EXTENDED STORAGE OF LOW-LEVEL RADIOACTIVE WASTE: POTENTIAL PROBLEM AREAS*

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ABSTRACT

If a state or state compact does not have adequate disposal capacity for low-level radioactive waste (LLRW) by 1986 as required by the Low-Level Waste Policy Act, then extended storage of certain LLRW may be necessary. In this paper, the issue of extended storage of LLRW is addressed in order to determine for the Nuclear Regulatory Commission the areas of concern and the actions recommended to resolve these concerns. The focus is on the properties and behavior of the waste form and waste container. Storage alternatives are considered in order to characterize the likely storage environments for these wastes. The areas of concern about extended storage of LLRW are grouped into two categories:

1. Behavior of the waste form and/or container during storage, e.g., radiolytic gas generation, radiation-enhanced degradation of polymeric materials, and corrosion.

2. Effects of extended storage on the properties of the waste form and/or container that are important after storage (e.g., radiation-induced oxidative embrittlement of high-density polyethylene and the weakening of steel containers resulting from corrosion by the waste).

The additional information and actions required to address these concerns are discussed and, in particular, it is concluded that further information is needed on the rates of corrosion of container material by Class A wastes and on the apparent dose-rate dependence of radiolytic processes in Class B and C waste packages. Modifications to the guidance for solidified wastes and high-integrity containers in NRC's Technical Position on Waste Form are recommended.

INTRODUCTION

The Low-Level Waste Policy Act (PL 96-573, December 22, 1980) established state responsibility to provide disposal capacity for low-level radioactive waste (LLRW), and it was envisioned that all states would be self-sufficient in this respect by 1986. In addition, the Act encourages the formation of interstate compacts which (subject to approval by Congress) may refuse waste from outside their respective compact areas after January 1, 1986. Should a state or state compact not have adequate disposal capacity by 1986, then extended storage of waste may be required until disposal means are available. The waste may be stored for a period of several months to several years at the site of waste generation (e.g., on site at a nuclear power plant), at the disposal facility, or at a state or regional facility dedicated to such extended storage.

LLRW storage needs that may result from the unavailability of disposal capacity is a relatively new radwaste management problem in the United States. Most nuclear power plants were not designed with onsite LLRW storage capacity of extended duration since, in accord with the customary procedure, it was assumed that the LLRW would be shipped to a disposal site whenever a truckload had accumulated. Similarly, most non-fuel-cycle LLRW generators have operated under the assumption that the waste would be shipped for disposal rather than stored. Extended storage of LLRW has not been necessary at the disposal sites since disposal of the LLRW has usually occurred within a few days after receipt.

The U.S. Nuclear Reglatory Commission (NRC) has considered[1] two types of storage for LLRW at a nuclear power plant:

(1) interim contingency storage, for up to five years, and

(2) long-term storage, for over five years.

Due to current uncertainties regarding the availability of LLRW disposal capacity, the NRC is aware that extended storage of LLRW may be pursued by nuclear power plants and by other NRC licensees which generate LLRW. (In this paper the term "extended storage" is generally considered to be identical to "long-term storage.")

Extended storage of LLRW is a relatively new undertaking in the U.S. In order to develop guidance for the extended storage of LLRW by NRC licensees and to help ensure the continued protection of public health and safety, the NRC has contracted with Brookhaven National Laboratory (BNL) to address the issue of extended storage, focusing on the waste form and container, but also considering storage alternatives in order to establish the likely range of storage environments that the wastes would encounter. In this paper, the major points of BNL's draft report[2] are summarized. Additional work in this area is anticipated.

Storage Environment Characteristics

The behavior of radioactive wastes, of the binder materials in which they are *immobilized, and of the* container materials will be affected by the environment within the storage facilities. The environmental variables considered are length of storage time,

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temperature, humidity, potential for wetting of the container, and radiation field. Unfortunately, explicit information about these variables is generally not presented in descriptions of LLRW storage facilities. It should be noted that not all of the facilities described are intended to be extended storage facilities.

The potential storage time is a variable significantly impacted by factors other than technical considerations. The storage space available and the rate of waste production are, of course, important, but social, political, and economic factors that affect the availability of disposal sites for LLRW are likely to be the major considerations in determining the length of time for which storage of LLRW may be needed. For the purposes of this paper, a 5- to 15-year storage period is considered.

