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Extended Storage of Low-Level Radioactive Waste: Potential Problem Areas

Prepared by B. Siskind, D. R. Dougherty, D. R. MacKenzie

Brookhaven National Laboratory

Prepared for
U.S. Nuclear Regulatory Commission

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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 report, extended storage of LLRW is considered in order to determine for the Nuclear Regulatory Commission areas of concern and actions recommended to resolve these concerns. The focus is on the properties and performance 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. Performance 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 embrittlement of high-density polyethylene and the weakening of steel containers resulting from corrosion).

A discussion is given of additional information and actions required to address these concerns.

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

1. 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. In addition, the Act encourages the formation of interstate compacts which (subject to approval by Congress) may refuse LLRW from outside their respective compact areas after January 1, 1986. Amendments to the Act are now before Congress, but the availability of disposal capacity for LLRW after January 1, 1986, remains uncertain. 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.

On-site LLRW storage needs arising from the unavailability of disposal capacity constitute a relatively new radwaste management problem in the United States. Most nuclear power plants were not designed with on-site LLRW storage capacity of extended duration since 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 site since disposal of the LLRW has usually occurred within a few days after receipt.

The U.S. Nuclear Regulatory Commission (NRC) has provided guidance for LLRW storage practices at nuclear reactor sites in Generic Letter 81-38.* In this document the NRC has considered two phases or time scales for storage of LLRW at nuclear power plants:

1. interim contingency storage, for up to 5 years, and

2. long-term storage, for over 5 years.

Because of current uncertainties regarding the availability of LLRW disposal capacity, the NRC is aware that extended storage of LLRW may be pursued by nuclear power plant licensees and by other NRC licensees who generate LLRW. (In this report, the term "extended storage" is generally considered to include both "interim contingency storage" and "long-term storage.")

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*Generic Letter 81-38 is reproduced as Appendix A.

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 of LLRW with special attention to 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. The dual objectives of this study are (1) to provide practical technical assessments for NRC to consider in evaluating specific proposals for extended storage and (2) to help ensure adequate consideration by NRC, Agreement States, and licensees of potential problems that may arise from existing or proposed extended storage practices.

The characteristics of the storage alternatives available for LLRW are reviewed in Section 2. Several different approaches to the design of extended storage facilities considered by utilities and nuclear service companies are included in this review, but, since much of the work on extended storage has been done abroad, non-U.S. storage concepts are also discussed. The storage environments are characterized to the extent possible in terms of parameters such as temperature ranges, moisture, radiation field, and expected storage time.

The properties and behavior of generic low-level waste stream types (e.g., ion-exchange resins, aqueous concentrates, filter sludges), radwaste binder materials (e.g., cement, bitumen), and container materials (e.g., polyethylene, carbon steel) are briefly reviewed in Section 3. This review emphasizes those characteristics deemed important for predicting the behavior of the waste forms and containers during storage and for assessing the effect of extended storage on waste form stability and container integrity after disposal.

The effects of storage conditions on the LLRW are evaluated in Section 4. The potential problem areas are identified and grouped into two categories for the purposes of discussion: 1) behavior of the waste form and/or container during storage and 2) effects of extended storage on properties of the waste form and/or container that are important after storage.

A summary of the report is given in Section 5. Technical details (absorbed dose and radiolysis calculations) and relevant legal documents and NRC staff technical positions are included as appendices.

2. STORAGE FACILITIES

In this section an overview of the alternatives for the extended or interim storage of low-level radioactive waste (LLRW) is presented, with emphasis on those alternatives which have been implemented by or proposed for nuclear power plant utilities in the U.S., although storage concepts proposed or implemented elsewhere are also considered. Since reduction of radioactivity by decay has been until recently a primary motive for storage of LLRW, some introductory comments comparing storage for decay with extended storage are presented in Section 2.1. This is followed in Section 2.2 by an account of extended storage concepts developed in the U.S. and abroad. Storage environments are characterized in Section 2.3.

2.1 Introductory Comments

There are several reasons for storing LLRW. Until recently the usual reason has been to allow for radioactive decay. Storage for decay is widely practiced by hospitals and universities. The possible unavailability of adequate disposal capacity for LLRW provides a second reason for storage of these wastes. An additional reason for extended storage is that existing disposal may become temporarily unavailable because of problems such as unavailability of transportation services. Storage is also practiced to consolidate waste for efficient shipment. Extended storage is defined here as the holding of LLRW for a specified period of time at or away from the waste generator's site before burial at a licensed disposal site. Some general aspects of these two types of storage and of the interim storage strategies (at or away from site of waste generation) are discussed in these introductory comments.

2.1.1 Storage for Decay[1]

The practicability of storage for decay requires that there be defined concentrations or quantities of radioactive isotopes below which the waste is considered to be suitable for disposal as nonradioactive waste. Such standards are available for liquid and gaseous effluents in 10 CFR Part 20. However, no such standards have been set for solid wastes.

A rule of thumb has been that, after a decay time equivalent to approximately 10 half-lives, the initial activities of "normal" industrial and medical radiochemicals have decayed sufficiently for environmental discharge (liquids and gases) under the restrictions of 10 CFR Part 20. This, plus the approximately one-year time limit normally practicable for storage for decay, limits consideration to isotopes with half-lives less than about 40 days as candidates for storage for decay. Radionuclides with half-lives between about 40 and 80 days are also potential candidates for storage for decay if additional precautions are considered, e.g., administrative inventory, shielding, security, final activity calculations, and final disposition. Many of the isotopes used in industrial, educational, and medical applications have less than 40-day half-lives, and storage for decay has been a standard practice. However, in the nuclear power industry, radwaste generally contains a mix of isotopes, some with long half-lives, and storage for decay has not been a

viable consideration. The NRC has provided guidance to medical, academic, and industrial licensees on storage for decay. See Appendix B.

2.1.2 Extended Storage

Extended storage of radwaste is likely to become necessary if access to disposal facilities becomes unavailable for a time. In this report, the time frame for extended storage has been conservatively estimated to be as long as 15 years. The following introductory comments on extended storage are from a recent assessment of LLRW management.[1]

Requirements for extended storage include:[1]

- Adequate space.
- Capability for volume reduction or waste solidification.
- Controlled access.
- Monitoring capability.

These requirements are more likely to be met at nuclear power stations than at industrial, educational, and medical facilities. For these and other small-volume generators, regional radwaste management centers are a possibility. Solid waste forms, which would also meet the criteria for final, permanent disposal, are the most desirable waste form candidates for extended storage.

Some of the probable consequences of extended storage include higher costs for waste management, increased occupational radiation exposure, and incentive to volume reduction. The higher costs of extended storage before final disposal could be alleviated somewhat by enhanced volume reduction and by storage for decay of some solid radwaste which could then be released for environmental disposal rather than to a radwaste facility. Currently, all solid radwaste must be disposed of in radwaste disposal facilities. Radwaste disposal site personnel were said to receive generally higher average doses than other radiation workers. The occupational radiation exposure of workers in an extended storage facility may also be higher than that for other radiation workers.[1] As seen in the following discussion, however, estimates of occupational exposure from the operation of extended storage facilities indicate that such exposure constitutes only a small portion of the total occupational exposure at nuclear power plants.

2.1.3 Occupational Exposure From Extended Storage Operations

In a study sponsored by the Atomic Industrial Forum, estimates have been cited by Martineit et al. for the annual exposure received by nuclear power plant personnel responsible for loading radwaste onto transport vehicles.[2] It is assumed in this study that these exposures would be received whether or not the radwaste is stored on site and also that the exposures received while unloading the waste from the transport vehicle at the on-site storage facility

equal those received while loading the vehicle. Only the latter contribute to the increase in plant personnel exposure from on-site operations. At the end of a 40-year storage period, it is assumed that the waste is removed from the storage facility, surveyed, and prepared for transport. Accounting for decay during storage and during the time required to unload the waste from the storage facility, the total expected occupational exposure for the complete removal of 40 years' accumulation of solidified radwaste has also been estimated (Table 2.1). The occupational exposure resulting from the implementation of extended storage could be increased above these values if it should become necessary to repackage the waste because of degradation or failure of the waste package during the storage period.

Table 2.1

Increased Radiation Exposure to Plant Personnel From Extended Storage Operations[a]

Scenario for Volume Reduction and Waste Solidification	Exposure (man-rem)	
	BWR	PWR
No Volume Reduction, Waste Solidified (Except Trash)		
Annual radiation exposure during storage operations	19.8	22.0
Exposure during removal of 40 years' accumulation of stored radwaste	206.	229.
Volume Reduction, Waste Solidified		
Annual radiation exposure during storage operations	19.8	22.0
Exposure during removal of 40 years' accumulation of stored radwaste	301.	334.
Volume Reduction, No Solidification		
Annual radiation exposure during storage operations	35.2	33.6
Exposure during removal of 40 years' accumulation of stored radwaste	649.	500.

[a]Information from Reference 2.

