks are now obtained and cesium activity has been reduced by a factor of 10-20 from previous levels. A few years ago the MFRP basin was contaminated with 200-300 ppm sodium nitrate when a cask containing sodium nitrate as antifreeze was drained into the basin. Rapid ionic breakthrough of the Powdex unit occurred following the incident and many changeouts were required to restore normal filter operation. The basin has since been modified to permit cask draining to the low activity waste tank.

Cartridge filters have much longer operating periods but remove none of the ionic impurities. Some sites change their cartridge filters as infrequently as once/year while other sites change filters 4-6 times/year, mostly during refueling outages when turbidity is high. The filter housings are generally discarded with the filters to reduce personnel exposure and to eliminate contamination spread. Because manual changeout requirements were causing one site too much personnel exposure, they eliminated their cartridge filters and rely on the ion exchange column to filter turbidity from the water; this is economically practical only if turbidity does not cause too high a pressure drop before the column is ionically exhausted.

Most sites ship their Powdex wastes as dewatered resin but a few sites are now incorporating the Powdex wastes in cement or urea formaldehyde during solidification of other liquid wastes. Cartridge filters are commonly shipped as dewatered waste but because of the high dose rates (5-50 rem/hour), it is becoming more common for them to be packaged in the solidification system. Typical waste volumes generated are shown in Table III. While the volume of a cartridge filter is small (10-15 litre), the volume of the downstream ion exchange resin column must be considered in the total purification system wastes generated. The Powdex systems generate 2-4 metre^3/year (75-150 ft^3) waste and the cartridge filter-mixed bed systems generate 0.5 - 3.5 metre^3/year (20-120 ft^3) with the median being at the lower end of the range. In addition, those Powdex systems with downstream mixed bed units also have waste contributions from these units. While this survey shows that strictly from the waste volume generation standpoint the cartridge filter-mixed bed system has the advantage, many other factors such as operating problems, exposure, and remote handling equipment must be considered. In general, where one type of filtration system is used in the primary cooling and radwaste systems, it makes sense to use that type of system in the fuel storage pool.

The newer ERDA fuel storage basins all utilize demineralized water in stainless steel lined or coated concrete basins and employ stainless steel or aluminum components. The waste management implications of these facilities are similar to the commercial facilities already discussed. In contrast, the older ERDA basins, such as the Savannah River (SRO) and Hanford reactor basins and the Idaho Chemical Processing Plant (ICPP) basin at the Idaho National Engineering Laboratory, have bare concrete walls, carbon steel piping, and do not employ demineralized water. These basins originally maintained clarity and radiation levels with a feed and bleed system discharging to a leaching pond. All except one have now been upgraded to recirculating systems. Needless to say, under these conditions the waste volumes generated while maintaining basin water purity and clarity are much greater than shown in Table III for the newer basins. In respect to filtration, numerous types of filters have been utilized and, after a considerable amount of experience and testing, the Savannah River reactor basins were fitted with backwashable dual media pressure sand filters.[4] Operation with these units has been very successful. ICPP has just converted their diatomaceous earth precoat filter system to a sand filter system and the N Reactor basin is scheduled for a similar installation. The SRO reactor basin filters are backwashed to a sludge collection tank where they are settled and decanted. About 4-8 metre^3 settled sludge/year are generated.

Ion Exchange

Examples of the types of ion exchange units utilized in fuel storage basins are included in Table II with the filter data. With the exception of the dual function Powdex units, already discussed, all ion exchange units are mixed bed resin columns with volumes generally ranging from 0.5 - 2.0 metre^3 (20-60 ft^3) of resin. A few of the basins have facilities to regenerate the resins but in practically all cases the resins are dewatered and discarded when depleted. The resins are normally replaced from once every two years to twice/year, depending on system ionic concentrations, radioactivity levels, resin volumes, and flow rates. Typical waste volumes (Table III) range from <1-4 metre^3/year. Volumes at the newer sites, which have lower fuel leak rates and fewer operating problems, are at the lower part of the range.

The volumes of mixed bed resin wastes generated during normal operating periods are quite low in comparison to the total wastes generated at modern reactor or reprocessing sites. However upsets in basin water chemistry control can significantly increase the volumes of wastes. The example of sodium nitrate contamination at the MFRP was previously discussed. At one reactor site, a small amount ($\sim$4 litre/min) of basin water leakage through the stainless steel liner is collected and treated by a small resin column. Due to leaching of impurities from the concrete this column must be replaced once/week. This small leak generates almost double the amount of waste generated by the basin purification demineralizer.