The temperature of the storage environment will vary only slightly in the large engineered structures for containerized radwaste that include heating, ventilation, and air conditioning (HVAC) systems in their design. A minimum temperature of 50°F (10°C) is explicitly mentioned by one utility for its LLRW storage facility.[3] An upper bound of 80°F (27°C) is conservatively estimated -- except during failure of the HVAC system, it is probably much lower -- and sharp variations, even within this temperature range, would not be expected. Since there is concern about drum corrosion due to humidity, the relative humidity is assumed to be below the critical value at which atmospheric corrosion becomes significant. For steel this value ranges from about 50% to 70%.[4] Temperature ranges for the indoor storage of resin waste in spent resin holding tanks at two other nuclear power plants range from 40°F to 90°F (4°C to 32°C) and 70°F to 100°F (21°C to 38°C).[5] At the other extreme, the wastes in a simple fenced-in concrete storage pad will be exposed to the outdoor temperature and the outdoor humidity, which over the course of a year in some locations may range from below -40°F (-40°C) to above 104°F (+40°C) and from 0% to 100%, respectively.[6]

For α and β radiation it may be assumed that, to a very good approximation, radiation emitted within the waste package is absorbed within the package. The γ-radiation field within a particular waste package will depend on the radiation emitted within the package itself and also on the γ radiation emitted by nearby packages. The γ radiation emitted within a particular package is generally not completely absorbed within the package itself. For example, at points of contact between two polyethylene containers loaded with γ emitters, the dose to the container material to a very good approximation will be the sum of the doses to those points for each of the two containers in isolation, i.e., when considering the dose to waste packages stored in proximity to one another, the γ-radiation field intensities of the individual packages should be superimposed. The dose to the contents of a waste package from the adjacent waste packages in a closely packed stacked array of such packages may be conservatively estimated by replacing the individual waste packages by an infinite medium. For example, the γ-ray dose to the contents of a stacked 55-gallon drum may be conservatively estimated by tripling the γ-ray dose to a 55-gallon drum in isolation. (It is assumed in making this estimate that all the drums in the stacked array contain the same concentrations of γ emitters.)[2]

The environmental variables discussed above are summarized in Table I for four categories of storage concepts. These four categories are large engineered structures, storage modules, shielded casks, and unshielded facilities. From the table, it may be seen that, based on the environmental variables, there are really only two important categories: large engineered structures and all other storage facilities. Some degree of environmental control is generally provided in the large engineered structures by means of a HVAC system.

TABLE I

Summary of Extended Storage Environments

	Large Engineered Structures	Storage Modules	Shielded Casks	Unshielded Facilities
Temperature (°C)	Controlled 10 to 27	Uncontrolled ambient -40 to +40	Uncontrolled ambient -40 to +40	Uncontrolled ambient -40 to +40
Relative Humidity	Controlled <50%	0% to 100%	0% to 100%	0% to 100%
Potential for Wetting Container	Negligible	Moderate	Moderate	Moderate to very large
Radiation Field From Adjacent Packages	Potentially significant. Factor of up to 3[a]	Slight	Slight	Slight

[a] Conservative upper bound for ratio of dose to infinite medium (approximating stacked drums) to dose to 55-gallon drum for γ-radiation.

For the other three categories of storage concepts, there is generally no environmental control other than that provided by the module or cask walls. In the extreme case of some unshielded facilities, there may not be any protection for the containers from rain or snow. The temperature and humidity of the storage environment will be that of the ambient air and will depend on local climate and weather.

Waste and Waste Package Characteristics

The properties and behavior of low-level radioactive waste streams, solidification agents, and container materials are reviewed, with the emphasis on those characteristics important for addressing the effects of extended storage on the waste and waste package. Because of the varied nature of non-fuel-cycle wastes, generic waste stream descriptions are not possible. Concerns such as radiolytic gas generation, production of corrosive liquids, and biodegradation will be relevant to particular non-fuel-cycle wastes, but except in special cases no general accounts of properties and behavior are available. Despite this general lack of characterization, if the activity of a non-fuel-cycle waste is Class B or Class C, then stabilization, either by incorporation into a binder material or by use of a high integrity container (HIC) is required, and thus much of the discussion below will be applicable. Therefore the characterization of waste streams in this paper deals only with fuel-cycle wastes. An overview of the properties and behavior of the wastes, binders, and solidification agents is presented below.