In a site-specific safety analysis(3) for a large engineered structure for interim storage of LLRW, an assessment has been made of the increased occupational exposures to on-site personnel from the activities associated with interim storage of LLRW at the Susquehanna Steam Electric Station (SSES). The SSES consists of two BWR units, rated at about 1000 MWe each and having an estimated combined LLRW production rate of 60,000 ft^3/yr. Only operations associated with the interim storage facility were considered in making the assessment; normal waste-handling operations independent of interim

storage were not included. The estimates of increased exposure associated with the SSES storage operations, presented in Table 2.2, are considerably lower than the AIF estimates given in Table 2.1, especially when normalized to a man-rem-per-MWe basis. The disparity between the two sets of estimates may be partly due to differences in the proposed storage periods. In the AIF scenario, the total LLRW accumulated over a 40-year operating period is removed from storage at the end of 40 years. In the SSES safety analysis, the maximum amount of LLRW stored is that generated by four reactor-years of operation per unit. In addition, the SSES safety analysis contains explicit mention of the minimization of exposure to on-site workers by use of concrete shielding, shielded loading equipment, minimization of the number of operating personnel, and controlled access to the storage facility. To put figures in Table 2.1 and 2.2 into perspective, they should be compared with the annual collective doses by work function at light water reactors. Some of these latter data from 1981 are presented in Tables 2.3 and 2.4. As may be seen from the data in the tables, the occupational doses associated with storage of LLRW are only a small portion of the total occupational dose at commercial nuclear power plants.

Table 2.2

Increased Occupational Exposure to On-Site Personnel From the Interim Storage of LLRW at the Susquehanna Steam Electric Station[a]

Category	Annual Estimated man-Rem
Transporting waste containers to the storage facility	0.6
Loading solidified waste containers into vaults	0.5
Inspection of stored solidified waste containers	0.02
Solidified waste containers off-loading	0.4
Trash handling exposures	
Trash container loading	1.3
Trash container off-loading	1.3
Total	4.1

[a]Information from Reference 3.

Table 2.3

Dose Information Reported by
U.S. Commercial Light Water Reactors for 1981[a]

	Number of Reactors Reporting	Annual Collective Doses (man-rem)	Gross MW-yr Electricity Generated	Average Man-rem per 1000 MWe[b]
BWR	26	25,471	10,899	2300
PWR	44	28,671	20,552	1400

[a]Information from NUREG-0713, Vol. 3, "Occupational Radiation Exposure at Nuclear Power Reactors," 1981.(4)
[b]Rounded-off to nearest 100.

Table 2.4

Percentages of Annual Collective Dose at
LWRs by Work Function for 1981[a]

Work Function	% of Total Man-Rem per Function	
	BWR	PWR
Reactor operation and surveillance	7.5	9.8
Routine maintenance	42.2	28.7
In-service inspections	3.7	6.5
Special maintenance	33.1	44.9
Waste processing	11.0	3.2
Refueling	2.5	7.0

[a]Information from Reference 4.

2.1.4 Storage Strategies

In devising strategies for the extended storage of LLRW, it should be noted that such storage may be implemented at the site of waste generation, at some interim holding facility (e.g., a state or regional facility or a facility operated by a commercial waste broker) away from the site of waste generation, or at the disposal site. Other distinguishing elements among possible storage strategies are the timing and location of the waste stabilization or immobilization operations. Stabilization at the waste generation site may be

performed before, during, or after the extended storage period. Packaging for disposal prior to storage is assumed in the NRC's guidance to power reactor licensees.* The effects of storage on the waste are important considerations for subsequent handling, shipment, and disposal operations. (See Section 4.1, below.) The possibility of two or more periods of extended storage (e.g., at the waste generation site and then at an interim facility) may have to be considered in the event adequate disposal capacity remains unavailable. Storage of containers of waste in large structures and direct storage in transport containers are the concepts most applicable to the three basic storage strategies. The various storage concepts are discussed in the next section.

2.2 Approaches to Extended Storage

Concepts and designs for facilities for the extended storage of LLRW have been considered both in the U.S. and abroad. In the following discussion an overview is given of some of the concepts which have been developed and of storage facilities planned or under construction. First, an account is given of extended storage concepts developed in one of the early (1978) U.S. studies of extended storage of LLRW. The conceptual designs for storage facilities offered by several engineering and nuclear service vendors are then briefly described. Following this discussion, the extended storage plans of particular power plant operators which were found in the open literature are outlined. Finally, a brief survey of LLRW storage outside the U.S. is presented, once again based on information from the open literature. When available, information on the environmental parameters associated with these alternatives is given.

2.2.1 Early Approaches to Extended Storage in the U.S. -- The AIF Study

Nuclear power plants were generally not designed with significant on-site storage capacity for LLRW. It was assumed that the LLRW would be shipped whenever a truckload had accumulated,[5] although spent ion-exchange resins have been stored on-site in unsolidified form for as long as one year and can be stored for 3 to 4 years in spent resin holding tanks.[6] By 1978, however, three of the commercial shallow land burial (SLB) sites had been closed and the availability of the remaining three SLB sites was increasingly subject to restrictions and uncertainty. It, therefore, became necessary to consider the on-site retention of LLRW at nuclear power plants, at least on an interim basis. In response to this situation, the Atomic Industrial Forum (AIF) commissioned an investigation of the technical requirements for the economics of on-site management alternatives for LLRW; the results of this study appeared in 1978 as a report hereinafter referred to as the AIF study or the AIF report.[2] Design criteria and conceptual designs for several kinds of radwaste storage facilities are presented in the AIF study and possible

*U.S. Nuclear Regulatory Commission, "Storage of Low-Level Radioactive Wastes at Power Reactor Sites (Generic letter 81-38), "Enclosure: Guidance Document, November 10, 1981. Reproduced in Appendix A to this report.

problems with the radwaste containers are also considered. Several generic storage modes are discussed, i.e., unsolidified radwaste stored in bulk, contaminated compacted trash, and containerized radwaste. It should be noted that the discussion of radwaste form and radwaste containers in the AIF study predates the publication of 10 CFR Part 61, the licensing requirements for the land disposal of radioactive waste, and the subsequent guidance for waste form and high integrity containers given in the NRC's May 1983 Technical Position on Waste Form.

The design criteria considered in the AIF report include such items as seismic resistance, dose-rate requirements, wind and snow loading, selective retrievability of individual storage containers, and monitoring and control of radionuclide releases, but in the present context of waste form, waste container, and storage environment, the following criteria from the AIF study are of particular concern:

- Design Life of the Facility - Two distinct lifetimes are considered for a generic storage facility, namely, 40 years (the projected life of the power plant) and 165 years. (No explanation is given for the 165-year figure.)

- Segregation of Radwaste During Storage -

  a. Containerized Storage - Separation of containers of different metallic materials may be necessary because of the possibility of galvanic corrosion when different metals are in contact, particularly if the contacting surfaces are wet.

  b. Bulk Storage - Wet wastes such as dewatered resins must not come in contact with wastes that should be kept dry such as incinerator ash and dried liquid residues.

In the AIF's conceptual design for bulk storage, the storage facility consists of rectangular silos constructed of 2- to 3-ft-thick reinforced concrete lined with stainless steel. A sheet metal enclosure provides protection against severe weather conditions. Support systems include the equipment for placing the waste into or removing it from storage (e.g., shielded piping), as well as lighting and ventilation systems. Heating or cooling systems are not indicated in the conceptual design. Two sizes of storage silos are considered which have approximate internal volumes of 11,000 ft^3 (external dimensions 28 x 28 x 25 ft high) and 45,000 ft^3 (external dimensions of 51 x 51 x 27 ft high). Two advantages are given in the AIF study for bulk storage of nonmonolithic radwaste, viz., (1) the cost of storage is reduced and (2) optimum packing efficiency is achieved. Potential problem areas identified in the study are (1) wetting of the dry waste during transport to storage, as well as during storage and (2) difficulties in removing the wet waste from storage.

A conceptual design for a storage facility for compacted contaminated trash is also discussed in the AIF report. This facility is expected to serve two basic functions viz., (1) to provide the shielding to reduce the generally

low dose rates (≈20 mrem/h at contact) of compacted trash to acceptable levels outside the building and (2) to protect the storage containers from the weather. Facility operation consists of delivering the waste, packaged in 55-gallon drums, to the entrance of the building for transfer by fork lift. The support systems consist of lighting, exhaust fans, and, if severe winters are likely, a heating system. (No specific temperature limits are mentioned.) The steel frame and block wall building measures 60 x 83.5 x 21 ft high internally with 1-ft-thick walls and a total internal volume of 105,210 ft^3. About 3 ft of space at the top of the inside volume is reserved for support system connections, allowing about 56,000 ft^3 of waste to be stored in stocked 55-gallon drums. A similar but smaller (54,000 ft^3) in-ground concrete storage facility is also discussed in which about 27,500 ft^3 of compacted trash packaged in 55-gallon drums could be stored.