Ion exchange requirements at the older ERDA basins are much more severe than at the more modern basins. At the Savannah River Operation reactors, a portable 1.7 metre^3 (60 ft^3) mixed bed ion exchange unit is employed. Due to the large amounts of nonradioactive impurities present from the well water makeup stream and along with those leached from the concrete and corrosion products, the columns are depleted in 1-4 days. At this frequency, it is uneconomical to discard the resins; therefore, they are transferred to a central facility for regeneration.

A different approach was taken at the ICPP basin several years ago when this bare concrete basin was placed on recirculation. As the radioactivity levels gradually increased, primarily due to ^{137}Cs, disposable units of clinoptilolite,a natural mineral specific for cesium removal, were put into service. By removing only the cesium, very low volumes of waste were generated. These original units were later upgraded with larger units containing Zeolon-900, a synthetic mineral even more effective for cesium removal, and a cation unit in the sodium form was added to remove strontium and to maintain a neutral pH in the basin water to minimize concrete leaching.

Because of the high concentrations of short half-lived radionuclides present in the N Reactor basin following a fuel discharge, a sand filter followed by two regenerable 1.7 metre^3 (60 ft^3) cation columns and a regenerable 1.7 metre^3 anion column is being designed for installation. This type of ion exchange system will provide water of near-demineralized purity. Pilot scale tests have shown that this purity of water will not cause significant deterioration of the bare concrete walls but the concrete leach products will cause an additional 6-8 column depletions each year beyond those required to control the radioactivity.[5] With the expected depletion rates, it is more economical to regenerate the resins than to discard them.

C. Non-Routine Wastes From Fuel Storage Basins

Non-routine wastes are defined as those wastes that are generated only once in several years but provide a significant volume when they do occur. The programs at many fuel storage basins to replace existing fuel storage racks with new racks permitting up to three times existing storage capacity are a current example. The fuel is normally transferred to one end of the fuel pool, the vacated portion isolated and drained, and the old racks are cut, loaded into casks and shipped to burial sites.

Removal of sludge from basin floors after many years of service has been necessary at several sites because these sludges are stirred up and restrict visibility during fuel handling operations. At the older ERDA basins these sludges, which are primarily dust, dirt and iron and aluminum corrosion products, have reached several inches in thickness. The problem isn't nearly as severe in the more modern basins with stainless steel liners and equipment but in several cases the sludge has built up to the point requiring vacuuming. The sludges absorb large amounts of radionuclides and special care must be taken to design vacuum systems to keep exposures low. Plutonium isotopes, which are not routinely analyzed, can absorb in the sludges and could require the wastes to be classified as retrievable storage waste. Any waste with total plutonium concentrations greater than 10 nonocuries/gram must be placed in retrievable storage at U.S. burial sites. At the Hanford KE Basin where the dissolved plutonium concentrations are increasing awaiting installation of the ion exchange columns, it is expected that 10 nanocuries Pu/gram will be exceeded on the mixed bed resins if they are operated to complete exhaustion, requiring that the resins be classified as retrievable storage waste. For this case, one alternative would be to cease operation before the limit is reached so that retrievable storage would not be required. This alternative, however, would generate significantly more wastes.

Biological growths can cause gross complications in any storage basin and deserve special mention.

Biological control problems are rather infrequent in the newer basins which employ high purity demineralized water, but during system upsets or in basins using lower purity water the problem can be extreme. One basin surveyed uses a unique approach by maintaining the water temperature below 13°C with a water chilling system and has effectively controlled biological growths without the use of biocides. Of the many alternative biocides commercially available that we have tested, most are not economically competive with chlorination, even considering the effects of chlorination on resin depletion. Most biocides tested caused an unacceptable clouding of the basin water that restricted visibility through a 6 metre water depth.

Other examples of non-routine wastes are components and debris remaining from in-reactor tests or dissassembled fuel components. These normally are of small volumes but may be extremely radioactive and require special collection and disposal.

REFERENCES

[1] STRAUSS, S.D., Meeting the squeeze on nuclear fuel storage, Power, February 1976, 53.

[2] United States Energy Research and Development Administration, "Alternatives For Managing Wastes From Reactors and Post-Fission Operations in the LWR Fuel Cycle", ERDA-76-43, 1, (1976), 2.35.