Waste Streams: Ion-Exchange Resins

- Radionuclide loadings on spent ion-exchange resins vary, typical loadings at different reactors ranging from 0.1 to 30 Ci/ft^3 and maximum loadings from 0.3 to 60 Ci/ft^3. Dose rates to the resin for a loading of 10 Ci/ft^3 are estimated to range from 10^2 to 10^3 rad/h. Based on the guidance given to LLRW generators in the NRC Technical Position on Waste Form,[8] the accumulated dose to the resins should not exceed 10^8 rad.

- A variety of radiation effects have been identified which may be of significance for the storage of spent ion-exchange resins, especially if the 10^8 rad accumulated dose limit recommended in the Technical Position is exceeded. It should be noted that these radiation processes may be affected both quantitatively and qualitatively by the partial pressure of oxygen and by the dose rate.[9,10]
  - Irradiation of ion-exchange resins may produce and release chemically active substances that can adversely affect the binder and container materials.
  - Radiolytic generation of gases has been observed. The predominant gas is hydrogen, which may pose a flammability or explosion hazard under certain storage conditions. In addition, the generation of other gases, such as carbon dioxide, methane, and trimethylamine, as well as the uptake of oxygen have been reported as resulting from the irradiation of ion-exchange resins.
  - Irradiation of ion-exchange resins prior to their solidification in cement has been reported to improve the compressive strength and immersion resistance of the resulting waste forms.
- Agglomeration of and gas buildup in unsolidified ion-exchange resins during storage has been attributed to biodegradation.[5,7,11]

Waste Streams: Other LWR

- The radiolytic generation of gases (predominantly hydrogen) and of corrosive substances has been observed in cellulosic materials.[12-14]
- Because much of this waste consists of organic materials (e.g., cellulose) biodegradation is likely if the wastes have not been self-sterilized by radiation. However, specific information on the nature of the biodegradative products (e.g., gases, corrosive materials) and their effects, if any, on binder and container materials does not seem to be available.

Binder Materials: Cement

- Radiolytic gas generation has been observed in waste forms consisting of low-level waste solidified in cement. This generation of gas has been attributed to radiolysis of water in the cement. Once again, the gas is predominantly hydrogen.[15]
- The relative humidity of the atmosphere in which the cement is stored may affect the compressive strength and leaching characteristics of the cement waste form since the cement may still be curing during at least the early part of the extended storage period.[16]
- Freeze-thaw cycling can damage cements which contain sufficient amounts of freezable water, particularly if mitigative measures have not been taken (e.g., air entrainment or the use of additives).[17]
- Certain of the products of waste radiolysis, such as low-molecular weight organic acids, have been found to attack cement.[17]

Binder Materials: Bitumens

- Radiolytic generation of gas has been observed in bitumens.[18] Once again, the major component of the gas is hydrogen, which may pose a flammability or explosion hazard under certain storage conditions. The G-values for gas generation depend on dose rate and the presence of oxygen.
- Biodegradation of bitumens has been observed. There is some evidence that corrosive substances may be produced as a result of biodegradation of bitumens.[19]

Binder Materials: Thermosetting Organic Polymers

- Radiolytic gas generation has been observed from at least one thermosetting organic polymer, vinyl ester-styrene, but the details are proprietary. There does not appear to be any information on the radiolytic generation of corrosives from this category of binder materials.
- During short-term small-scale testing, a small amount (<0.4 volume percent) of a free liquid (pH=5) has been observed on the surface of waste forms consisting of simulated LWR waste streams solidified in vinyl ester-styrene. Thermal cycling of these small-scale waste forms increases the amount of this free liquid (to 1.3 volume percent).[20]