A storage concept for containerized radwaste which requires remote handling by an overhead bridge crane is also presented in the AIF study. It is noted in this study that solidification of the containerized radwaste prior to storage would preclude the use of cheaper, more efficient solidification techniques which may be developed during the period of storage, but since the same containers can be used for both waste forms, the same storage facility concept is used for both monolithically solidified and unsolidified wastes. The storage facility consists of one or two storage modules with 100,000 to 600,000 ft^3 of total storage volume enclosed by 2.5- to 3-ft-thick walls. The stored containers will be stacked. A ventilation system is considered necessary under worst-case conditions to remove hydrogen gas resulting from the radiolytic decomposition of solidified radwaste. No information is presented about temperature control systems. The containers used for the interim storage of radwaste are also discussed in the AIF study. These factors are considered important for maintaining the structural integrity of these containers:

(1) <u>The Loads Resulting From Stacking of the Containers</u>

It is found that 55-gallon drums may be stacked in layers of seven to ten depending on the wall thickness of the drums (18 gauge or 16 gauge) and their orientation (horizontal or vertical). For example, it is stated in the AIF study that a vertical 55-gallon drum (DOT Specification 17H, 18 gauge) can withstand a load of 13,000 pounds before it starts to fail. It is further stated that this load will not be exceeded for waste with a density of 150 lb/ft^3 in drums stacked eight high. The maximum weight of seven 55-gallon drums filled with 150 lb/ft^3 waste totals about 7720 lb. (The maximum value of the density for cement-solidified LLRW is about 150 lb/ft^3 or 2.4 g/cm^3. Also, the "authorized" gross weight for a 17H 55-gallon drum is only 840 lbs.*)

*The "authorized" gross weight appears to be based on the 4-ft free drop test. See "Certification of ERDA Contractors' Packaging With Respect to Compliance With DOT Specification 7A Performance Requirements." Phase II Summary Report, MLM-2228, June 12, 1975.

(2) Corrosion of the Container Materials

To prevent corrosion on the inner surface of the container by wet wastes, it is recommended in the study that the containers be either lined with a corrosion-resistant material or fabricated of stainless steel. Discussions of the corrosion of the inner surfaces of containers by wet wastes and of the mitigation of such corrosion by the use of coatings or liners of corrosion-resistance materials are given below in Section 3.3.

(3) Gas Evolution From Radiolysis or Biodegradation of the Radwaste

It is suggested that overpressurization of containers be avoided by including venting or a pressure relief valve in the container design. Discussions of radiolytic gas generation from waste and binder materials are given below in Sections 3.1 and 3.2, respectively. Calculations of radiolytic gas generation are presented in Appendix A. (The vented gas should be monitored for radionuclides and explosive mixtures.)

Also, in the event radwaste is stored in nonmonolithic form for extended periods prior to solidification for permanent disposal, it is necessary to ascertain whether the radwaste has undergone changes that will affect either the ease of retrieval for solidification or the solidification process itself. It is noted in the AIF study that dried residues of liquid radwaste and the ash from radwaste incineration are hygroscopic and will absorb large quantities of water, thus causing the waste to swell. It may then prove difficult to remove the waste from the containers for solidification. It is also noted that spent resins have been known to undergo physical and/or chemical changes to a form that is not amenable to slurrying: such "bridging" or "caking" of the resins has been attributed to the growth of microorganisms in the resin. No resolution was suggested for these problems.

2.2.2 More Recent Approaches to Extended Storage

Several vendors have presented conceptual designs for an engineered long-term on-site storage facility which are similar to the storage concept for containerized radwaste described in the AIF study. For example, Chauvin and Jurkin[7] have described a conceptual design developed by Sargent and Lundy for a facility to store solidified containerized LLRW. The nominal storage capacity of the building is 8000 55-gallon drums, which (according to these authors) is equivalent to 8 to 10 years' LLRW production from a typical two-unit power plant equipped with volume reduction (VR) by drying and incineration, or alternatively, to 2 years' production without VR. (The "typical" two-unit plant is not defined.) The facility is an above-grade reinforced concrete structure, measuring about 133- x 69-ft and is 43 ft high with the drum storage area occupying about 103 ft of the length of the structure (or about 305,000 ft^3). As in the AIF design, the drums are handled remotely by an overhead bridge crane. The drums are stored stacked in up to eight layers with steel grating between the layers for stabilization. A heating,

ventilation, and air conditioning (HVAC) system is explicitly mentioned, but no information is given about the temperature in the storage area. The air pressure inside the building is maintained slightly below outside pressure, and all exhaust ventilation is filtered and monitored for radioactivity.

Another design with some similarities to the AIF containerized radwaste storage concept has been developed by Cygna Energy Services and is described by Gardner et al.[8] The containers are a mixture of radwaste liners and 55-gallon drums or 100-ft^3 metal boxes for dry active waste. One interesting feature is the manner in which wastes are segregated by activity within the storage bay area. Liners with a contact dose rate of less than 100 mr/h are placed at the perimeter of the storage bay area, but higher-activity containers are located either in the interior of this area or in aboveground concrete vaults also within this area. Once again, a HVAC system is included in the design, but no information on temperature is presented. It should be noted that some of the waste packages could possibly be subject to accelerated degradation because of the increased radiation field from the radionuclides in neighboring packages. As discussed further in Section 2.3, Storage Environments, gamma radiation at a point is essentially additive when multiple sources are present.

A conceptual design for an integrated VR, solidification, and interim storage facility has been developed by Bechtel.[9] Relatively little information is given about the storage facility *per se*, but its size (approximately 200,000 ft^3) is intended to accommodate a minimum of 2 years' production of LLRW from a "typical" two-unit LWR power plant. The storage area is divided into two modules, one for "low-level" drum storage and the other for "intermediate-level" drum storage. (The criteria for "low" and "intermediate-level" wastes are not given.) A HVAC system is incorporated into this design as well, but temperature conditions are not discussed.

The above concepts for the on-site storage of containerized LLRW all share certain features. They are all essentially large concrete-shielded facilities with storage volume for several years of containerized LLRW production by the power plant unit(s). Provisions for such accessories as cranes, loading docks, HVAC systems, off-gas systems, and decontamination facilities are included in the design. On-site storage in modular storage casks is an alternative approach. On-site storage containers (OSSC), offered by ATCOR Engineered Systems, Inc., and described by Dufrane,[10] are steel-reinforced concrete structures similar in size and shielding characteristics to standard casks or vans used to ship LLRW. The vendor suggests that they be used bothto store and to ship the radwaste liners and/or 55-gallon drums in which LLRW is packaged at any particular reactor. Similar approaches to on-site interim storage of LLRW, i.e., storing the LLRW in on-site concrete storage containers that can also be used as the off-site transportation cask and high integrity container, have been developed by Nuclear Packaging, Inc,[11] and by Westinghouse Hittman Nuclear Inc. Steel casks for on-site shielded storage and disposal have been proposed, also by Westinghouse; each cask can hold several 55-gallon drums -- 3, 7, or 14 drums in the currently proposed cask sizes -- or one large container.[11]

Because of the varied physical and radiological properties of industrial LLRW it is difficult to generalize about the characteristics of an extended storage facility for such wastes. One industrial waste storage facility currently used for relatively short-term storage, described in a New York State study,[12] consists of a reinforced concrete building with shielded storage cells and a ventilation system. The limited 750-ft^3 storage capacity will hold 100 55-gallon drums for a few months, after which the waste must be shipped for disposal.

The basic design characteristics of a building developed for the storage of wastes with low volume and low specific activities typical of institutional wastes is also given in the New York State study.[12]. It is a simple concrete building with 12-inch-thick walls and a ventilation system. Drums and/or other containers are stored in 1300 ft^3 of storage volume. As in the industrial LLRW storage facility, the storage capacity is limited and continued operation of the facility depends on the availability of disposal capacity.

2.2.3 Specific Utility Storage Plans

The Electric Power Research Institute (EPRI) has sponsored a survey of the actions and plans of the nuclear utilities regarding on-site storage of LLRW and has reported[13] information valid as of 1983. The status of these plans and actions as given in the EPRI report is summarized in Tables 2.5 and 2.6.

In the EPRI report, the term "reinforced concrete structure" refers to a building which provides shielding as well as some degree of environmental control. The term "prefab structure" means a building with some degree of weather protection but not very much shielding. "Bunkers" are described as storage facilities set into the ground, with removable covers. No information is available on temperature control, but presumably the prefab structures and bunkers are less likely to be serviced by HVAC systems.

Table 2.5

On-Site Storage Facility Actions and Plans of Nuclear Utilities[a]

	PWR	BWR	Total
No Expansion Planned	13	5	18
Expansion Planned	17	9	26
Expansion Completed	6	4	10
No Information	15	8	23
	51	26	77

[a]Information from EPRI NP-3617.[13]

Table 2.6

Construction Status of
On-Site Storage Facilities at Reactor Sites[a]

	Completed	Not Completed
Reinforced Concrete Structures	8	18
Prefab Structures (concrete or metal panels)	1	5
Bunkers	2	0

[a]Information from EPRI NP-3617.(13)

It is not clear whether the reinforced concrete structures of the EPRI survey are necessarily large engineered storage structures as described above in Section 2.2.2, but simple butler-type buildings would seem to form a subset of the EPRI survey's second category of on-site storage facilities, i.e., prefab structures. Only 6 of the 34 on-site storage facilities of Table 1.2 are prefab structures; 2 are bunkers, and the remaining 26 are reinforced concrete structures. The results of the EPRI survey suggest that fewer than half the on-site storage facilities planned, under construction, or existing are simple butler-type buildings, at least as of the end of 1983.* In the range of existing and planned storage facilities and associated environments, simple butler-type buildings and large engineered structures fall near or at the ends of a "spectrum" of storage facility concepts. Specific examples of such facilities are described below.