[3] Wastes From Spent-Fuel Storage (Proceedings of the International Symposium on Management of Wastes From the LWR Fuel Cycle), CONF-76-0701, ERDA, Denver (1976)

[4] LANGHAAR, J.W., BELLAS, H.W., Water purification in fuel storage basins, DPSU 71-30-2A, E.E. duPont de Nemours and Company, April 1971.

[5] SCHNEIDMILLER, D., Concrete leaching studies for fuel storage basin recirculation, United Nuclear Industries, UNI-356, August 1976.

[6] KIBBEY, A.H., GODBEE, H.W., A critical review of solid waste practices at nuclear power plants, Oak Ridge National Laboratory, ORNL-4924, March 1974, 15.

TABLE I

RADIONUCLIDE CONCENTRATIONS IN FUEL STORAGE POOLS[(a)] μci/ml

Radionuclide	Current Generation Basins With High-Integrity Fuel	Early Generation Basins	Older ERDA Basins
^{3}H		1×10^{-3}	
^{54}Mn		1×10^{-5}	
^{58}Co	$5 \times 10^{-6} - 3 \times 10^{-5}$	$2 \times 10^{-5} - 1 \times 10^{-4}$	
^{60}Co	$5 \times 10^{-6} - 1 \times 10^{-4}$	$1 \times 10^{-5} - 1 \times 10^{-4}$	5×10^{-5}
^{90}Sr		2×10^{-5}	2×10^{-2}
^{131}I	1×10^{-7}	1×10^{-6}	
^{134}Cs	$1 \times 10^{-5} - 1 \times 10^{-4}$	3×10^{-4}	3×10^{-3}
^{137}Cs	$3 \times 10^{-5} - 1 \times 10^{-4}$	$5 \times 10^{-4} - 1 \times 10^{-3}$	3×10^{-2}
^{140}Ba		1×10^{-5}	
$^{144}CePr$			3×10^{-3}
Pu			2×10^{-5}
Total, μCi/ml	$1 \times 10^{-7} - 1 \times 10^{-3}$	$1 \times 10^{-3} - 1 \times 10^{-2}$	$1 \times 10^{-2} - 5 \times 10^{-1}$
Dose Rate[(b)], mrem/hr	<1	<5	5-50

(a) At equilibrium conditions; higher values are generally present immediately following reactor fuel discharge.

(b) Dose rates approach 40-100 mrem/hr on occasion during cleanup system upsets and following fuel discharges.

TABLE II

SURVEY OF SPENT-FUEL POOL CLEANUP SYSTEMS

SITE	FILTRATION	ION EXCHANGE	REGENERATE	REMARKS
BWRS				
Browns Ferry 1,2,3	Powdex	Powdex	No	Dual function
Dresden 1	D. Earth		No	
Dresden 2,3	Solka-floc	Mixed Bed	Maybe	Sludge present
Humboldt Bay	Cartridge	Mixed Bed	No	Sludge present
Monticello	Powdex	Mixed Bed	No	
Peach Bottom 2,3	Powdex	Powdex	No	Dual function
Shoreham 1		Mixed Bed	Maybe	
PWRS				
Connecticutt Yankee	Cartridge	Mixed Bed	No	
Ginna	Cartridge	Mixed Bed	No	Sludge present
Maine Yankee	Cartridge	Mixed Bed	No	
Palasides	Cartridge	Mixed Bed	No	
Pickering 1 & 2	None	Mixed Bed	No	Resin used as filter
Point Beach 1 & 2	Cartridge	Mixed Bed	No	
Rancho Seco	Cartridge	Mixed Bed	No	Pool empty
San Onofre 1	Cartridge	Mixed Bed	No	
Zion 1 & 2	Cartridge	Mixed Bed	No	
ERDA REACTORS				
Hanford KE, KW	Cartridge	Zeolite, M.B.	No	Sludge present
Savannah River K	Sand filter	Mixed Bed	Yes	Mobile IX, sludge
REPROCESSING				
Idaho Falls, ECF	D. Earth	Mixed Bed	No	Refrigeration
Idaho Falls, ICPP	D. Earth	Zeolite	Yes	Sand Filters now installed
Morris, MFRP	Comb. Precoat	Powdex,Zeolite	No	Dual function
Savannah River, RUBOF	D. Earth	Mixed Bed	Yes	Resin 8 yrs. old

X 10^{-1}

TABLE III

STORAGE BASIN PURIFICATION WASTE VOLUMES[a]

TYPE OF WASTE	TYPICAL VOLUMES
Filter Precoats (Powdex, Diatomaceous Earth Solka-floc)	2 - 5 $metre^3$/year[b] (20 $metre^3$ at one site)
Cartridge Filters	1 - 6 units/year[c] (20 units at one site)
Mixed Bed Resins	<1 - 4 $metre^3$/year[d]

NOTES

(a) Volumes are based on data from current generation commercial reactor sites. Total plant waste volumes are ∿200 $metre^3$/yr for BWR sites and ∿100 $metre^3$/yr for PWR sites.[6]

(b) Sludges are normally shipped in dewatered form, but are sometimes solidified in cement or urea-formaldehyde.