Container Materials: Carbon Steel

- The following generic types of corrosion are considered to be of concern in the degradation of steel LLRW containers during storage: uniform attack, localized attack (pitting and crevice corrosion), galvanic attack, dealloying attack, and cracking phenomena (stress corrosion cracking).[21]
- Corrosion by the atmosphere, generally in the form of uniform corrosion, results from the interaction of carbon steel container material with the atmosphere and depends on the temperature and the relative humidity. This is the familiar, if somewhat difficult-to-quantify type of corrosion which is commonly known as "rust". Rates of 0.1 to 0.5 mils per year (mpy) are reported for the atmospheric corrosion of steels in an industrial atmosphere; these values are ten-year averages with about half of the corrosion occurring during the first year.[4]
- Corrosion of carbon steel containers may occur as a result of chemical reactions with aggressive components of the waste.
  - Corrosion rates of 0.4 to 4 mpy have been reported for mild steel immersed in various simulated unsolidified LLRW.[10,22] (This is of relevance for carbon steel containers with Class A waste, which does not have to be solidified but only dewatered).
  - Corrosion of carbon steel embedded in solidified wastes has also been observed. It is minimal for steel embedded in cement.[22] Corrosion of metals in bitumen has been attributed to biodegradation.[19] A corrosion rate of about 0.01 mil/day is reported for mild steel embedded in waste

forms consisting of a chelating decontamination reagent solidified in vinyl ester-styrene.[23]

These corrosion rates can be used to estimate lifetimes for carbon steel drums containing these wastes (Table II).

TABLE II

Estimated Corrosion Lifetimes for 18-Gauge 55-Gallon Drums[a]

Waste Type	Measured Corrosion Rate for Carbon Steel (mils per year)	Estimated Drum Lifetime[b] (years)
BWR powdered resin waste[c]	<1	>50
BWR chemical regenerative waste[c]	<1	>50
Boric acid waste[c]	3 to 4	12 to 17
H^+ form cation resin (unirradiated)[d]	0.4	≈120

[a] Drum material is assumed to be carbon steel.
[b] Estimate is based on assumptions of uniform corrosion, 50-mil wall thickness, and complete corrosion of wall.
[c] Measured corrosion rate from Colombo and Neilson.[22]
[d] Measured corrosion rate from Swyler, Dodge and Dayal.[10]

Container Materials: Polyethylene

- High density polyethylene is resistant to attack by a large number of chemical reagents (at least in the absence of a radiation field).

- Irradiation of polyethylene under anoxic conditions promotes cross-linking to a greater extent than degradation. Irradiation in air produces radiolytic oxidation at the surface. This oxidized zone is believed to gradually penetrate into the bulk of the polyethylene material as the irradiation proceeds, eventually resulting in oxidative degradation of the material. In addition to the total dose, the dose rate is once again important in this radiolytic oxidative process, the rate of radiolytic oxidation apparently varying inversely with the dose rate. Furthermore, the rate-limiting step is thought to be an activated process, so that the rate of radiolytic oxidation will be temperature dependent.[24,25]

Potential Problem Areas

Potential problem areas for the extended storage of LLRW have been identified and are found to fall into two categories as follows:

1. Potential problems in the behavior of the waste, binder, and/or container material during storage:
    a. Radiolytic gas generation from waste, binder, or container material may result in:
        i. pressurization, causing damage to the waste form and/or container, and/or
        ii. a flammability or explosion hazard.
    b. Internal corrosion of shipping containers, whether of radiolytic or non-radiolytic origin, may result in the failure of the container during storage:
        i. by localized pitting attack, resulting in penetration of the container wall. (This is an important consideration for steel containers, especially carbon steel drums.)
        ii. by any of several attack mechanisms, resulting in loss of structural strength and eventually to collapse of stacked containers.
    c. Atmospheric corrosion of shipping containers, in particular, carbon steel containers, may result in failure of the containers during storage.
    d. Radiation-enhanced creep of polyethylene may result in the collapse of stacked containers during storage.
    e. Biodegradative production of gas and corrosives may result in pressurization and container failure, respectively, during storage.
2. Potential problems after storage because of degradation of the waste, binder, and/or container material during storage:
    a. As a result of corrosion of the shipping container during storage, the container may not meet DOT requirements for radwaste shipments and repackaging of the wastes for shipment may be necessary.
    b. Because of radiolytic cross-linking and radiolytic oxidative degradation of polyethylene, waste containers of this material may fail to meet the free drop and lifting load requirements for high integrity containers.
    c. Agglomeration of spent ion-exchange resins stored for extended periods in resin holding tanks, whether biodegradative or radiolytic in origin, may interfere with subsequent transfer and processing of the resins.
    d. An extended storage period may affect the leaching properties, mechanical strength, and immersion resistance of waste forms, especially those with cement binders.
    e. Temperature fluctuations (freeze-thaw cycling) may result in the loss of monolithic physical integrity of cement waste forms.
    f. The radiolytic production of corrosives from the wastes may result in the loss of monolithic physical integrity of waste forms, especially cement.