TVA(14)

Because of restrictions on the amount of LLRW accepted for disposal at the Barnwell facility, the Tennessee Valley Authority plans to manage the LLRW generated at Browns Ferry, Sequoyah, and Watts Bar Nuclear Plants by constructing on-site storage facilities in possible conjunction with VR facilities. Temporary storage of some LLRW has been provided at Browns Ferry and Sequoyah. Compacted dry active waste has been stored temporarily in an existing cable warehouse, which consists of a concrete pad, timber roof, and support beams without walls. The remainder of the waste, consisting largely

*On the other hand, informal comments made by nuclear utility staff at two meetings -- Waste Management '85 in Tucson in March and the June '85 ANS meeting in Boston -- indicate that many utilities are building simple butler-type structures. It is not clear whether there is any inherent contradiction between the results of the EPRI survey and the information made available to the NRC. Perhaps the EPRI survey is already outdated.

of powdered resin sludges, was being shipped to Barnwell for burial. A long-term storage facility constructed at the Browns Ferry Plant to hold all the LLRW generated for the life of the plant was designed as a series of above-ground vault-like modules constructed of reinforced concrete. Wastes may be emplaced into or removed from each module by means of a mobile gantry crane. Each module measures 244 x 34 x 20 ft high. There are now four such modules at Browns Ferry holding 29,000 ft^3 each for about one year of storage total. For shielding purposes, the thickness of the outer walls of the modules for trash and for resins are 24 and 42 inches, respectively. A similar storage facility has been constructed at the Sequoyah Plant. HVAC or other environmental control systems do not seem to have been included in the design of the facility. These TVA extended storage facilities seem to fall into the category of "storage bunkers", as the term is used in the EPRI survey.

Detroit Edison - Fermi 2[15]

Commercial LLRW burial sites may not be available for Fermi 2 wastes after 1986. Because the storage space provided at the Fermi 2 reactor is sufficient for only a few months' production of LLRW it was decided in June 1981 to build an on-site storage facility (OSSF) to handle up to five years' production. According to Detroit Edison, the OSSF at Fermi 2 is one of the first extended storage facilities in this country. As of August 1983, the OSSF was near completion and the expected total cost was about $7 million. An account of Detroit Edison's plans for the on-site storage of LLRW at the Enrico Fermi 2 nuclear power plant follows.

The new building was located adjacent to the drum-exit area of the existing radwaste building in order to minimize handling of the drums. Thus, one building is used for both on-site storage and loading of trucks for shipment of wastes to a shallow land burial site (when one becomes available). The building also could be expanded if more storage space should be needed in the future. The entire storage building is located within the site security perimeter.

Three specific criteria were applied to the facility design:

1) simplicity,

2) efficient use of storage space,

3) passive operation, i.e., minimal reliance on mechanical or electrical operation.

A modular design was considered in which the storage area was broken up into many small cells, each containing a 3x3 array of drums. This honeycomb design was abandoned in favor of a significantly less expensive open warehouse in which the drums are stored in a few large open areas.

The design capacity of the OSSF is about ten thousand 55-gallon drums. The OSSF is divided into four sections:

1) a storage area for drums containing liquid waste solidified in asphalt (four compartments),

2) a storage area for dry active waste (two compartments),

3) a central truck bay,

4) an annex for offices, control room, dry active waste compaction, and empty drum storage.

The separation of storage areas into compartments allows for the segregation of drums by radioactivity level and/or waste type. The smallest component in the first section is a "test" drum-storage cell in which drums from certain batches of solidified waste may be set aside for periodic inspection. Up to eight layers of drums may be stacked in the storage areas by use of a 10-ton overhead crane, which can be positioned from the control room by means of TV monitors.

The reinforced concrete building is 212 ft long, 67 ft wide, and 54 ft high. The floor is 4 ft above grade and 2 in. above the maximum flood elevation for the site. The OSSF contains a HVAC system designed to maintain a minimum temperature of 50°F. No maximum temperature was mentioned. (The OSSF is thus not completely passive despite the design criterion of passive operation noted above.) Fire protection is provided by an overhead sprinkler system above all storage areas. Radiation shielding has been incorporated into the design to ensure that the dose to personnel during maintenance, operation, and any future expansion will be as low as reasonably achievable (ALARA).

According to Detroit Edison's description, the Fermi 2 OSSF provides the drums of LLRW with a relatively uniform, noncorrosive environment. (Accident conditions, such as earthquakes, tornadoes, fires, and long-term power failures, are not considered here.) Temperature fluctuations should be minimized and low humidity maintained by the HVAC system. Wetting of the waste container and waste should not be a problem under ordinary circumstances. The inclusion of a "test" drum-storage cell is useful, but for any degradation process dependent on the radiation field it should be noted that the radiation field in the center of a stacked pile of drums of solidified liquid waste may not be duplicated in the test cell. Also, the compressive stress on a drum at the bottom of a stack of eight drums may not be duplicated unless drums in the test cell are also stacked.

Susquehanna Steam Electric Station[16,17]*

At the Susquehanna Steam Electric Station (SSES), operated by the Pennsylvania Power and Light Company (PP&L), the low-level radioactive waste

*Information also obtained during visit to SSES, Berwick, PA, July 15, 1985.

holding facility (LLRWHF) is intended for contingency storage when off-site disposal facilities are not available. The SSES consists of two BWR units, rated at 1050 MWe each, with an estimated combined LLRW production rate of 60,000 ft^3/yr. This waste will be packaged in steel liners or 55-gallon drums and stored for up to four years per unit or until waste can be shipped to off-site disposal sites.

The LLRWHF is a structural steel frame building with uninsulated metal siding and roofing to provide weather protection. Its overall dimensions are 240 by 290 ft with the long axis oriented north-south. The building height at the slightly peaked centerline is 42 ft. The building encloses a system of reinforced concrete waste storage vaults. For initial facility operation, two concrete vaults are provided for storage of waste liners and trash and are located in the western half of the building. During this initial operation, the eastern half will remain an open unoccupied area. Another concrete vault will be constructed over the eastern half of the building at a later date to accommodate additional trash storage.

The reinforced concrete vaults provided for initial operation consist of 17-ft-high concrete walls, 30 in. thick on the north, west, and south sides and 24 in. thick on the east side. An 18-in.-thick wall divides this area into two separate vaults. This entire area is covered by 18-in.-thick pre-cast concrete panels with a total of 395 circular openings, each with a concrete plug which will be individually removed while a waste liner is being placed into or retrieved from storage. (The procedure for emplacing liners through these openings is briefly described below.) These pre-cast panels are supported by a structural steel framing system. For initial operation, waste liners will be stored in the west vault and trash in the adjacent vault. An inspection area is provided for waste liner inspections at any time. Trash will be stored temporarily in the adjacent vault, which is accessible by a forklift truck. In addition, an emergency exit is provided in the southeast corner of the interim trash storage vault. When this vault is converted to waste liner storage, an inspection station will be added and the emergency exit closed.

When either the interim trash storage vault or the liner storage vault approaches half-full capacity, construction will begin on an additional concrete vault in the eastern portion of the building. The additional vault area will be enclosed by 30-in.-thick concrete slab supported on metal decking over a structured steel framing system.

An 18-in.-thick concrete wall is located along the north side of the truck bay. This wall is 11 ft high in one section and 23 ft high in another and provides shielding during DAW container and waste liner storage or retrieval.

A control room located at the northeast corner of the facility has 18-in.-thick concrete walls on the south and west sides and metal siding with insulated sheetrock walls on the north and east. The ceiling consists of insulated acoustical panels below the metal roofing.

A curb around the perimeter of the building, i.e., inside the metal shell but outside the storage vaults, will contain any liquid such as rainwater or fire sprinkler water that may be introduced in to the building. The curb is designed to retain the volume of fire protection water that would be released if all the sprinklers were actuated for one-half hour. A system of floor drains for drainage of additional flow will route such water to a sump in the offloading area so that it can be sampled for contamination and collected for disposal. Ramps are provided for vehicular traffic over the curb. The floor, curbing, sumps, and shield walls of the facility will be coated with a decontaminable material to a height equal to the height of the curbing.

The facility is equipped with an active ventilation system designed such that it moves air generally upward, away from the equipment operators, removes noxious or irritating exhaust fumes whenever internal combustion engine powered machinery is operating inside the facility, and prevents excessive heat buildup from the roof in the summer. The ventilation system for the storage areas and truck bay will not provide any heating, air conditioning, or humidity control for the facility. The facility room, however, is provided with both heating and air conditioning, which normally will be required when occupied while LLRW is being loaded or unloaded at the facility.