(c) Housings are normally discarded with the cartridges. When dose rates are high they are sometimes solidified in cement or urea-formaldehyde, thus increasing waste volume.

(d) Most sites are in the lower third of the range. Resins are normally shipped in dewatered form but can be solidified. Older ERDA sites not employing high-purity water usually regenerate the resins.

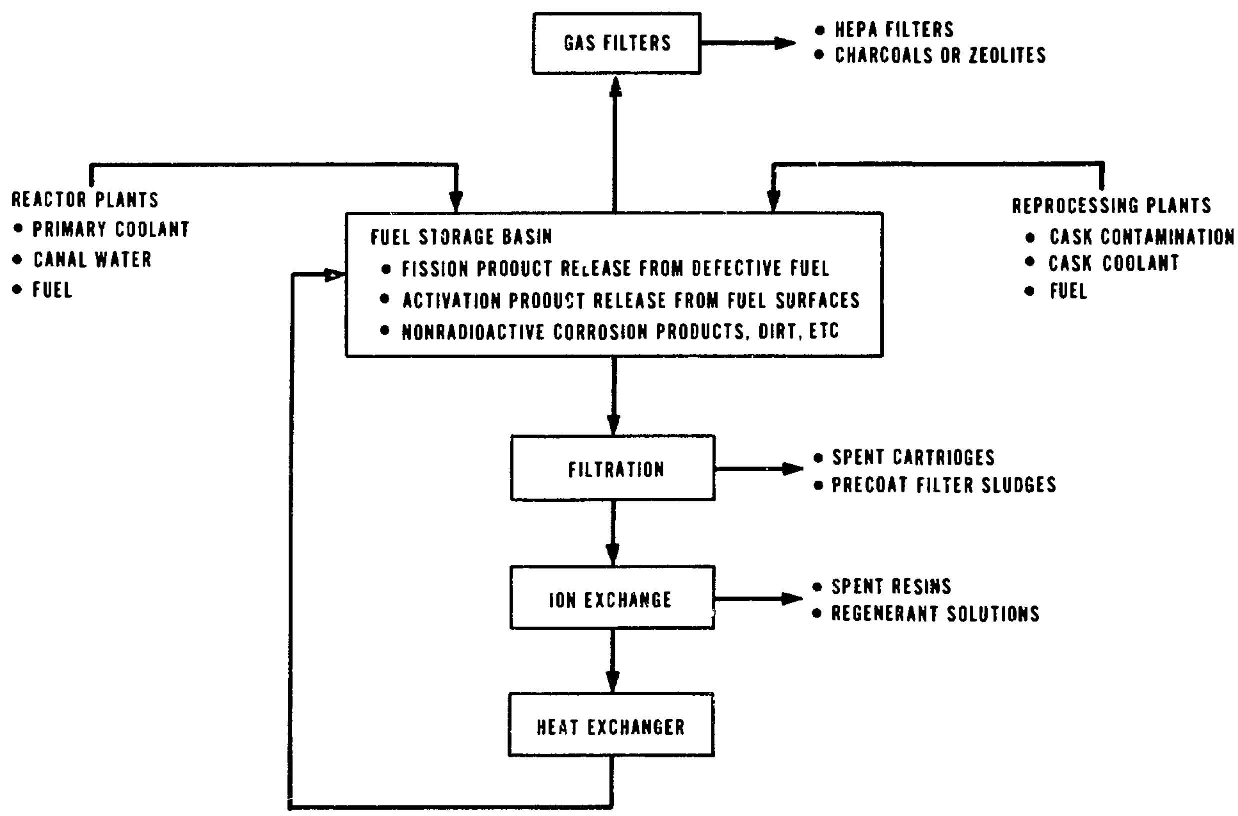

FIGURE 1

ROUTINE SOURCES OF WASTES FROM FUEL STORAGE BASINS