Conclusions: Additional Information Needs and Recommendations for Further Work

The general needs for additional information may be summarized in the following recommendations for further work:

- The corrosion rates of carbon steel immersed in a variety of dewatered low-level wastes need to be determined in order to estimate the storage lifetimes of containers of Class A waste. These rates may be determined in the absence of radiation since non-radiolytic corrosion processes are thought to predominate in Class A wastes.

- The corrosion rates of carbon steel embedded in solidified low-level wastes need to be further investigated in order to estimate the storage lifetimes of steel containers with solidified wastes. a) The effect of irradiation on these corrosion rates should be determined. The radiation field should, to the extent practicable, simulate the expected radiation environment. The partial pressures of hydrogen and oxygen expected inside containers should also be simulated as closely as possible. Acceleration of testing by increasing the dose rate should take into account the effects of dose rate and radiation-enhanced oxidation to the extent practicable. b) The biodegradative contribution, if any, to corrosion of steel in solidified wastes should be determined.

- Further work on the rates of radiolytic gas generation in binder materials is needed in order to assess the potential for flammability or explosion. Radiolytic gas generation is not anticipated to be a problem in properly designed cement binder applications. Data on such gas generation, however, seem to be particularly scarce for the thermosetting organic polymer binders. Acceleration of testing by increasing the dose rate should take into account dose-rate effects (including "apparent" dose-rate effects such as the time needed for diffusion of the radiolytically generated gas through the binder matrix) to the extent practicable.

- The rates of biodegradative gas generation in representative dewatered Class A wastes and of radiolytic gas generation in representative dewatered Class B and C wastes need to be determined in order to assess the potential for pressurization of or the release of flammable and explosive gases from the waste containers. Acceleration of testing by increasing the dose rate should take into account dose-rate effects and radiation-enhanced oxidation to the extent practicable.

- The rates of radiolytic degradation of polyethylene high-integrity container materials need to be determined. The effect of dose rate and the partial pressure of oxygen on radiolytic degradation of polyethylene should be considered, especially if testing is accelerated by increasing the dose rate. Also, since the rate-limiting step of the radiolytically enhanced oxidation reaction is believed to be a thermally activated process, the effect of temperature on the rate of this reaction may need to be investigated.

- The resistance of polyethylene to corrosion by representative Class B and C wastes in a radiation field needs to be established, even if such wastes are compatible with polyethylene in the absence of a radiation field. The emphasis should be on any such wastes containing oxidizing components. Acceleration of testing by increasing the dose rate should take into acccount dose-rate effects to the extent practicable.

- The effects of the temperature and the humidity of the extended storage environment on the curing and thus on the mechanical strength, immersion resistance, and leaching properties of cement waste forms need further investigation.

- The above considerations regarding corrosion and oxidative degradation of carbon steel and polyethylene will need to be extended to other LLRW container materials as they come into use. At present, high-integrity containers fabricated from a duplex stainless steel alloy, Ferralium 255, and from a fiberglass-reinforced plastic are being considered by vendors.[26,27]

In the Technical Position Paper (TP) on Waste Form,[8] the NRC has provided guidance to waste generators on test methods and results acceptable to the NRC staff for implementing the 10 CFR Part 61 waste form requirements. Because large scale extended storage of LLRW would represent a relatively new radwaste management activity in the United States, it is not explicitly addressed in the TP. [The effects of a storage period--but not explicitly an extended storage period of greater than five years--are considered in a few places in the TP. For example, it is specified that the thermal loads from storage as well as from processing, transportation, and disposal should be considered in the design of high-integrity containers (HICs). Also,storage is identified as one of several factors to be considered in a quality assurance program for HICs.] Modifications and additions to the TP are considered appropriate in order to take account of the potential problems which may need to be addressed if extended storage of LLRW is pursued because of inadequate disposal capacity. For example, it is recommended that the following concerns (which are somewhat overlapping) be addressed in the TP:

- The effect of the storage environment on the behavior of the waste, waste form, or container material during and after storage.

- The effect of dose rate on the results of radiation testing of wastes, waste forms and container materials.

- The accumulation of combustible or otherwise hazardous gases.

- The load on a waste container resulting from the waste stacked above it.

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