Containers of cemented waste are placed under a shield bell and onto a flat bed truck at the SSES solidification facility for transit to the LLRW Holding Facility. After the flat bed truck arrives at the facility and is in the storage location, the 10-ton monorail hoist is used to lift the vault ceiling shield cover plug. Then the bridge crane is maneuvered to place the shield bell into the position vacated by the shield cover plug. The waste container is then lowered into its storage position by a chain hoist inside the shield bell. After the plug is replaced the shield bell is returned to the truck bay. The procedure is reversed to retrieve a container for inspection or offsite shipment for disposal.

Fifty-five gallon steel drums containing trash are placed on pallets and loaded onto a truck at the SSES radwaste building and transported to the holding facility, where the LLRW is unloaded and maneuvered into the interim or final trash storage vault with a battery-operated forklift. During the initial phase and throughout the life of the LLRWHF, the low-level DAW and cemented waste will be segregated. DAW will be stored in the trash storage vault and cemented waste in the solidified waste storage vaults. The waste containers will be further segregated within the vaults to take maximum advantage of the self-shielding properties of the waste material and to minimize exposure. To the extent practicable, waste stored in the trash storage vault will be arranged with containers having contact dose rates $\leq$30 mrad/h along the vault walls and on the top layer of the storage area and containers with higher dose rates stored underneath. Similarly, cemented waste stored within solidified waste storage vaults will be arranged with containers having contact dose rates $\leq$3 rad/h stored next to the vault walls and on the top layer. Containers with a contact dose rate >3 rad/h will be stored inside this perimeter.

After one year's storage, waste containers will be inspected quarterly. A container will be moved out of its storage location to the inspection station where it will be visually inspected for deterioration, leakage or other conditions which might preclude shipment and disposal without repackaging. Inspections required by 49 CFR Part 170 can also be performed at these stations before the waste is shipped offsite for disposal.

Because of the nature of dry trash wastes, and because of precautions to prevent container contact with water, PP&L expects corrosion to be minimized. Also, PP&L believes that this type of waste in steel container can be stored for four years without loss of container integrity. Therefore, no integrity monitoring program is planned for the trash containers. At the time of shipment offsite, the containers will be inspected to determine if they are acceptable for transportation and receipt at the disposal site. If problems occur, PP&L has the capability to repackage the waste at the SSES radwaste building.

Robert Emmet Ginna Nuclear Power Plant*

The LLRW storage facility at the Ginna Nuclear Power Plant is the upper radwaste storage building (URSB), where wastes packaged in 55-gallon drums and LSA boxes are kept until they can be shipped. The URSB is a sheet metal building with a concrete base and is about 90 ft long and 50 ft wide. There is about 16 ft of useful storage height on allowing for operation of the overhead crane in the building. There are no heating or cooling systems in the URSB. A concrete pit measuring about 57 ft x 15 ft and 8 ft deep was at one time a storage bunker for high activity waste. A 6-ft-high concrete wall surrounds the entire URSB and, at the rear of the URSB, a 5-ft-high lead barrier provide radiation shielding on the north wall for guard towers. Also, lower activity drums are stacked around the occasional higher activity drum -- e.g., one containing cement-solidified evaporator bottoms -- to provide shielding. Volume reduction of the dry activated waste at the plant is accomplished with a mobile shredder/super-compactor system and a drum compactor. The URSB has a storage capacity for 5 years' production of dry active waste. The URSB also serves as a test area for contaminated equipment used during outages, but in the event radwaste could not be shipped off site, no equipment testing would be performed in the URSB in order to assure the availability of the URSB for storage of radwaste. (The contaminated equipment is stored elsewhere.)

Ion-exchange resins are currently stored in two spent resin holding tanks. The resin beds are renewed on the basis of their decontamination factor rather than a set time period. The resins have been transferred from the holding tanks for dewatering and immediate shipment in high integrity containers to a disposal facility. The usual radiation field for the resins during transfer is from 40 to 50 R/h, but readings as high as 100 R/h have been observed.

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*Information obtained during visit to Ginna Plant, Ontario, NY, June 26, 1985 and in a telephone conversation between F. J. Mis, Rochester Gas and Electric, and B. Siskind, BNL, August 22, 1985..

If sufficient low-level radwaste disposal capacity for the Ginna plant should become unavailable, contingency storage plans include construction of additional prefabricated buildings for dry active wastes, and expansion of the storage capability of the spent resin holding tanks. Corrosion of steel drums in storage is not expected by the Ginna Plant staff, nor is caking or bridging of the stored resins. No corrosion of the drums kept in the URSB has been noted. In any case, the use of a plastic lining inside the drum to facilitate repackaging in the event the drum completely corroded away has been considered. No difficulties have been encountered with transfer of spent resins from the holding tanks. The resins in the holding tanks are kept wet and can be sparged with nitrogen gas to facilitate dispersal in the event bridging or caking should occur.

Calvert Cliffs Nuclear Power Plant*

At the Calvert Cliffs Nuclear Plant, LLRW is stored in an area that is an integral part of the Materials Processing Facility, which is a radwaste volume reduction and decontamination facility. The area provides 75,000 ft^3 of storage volume, which at the expected rate of waste production [mostly dry active waste (DAW)] of 15,000 ft^3/yr is sufficient for 5 years. The walls of the storage area consist of 8-in.-thick poured concrete, but no temperature control is provided. There is heat during cold weather in the processing area. The unit has a ventilation system; however, the air pressure in the storage area is slightly negative relative to the processing area.

DAW wastes are packaged in 55-gallon drums or 4-ft x 4-ft x 6-ft steel boxes for storage or shipment. Spent resins are packaged in HDPE HICs for shipment. The planned strategy regarding storage and shipment is to ship higher activity waste (Classes B and C) and store lower activity waste (Class A). The DAW is Class A. About 660 ft^3 of sandblasting grit contaminated with 10^{-5} μCi/cc Cs-137 and 10^{-6} μCi/cc Co-60 is being stored. (If these contamination levels were in aqueous waste, the radionuclide content of the waste would be below regulatory concern according to 10 CFR Part 20.) Some contaminated oil (less than 10^{-4} μCi/cm^3) is being stored but will be incinerated. Should disposal capacity become insufficient or unavailable for the plant's Class B and Class C wastes, a contingency plan involving on-site modular storage of these wastes may be implemented.

2.2.4 LLRW Storage Outside the U.S.

Storage of LLRW has been implemented in other countries. An overview is given below of some of the storage concepts in use outside the United States.

*Information obtained during visit to Calvert Cliffs Plant, Lusby, MD, June 14, 1985.

2.2.4.1 Italy

Since 1960, CAMEN, the Center for Military Applications of Nuclear Energy in Italy, has solidified relatively low-level waste with concrete inside cylindrical containers made of concrete. The waste has been stored in a monitored open bed without further protective covering. A report[18] gives the results of a study of the state of preservation of some 400 containers placed in storage through 1979.

Two methods were used for assessing container performance:[18]

1. Visual inspection of the condition of the outer surface. This inspection made it possible to identify and classify alterations found in the concrete of the container which might also involve the solidified waste, e.g., segregation of components within the waste form, laitance, visible cracks, and leaks.

2. A systematic survey of transferable contamination which might be present on the surface of each container, using "smear" techniques described in IAEA standards for shipping radioactive material.

Visual inspection showed that alterations of the concrete stabilized within a few years, regardless of the method of container manufacture. The damage caused by atmospheric agents and seasonal cycles was slight, in keeping with observations during management of the site that most of the observed alterations appeared early, during the period of aging of the concrete and the very earliest part of the storage period. The contamination survey showed that only a very small number of containers had detectable removable surface contamination. Only one container had sufficient surface activity to be classified as contaminated by the CAMEN radiological protection standards, i.e., $>5\times10^{-5}$ $\mu Ci/cm^2$ surface radioactivity.

It was concluded[18] that under the storage conditions at CAMEN

- the release of radioactivity tends to exhaust itself over time;

- after an initial settling period, the characteristics of the containers do not vary appreciably; and

- containers made of concrete, when adequately built and aged, will fulfill the requirements for radiological safety for several decades.

Since the storage at CAMEN is exposed to the elements, it may be concluded that concrete containers of good quality stored in an enclosed structure should provide at least as much protection as those in this study. There are some limitations on this conclusion:

- As stated in the CAMEN report, the containers must be adequately built and aged. ("Adequate" building and aging are not specified.)

- Release of radionuclides could occur during transport of concrete containers to a disposal site after storage if there is a loss of integrity. The container must meet DOT requirements, e.g., DOT drop test. (The modular reinforced concrete storage casks described above in Section 2.2.2 are essentially overpacks for waste containers. The CAMEN concept, however, involves LLRW solidified in concrete inside cylindrical concrete containers.)

- No information is given in the CAMEN report on the temperature extremes to which the containers would be exposed, but the annual and even daily temperature variations are probably greater in some parts of the United States. (A discussion of the resistance of concrete to weathering is given below in Section 3.2.)

2.2.4.2 Canada

In Canada renewable licenses are issued for radioactive waste storage facilities on the basis of a maximum 50-year storage period. If wastes will not have decayed to a de minimis* activity level by the end of this period, they must be stored retrievably. The electric utilities operating nuclear reactors handle their own LLRW, while the federal government, through the crown corporation Atomic Energy of Canada, Ltd. (AECL), handles all the rest of the LLW produced both at the federal government laboratories and by academic, institutional and industrial generators. Both the utilities and AECL have been storing the bulk of this waste. They are currently investigating methods for permanent disposal, since waste containing long-lived isotopes will eventually have to be disposed of, and it is less costly to dispose of it in the first place than to have to retrieve and dispose of it later.[19]

AECL[19]

Most of the waste handled by AECL is incinerated (in a controlled air incinerator) or baled, and the residue stored in engineered facilities. The next step is the immobilization of the residue by bituminization. This is considered a necessary treatment before disposal. The volume reduction techniques (incineration and baling) cut in half the need for new storage facilities from the recent usage of 3000 m^3/yr. On the basis of the annual rate of generation the most important radionuclides associated with the wastes (and their annual rates of generation for AECL maintenance waste) are Cs-137 (10 Ci/yr), Sr-90 (9 Ci/yr), H-3 (100 Ci/yr) and Co-60 (70 Ci/yr).

Wastes containing nominally more than 0.1 Ci/m^3 of fission or activation products, and certain long-lived radionuclides, are stored in engineered structures buried in-ground at least one meter above the water table. Various designs of reinforced concrete structures have been used ranging from rectangular bunkers (61 x 4.9 x 2.4 m deep) to the currently used cylindrical

*The term "de minimis" is not further defined in Reference 19.

designs 0.15 to 6 m diameter and 4 to 5 m deep. Each facility is fitted with a removable weatherproof cap.

Nuclear-Electric Utilities[19]

Ontario Hydro operates a centralized waste management facility at its Bruce Nuclear Power Development site which receives waste from nine reactors at a rate of about 3000 m^3/yr. The waste is volume-reduced by incineration or compaction using equipment similar to that employed by AECL (controlled air incinerator and in-drum baler). Initially, LLRW was stored in concrete trenches while LLRW (such as ion-exchange resins) was stored in waterproof concrete tile holes. Both types of facilities are set in the ground well above the water table. The latest facilities have been constructed above ground in a prestressed, prefabricated concrete building. The cost of storing LLRW in this way is reduced by a factor of 3 compared with in-ground trenches, and land usage is reduced by a factor of 10.

It appears that the extensive experience which the Canadians have had with storage of low-level radioactive waste has been essentially all positive. They are moving towards a waste management system involving permanent disposal, for one thing because there is a limit on the storage time permitted. However, they plan to characterize and segregate wastes according to their hazardous lifetimes, and those whose lifetimes are short enough can still be stored to decay to de minimis levels. (See the discussion of storage for decay in Section 2.1, above.) Such a strategy has much to recommend it since it is less costly than permanent disposal of all radwaste and appears to be reasonable in terms of health risks.

2.2.4.3 Scandinavian Countries[20]

Sweden is currently storing low- and medium-level radwaste prior to final disposal. The estimated storage time ranges from 5 to 50 years. All stored radwaste is solidified in either bitumen or cement and packaged primarily in 0.2 m^3 (55-gal) drums. Some cemented waste is also packaged in 1 m^3 (interior volume) reinforced concrete containers. The waste is stored aboveground in shielded warehouses and belowground in engineered rock caverns.

The Scandinavian countries have chosen to use the Swedish-designed storage methods as reference systems for temporary radwaste storage. The storage facilities include different kinds of storage buildings for bituminized and cemented wastes and an underground rock cavern for both bituminized and cemented wastes.

Bituminized Waste Storage (BWS)

The BWS building is ventilated and has smoke detectors and a CO_2 fire-extinguishing system. The circulation-type (not once through) ventilation system includes filters to control airborne particulate activity and dehumidifiers to control corrosion of the painted mild steel drums. The drummed waste is passed through an airlock into the ventilated storage room. Drum handling

is accomplished with an overhead crane. Mirrors and closed-circuit TV permit semiremote operation of the crane. The drums are stored in pits. Each pit contains 24 drums arranged in six layers of four drums in a square array. A steel plate is placed between every second or third layer of drums for stability. A removable concrete lid, which closes each pit as it is filled, provides shielding and comprises part of the floor of the storage room.

Cemented Waste Storage (CWS)

The much simpler CWS building is essentially a shielded warehouse in which the cemented waste, in either steel drums or 1-m^3 reinforced concrete boxes, is stacked up to the ceiling. The waste is handled by a forklift with a shielded cabin. The CWS building uses only natural ventilation through labyrinth-shielded vent openings in the walls of the building. Neither filtering of the air, nor forced circulation, nor any other air conditioning is provided for.

Rock Cavern Storage (RCS)

The RCS system consists of tunnels mined in rock at a depth from the surface of approximately 30 m, depending on rock quality. RCS may be used for final disposal, but is considered temporary until the decision is made to seal the cavern. The cavern is designed to store both cemented and bituminized wastes. However, bituminized waste drums are packaged in the 1-m^3 reinforced concrete boxes for RCS to alleviate any fire hazard. Waste is handled in the storage area by an overhead crane and remote closed-circuit TV. The RCS system is ventilated and drained of water. The drained water is also treated.

A safety analysis of the storage systems included the following considerations:

- Normal events: No problems are anticipated with the waste packages due to impacts, scrapes, etc. involved with normal handling procedures.

- Environment: The 5- to 50-year storage time frame means that temperature and humidity may need some control where corrosion and/or freeze-thaw cycling may affect waste stability. For bituminized waste, corrosion of the drum is not allowed since the drum provides mechanical stability. Dehumidifying the air and protecting the drums with a suitable paint is felt to provide adequate protection from corrosion for bituminized waste drums.
  Cemented waste drums can tolerate some corrosion since the drum is not required for mechanical stability of the cemented waste.

- Processes in waste which may cause degradation: Neither radiation damage nor biodegradation is considered significant for low- and medium-level bituminized and cemented radwaste over a 50-year period. (It should be noted that there is some evidence that biodegradation may contribute to the corrosion of metals embedded

in bitumen. See the discussion below in Section 3.2.) Swelling may cause breaching of a waste container and should be allowed for or limited. Swelling of up to 5% can be tolerated by bituminized waste since the drum normally has 8 to 10% free space and bitumen can deform. However, it was noted that water immersion can lead to a greater than 5% swelling in bituminized waste. Swelling of concrete waste is more serious. Swelling of more than 0.3% by volume could lead to cracking of the waste form and concrete box or rupture of a drum. To alleviate this the concrete molds are lined with a 1-cm foam plastic mat to absorb minor swelling. (The swelling of bituminized and cemented waste forms on immersion in water is discussed below in Sections 3.1 and 3.2.)

- Abnormal events: Abnormal events during storage and related handling include high-impact collisions, dropping and fire. If collision and/or dropping velocities do not exceed 55 km/h (i.e., a drop height of 14 m), no significant release of activity is expected from waste packages. Very little dust-sized particulate matter results from such impacts, and cleanup of larger debris should be a trivial matter in the isolated storage area. Bitumen fires are hard to ignite, but the possibility must be considered. Thus fire-extinguishing systems are required for BWS. Possible causes of bitumen fires included sabotage, an airplane crash followed by a fuel fire, a large forest fire, and failure of overhead cranes. The probability of sabotage was said to be difficult to estimate, but very low. The probability of an airplane crashing into a BWS building was estimated as being less than 10^{-7} per year, which was considered an acceptable risk. The final two possibilities were considered to be remote.

2.2.5 Generic Classification of Extended Storage Facilities

From the preceding overview of the various approaches to the problem of extended storage of LLRW, it may be seen that a wide variety of solutions to the problem have been proposed. To facilitate the discussion of storage environments in Section 2.3, a comprehensive generic classification of the LLRW extended storage concepts is needed. Such a classification has been given in New York State's recent study of LLRW management:[12]

- Shielded Storage Buildings - These are permanent buildings designed specifically for the extended storage of LLRW. They are generally provided with separate shielded areas for the storage of dry active waste and solidified waste. Overhead bridge cranes are used for remote handling of the waste packages. Building ventilation systems are typically provided. This concept is essentially the same as containerized storage in large structures, the concept presented in the AIF study.

- Shielded Storage Modules - These are permanent concrete structures at least partially aboveground. Waste containers are emplaced or

retrieved from above by a gantry crane. This approach is used by the TVA. (See Section 2.2.3 for a description of the TVA facility.)

- Shielded Casks - These are cylindrical all-weather concrete containers that can be located outdoors and that are designed to hold waste drums or liners.

- Unshielded Facilities - These can range from minimal unshielded facilities, e.g., simple fenced-in outdoor concrete pads, to unshielded prefab structures, e.g., sheet metal or concrete buildings designed for containerized waste storage. Unshielded facilities are generally intended for low-specific-activity wastes. The waste containers are generally handled by means of hand dollies, forklift trucks, or cranes. These facilities are generally used for storage for decay rather than extended storage.

Storage facilities based on all the above storage concepts are currently in use or proposed for use at nuclear power plants in the United States. It should be noted that storage of radwaste in facilities based on these concepts is by no means limited to nuclear power plant radwaste; non-fuel-cycle LLRW may also be stored in such facilities.

Storage concepts not in these four categories, e.g., the Scandinavian rock cavern storage concept, do not seem to be under active consideration in the United States and will not be considered further in this report. The above four categories seem to adequately span the range of extended storage concepts proposed or actually under construction in the United States, and they will be used in the discussion of storage environments in Section 2.3.

2.3 Storage Environments

The properties and 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 nature of these effects for specific waste forms, binder materials, and container materials will be discussed below in Section 3.1, 3.2, and 3.3, respectively. In this section the following environmental variables that may significantly affect the properties and behavior of the wastes and associated materials are considered: temperature, humidity, potential for wetting, radiation field, and length of storage time. The ranges for these variables in four generic storage concepts will be deduced from the information about these storage concepts presented in the preceding section. Unfortunately, explicit information about these variables is generally not presented in descriptions of LLRW storage facilities.

The temperature of the storage environment will vary only slightly in the large engineered structures for containerized radwaste that include HVAC systems in this design. As noted above, a minimum temperature of 50°F (10°C) is explicitly mentioned by one utility for its LLRW storage facility.[15] An upper bound for the temperature of 80°F (27°C) is conservatively estimated

here and sharp variations even within this temperature range would not be expected. The HVAC system in that facility is also designed to protect the stored drums from corrosion due to humidity. No numerical values for the relative humidity are given, but it will be assumed to be below the critical value at which atmospheric corrosion becomes significant. For steel this value ranges from about 50% to 70%[21] (see Section 3.3, below). Outdoor temperatures, in contrast to the controlled temperatures within the storage facility, have ranged from as low as -16°F (-27°C) in January to as high as 102°F (34°C) in July near the site of the facility.[22] 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] It is not clear whether the temperature of the storage environments is controlled to any extent at these last two facilities. On the other hand, in the simple fenced-in concrete storage pad, also described in the preceding section, the waste will be exposed to the outdoor temperature. Similarly, the humidity in the simpler storage concepts will vary with the outdoor humidity. Except under abnormal operating conditions, the potential for wetting is significant only for waste stored outdoors or in very simple structures (as in the case of the concrete storage containers at CAMEN in Italy. See Section 2.2, above).

For α and β radiation it may be assumed to a very good approximation that 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 containers 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. The dose to waste packages stored in proximity to one another should include the contribution from γ-emitting radionuclides in nearby packages. The effects of radiation fields are considered further in Section 4 and Appendix C.

The storage time is a variable determined chiefly 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 LLRW must be stored. In this report, a 5- to 15-year storage period is assumed.

The environmental variables discussed above are summarized in Table 2.7 in a largely qualitative manner for the four categories of storage concepts described in the New York State LLRW study.[12] 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

heating, ventilation, and air-conditioning (HVAC) system. The temperature limits in such a facility, as discussed earlier in this section, are conservatively taken as 50°F (10°C) and 80°F (27°C). Similarly, the relative humidity is assumed to be less than 50%. The potential for wetting of the waste in a large engineered storage structure is negligible except under accident conditions, e.g., activation of sprinkler systems in case of fire. (This can be reduced even further if a CO_2 fire extinguishing system is implemented as in one of the Scandinavian storage facilities. See Section 2.2.) The effect of the radiation field from adjacent waste containers may be conservatively estimated by approximating the stacked waste containers by an infinite absorbing medium with an activity concentration equal to that of the waste. See Appendix C for this calculation. (Note that the absorbed dose, an intensive quantity, is defined as the imparted energy per unit mass.) For Co-60 the absorbed dose for a continuous medium is 2.7 times greater than for a 55-gallon drum. For Cs-137, it is 2.1 times greater. For Sr-90, a pure β emitter, the two absorbed dose values are the same. As a conservative upper bound, the γ dose to a continuous medium may be taken as three times the dose to a 55-gallon drum from the same γ-emitting isotope at the same concentration.

For the other three categories of storage concepts, there is generally no environmental control other than that provided by the module on 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. Outdoor temperatures may range from below -40°F (-40°C) to above 104°F (+40°C), although daily variations are much less and depend on location and season. The relative humidity also depends on the season, location, and time of day and range from below 30% to 100%. See, for example, Weather Atlas of the United States[23] for details regarding specific locations at specific seasons. Very conservative limits of -40°F (-40°C) and +104°F (+40°C) and 0% and 100% have been taken for the ambient temperature and relative humidity ranges, respectively. Because insolation of the storage facility may increase the temperature to greater than ambient, an upper limit of 140°F (+60°C) is given in Table 2.7. The potential for wetting is negligible to very large, depending in part on how well the modules or casks are sealed and on how well they are protected from the elements and on the local rainfall. The radiation field from adjacent packages is potentially significant if several packages are placed in any given module or cask.

Table 2.7

Summary of Extended Storage Environments

	Large Engineered Structures	Storage Modules	Shielded Casks	Unshielded Facilities
Temperature (°C)	Controlled 10 to 27	Uncontrolled, near ambient[a] -40 to +60	Uncontrolled, near ambient[a] -40 to +60	Uncontrolled, near ambient[a] -40 to +60
Relative Humidity	Controlled <50%	0% to 100%	0% to 100%	0% to 100%
Potential for Wetting	Negligible	Negligible to moderate	Negligible to moderate	Negligible to very large[b]
Radiation Field From Adjacent Packages	Potentially significant. Factor of up to 3[c]	Potentially significant[c]	Slight to potentially significant[d]	Potentially significant[c,e]

[a]Insolation may increase temperature to greater than ambient.
[b]Negligible in prefabricated structures. Very large in uncovered storage pad.
[c]Conservative ratio of dose to infinite medium (approximating stacked drums) to dose to 55-gallon drum for γ radiation. Assumes multiple packages in storage structures.
[d]Slight if only one waste package per cask. Potentially significant if cask contains several packages.
[e]Not significant if such facilities are used only for storage of low-specific-activity waste.

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3. PROPERTIES AND BEHAVIOR OF WASTES, BINDERS, AND CONTAINER MATERIALS

In this section, the properties and behavior of low-level radioactive waste (LLRW) streams, solidification agents, and container materials are reviewed. The emphasis is on those characteristics of these materials that may be important for predicting the behavior of the waste forms and containers during extended storage and for assessing the effect of extended storage on waste form stability and container integrity during transport and after disposal. In addition to ordinary chemical processes which may degrade the performance of the binder or container materials (e.g., atmospheric corrosion of carbon steel containers), the effects of the radiation field on the properties, and the behavior of the waste package materials are also addressed.

In 10 CFR Part 61 Subpart D, LLRW is classified according to the concentration of radionuclides. Class A waste must meet certain minimum requirements for near-surface disposal and Class B and Class C wastes, with progressively higher concentrations of certain radionuclides, must meet more rigorous requirements for disposal. Class A wastes are mostly trash, paper, plastic, low-specific activity resins, and various institutional wastes; Class B wastes tend to be evaporator concentrates, resins, and spent filters; Class C wastes include certain wastes generated by industry, e.g., rejected or spent radiation sources, as well as discarded activated parts and equipment from commercial nuclear power plants.[1,2]

In the discussion of waste streams, emphasis is on the radiolysis of the wastes, since the resulting production of gas and corrosive substances could, in principle, adversely affect the mechanical integrity of the binder material, corrode or pressurize the container, or create an explosion or flammability hazard in the storage facility. In Section 3.1, an overview is given of the available information about the radiolysis of these wastes. Calculations of the radiation dose to the waste package from the radionuclides are presented in Appendix C.

Radiolytic gas generation is also a concern in the consideration of binder materials during storage. In addition, the effects of temperature and humidity on the behavior and properties of the binder materials may also be important. In Section 3.2, a survey is given of the effects of radiation, temperature, and humidity on cements, asphalts (or bitumens), and thermosetting organic polymers.

Degradation of containers during storage may lead to container failure during storage and may compromise the ability of the containers to meet requirements for handling and shipping. In Section 3.3, the corrosion of carbon steel in contact with the wastes and by the atmosphere is discussed. It should be noted that even in the absence of a radiation field, the chemical compatibility of dewatered or solidified LLRW with the storage container material is of concern. It is assumed in this report that once the LLRW is placed in a container, it will not be repackaged unless its container deteriorates during storage. It is further assumed that, because of the additional

handling and resulting occupational exposure to radiation, such repackaging of the waste for transport and disposal is undesirable and is to be avoided. Both the minimum and stability requirements include limitations on the free liquid in the waste. The waste must, therefore, be dewatered, packed with sufficient absorbent material, or solidified. If Class B or Class C waste is dewatered but not solidified, a high integrity container as described in the NRC Technical Position Paper on Waste Form will be required. If such waste is solidified, a container such as a carbon steel drum may be used. The chemical resistance of high integrity container materials and concern about the corrosivity of solidified waste forms towards carbon steel are discussed below in Sections 3.2 and 3.3. Some data on the corrosivity of various simulated LLRW toward carbon steel in the absence of a radiation field, particularly relevant for the extended storage of Class A wastes, are also discussed. Finally, the radiolytic degradation of polyethylene is considered. Where the effects of radiation -- on the waste, on the binder, or on the container material -- are discussed, it should be noted that the effect of the radiation dose rate, particularly in the presence of oxygen, may be as important as the total radiation dose.

It must be emphasized that nonradiolytic effects are likely to be the primary concern for most LLRW packages. Based on the concentrations of radionuclides, most LLRW packages are found to contain Class A waste. For example, according to a recent study by New York State,(1) the LLRW volumes generated by the commercial sector (i.e., commercial nuclear power plants, academic and medical institutions, and industries) may be categorized as follows: 60% Class A, 30% Class B, and 10% Class C. Even higher percentages of Class A waste were estimated in a survey carried out by BNL for the NRC.(2) The 16 nuclear power plants responding to the survey all reported that over 80% of their LLRW volume shipped off site in 1984 was Class A. Based on recent estimates attributed to NRC,* Class C wastes comprise only about 1 volume percent of the nation's LLRW but contain up to 80% of the activity at the time of disposal. Here, it should be stressed that the information in this chapter is based on the results of tests and experiments that in many cases, particularly for phenomena involving radiation, were carried out under worst-case (or even beyond realistic worst-case) conditions in order to accelerate testing or for the sake of conservatism.

3.1 Waste Streams

The characterization of low-level radioactive waste streams used in this report follows that developed in the Draft Environmental Impact Statement (DEIS) for 10 CFR Part 61.(3) In the DEIS, 25 low-level radioactive waste streams are identified which are considered to constitute the bulk of such waste to be generated by the year 2000. These waste streams, divided according to source into five categories, three fuel cycle and two non-fuel-cycle,

*"No Technical Reasons to Separate Class C From Other LLW, House Subcommittee Told," Nuclear Waste News, July 25, 1985, p. 181.

are listed in Table 3.1. In order to facilitate the assessment of the extended storage of LLRW, the 25 waste streams may be grouped in an alternative manner according to similarities in their macroscopic characteristics:

- Light Water Reactor (LWR) Process Wastes
- Trash
- Low-Specific-Activity (LSA) Wastes
- Special Wastes

LWR process wastes are usually wet wastes specific activities comparatively higher than either the trash group or the LSA group. The trash group contains most of the combustible LLRW generated. The LSA group includes all the streams containing comparatively small activities that are not included in the LWR process waste group or the trash group. The "special" waste group includes streams that contain relatively high concentrations of radioactivity and are small in volume when compared with the other three groups.[3,4] The waste streams comprising these four groups are listed in Table 3.2. A fifth group includes potentially large but unpredictable streams such as those from power plant decommissioning and nuclear fuel reprocessing activities. Since it is not known when (or if, in the case of reprocessing) such wastes will be generated, they are not considered further in this report.

The LWR process waste streams are the best characterized of all the LLRW streams.[3,4] In the following discussion of the characteristics and properties of waste streams, attention is given to ion-exchange media and, in particular, the behavior of commonly used organic ion-exchange resins in a radiation field because of the relatively large amount of work that has been done in this area. A brief account is then given of the other LWR process wastes.

The following discussion of waste stream characteristics deals only with fuel-cycle wastes, with emphasis on radiolytic properties important for Class B and Class C wastes. 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 (e.g., LLRW generated by Union Carbide Corporation, New England Nuclear Corporation, and the 3M Corporation and described in recent reports by BNL for the NRC[5-7]) no general accounts of properties and behavior are available. Even for these special cases, the descriptions refer to wastes generated prior to the effective date of 10 CFR Part 61. If a non-fuel-cycle waste is Class A, it is likely to be stored in dewatered form in its shipping container; if 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. In either case, much of the discussion below in Sections 3.2 and 3.3 will be applicable. The results of a recent survey of LLRW generators[2] suggest that only a small proportion (<5%) of the non-fuel-cycle LLRW volume is likely to be greater than Class A.

Table 3.1

Waste Sources and Streams[a]

Nuclear Fuel Cycle
Central Station Nuclear Power Plants
Ion-Exchange Resins
Concentrated Liquids
Filter Sludges
Cartridge Filters
Compactible Trash
Noncompactible Trash
Non-Fuel Reactor Core Components
Decontamination Resins
Fuel Fabrication Facilities
Process Wastes
Compactible Trash
Noncompactible Trash
Uranium Hexafluoride Plants Process Wastes
Non-Fuel Cycle
Institutional Facilities
Liquid Scintillation Vials
Absorbed Liquid Waste
Biowaste
Trash
Industrial Facilities
Waste from Isotope Production Facilities
High Activity Waste
Tritium Production Products Manufacturing Waste
Sealed Sources
Accelerator Targets
Source and Special Nuclear Material Waste
Source and Special Nuclear Material Trash
Low Activity Waste from Various Sources
Low Activity Trash from Various Sources

[a]Reference 3.

Table 3.2

Waste Groups and Streams[a]

Group	Stream
I. LWR Process Wastes	Ion-Exchange Resins[b] Concentrated Liquids[b] Filter Sludges[b] Filter Cartridges
II. Trash	LWR Compactible Trash[c] LWR Noncompactible Trash[c] Fuel Fabrication Compactible Trash Fuel Fabrication Noncompactible Trash Institutional Trash[d] Industrial Source & SNM Trash[d] Industrial Low Trash[d]
III. Low Specific Activity Wastes	Fuel Fabrication Process Wastes UF_6 Process Wastes Institutional LSV Waste[d] Institutional Liquid Waste[d] Institutional Biowaste[d] Industrial Source & SNM Waste Industrial Low Activity Waste
IV. Special Wastes	LWR Non-Fuel Reactor Components LWR Decontamination Resins Waste from Isotope Production Facilities Tritium Production Waste Accelerator Targets Sealed Sources High Activity Waste

[a]Reference 3.
[b]Further subdivided into BWR and PWR.
[c]Further subdivided into BWR and PWR.
[d]Further subdivided into large facility and small facility.

3.1.1 Ion-Exchange Media

Ion-exchange media, which are used in LWRs to remove the radioactivity dissolved in liquids, usually consist of organic resins or, in some cases, inorganic zeolites. Spent ion-exchange media must be dewatered or solidified for disposal but are typically handled at the LWR in the form of a slurry. This slurry may be dewatered in the shipping container prior to shipment.

Dewatering removes free water only; dewatered ion-exchange media may still contain as much as 55% water by weight in the form of absorbed, adsorbed, and interstitial water. Structural stability for disposal of dewatered Class B or Class C ion-exchange media may be provided by a suitable high-integrity container. Alternatively, the spent media may be stabilized by incorporation into a solid matrix consisting of binder materials such as those discussed in Section 3.2.[3,4]

Dried ion-exchange resins are extremely hygroscopic and swell on absorption of water. This swelling can compromise the physical integrity of the waste form. For example, Bonnevie-Svendsen et al.[8] have presented the results of studies on the incorporation of spent ion-exchange resins into bitumen. These workers noted that up to 60% (presumably by weight) dry resin could be incorporated into bitumen without impairing the water resistance of the waste form products; for products with more than 60% dry resin, however, the waste form was observed to swell when it was put in contact with water. Similarly, Colombo and Neilson[9] report swelling, cracking, and loss of physical integrity of bitumen samples as a result of immersion in water during leach testing; these bitumen samples contained sodium sulfate concentrations ranging from 17.5 to 44.3 weight percent. In both cases, the swelling of the bituminized waste forms is attributed to the uptake of water by the hygroscopic waste incorporated into the bitumen.

3.1.1.1 Radiation Effects on Ion-Exchange Resins: General

In a survey by BNL of U.S. nuclear utilities, some information was obtained on the radionuclide loadings on spent ion-exchange resins.[10] Large variations found in both the total activity and the radionuclide species making up the resin loading were presumably due to different operating conditions at different plants. The loadings for spent coolant cleanup resins, both maximum and typical, show more than a hundred-fold variation, the maximum loadings reported ranging from 0.3 to 60 Ci/ft^3 and typical loadings from <0.1 to 30 Ci/ft^3. Also, the distribution of the relatively long-lived radionuclides varies from predominantly activation products (e.g., Co-60, Mn-54) to largely fission products (Cs-134, Cs-137). Relative to the dose from Co-60, and Cs-137 and, possibly, Sr-90, the long-term radiation doses delivered to the resin by the other radionuclides is small because of their shorter half-lives. The dose to resins loaded to about 10 Ci/ft^3 with one of these isotopes is estimated to be about 10^8 rad, and the dose rates are estimated to fall between 10^2 and 10^3 rad/h.[10] In addition, according to the NRC Technical Position on Waste Form, resins should not be generated with radionuclide loadings that will produce $>10^8$ rad total accumulated dose. This specification, which was formulated to minimize adverse radiation degradation effects after disposal, will also limit the total dose to the waste package during storage. It should be kept in mind, however, that if radionuclides such as Cs-137, Co-60, and Sr-90 predominate (with half-lives of 30, 5.3, and 28.1 years, respectively), much of this 10^8 rad dose will be accumulated during a 5- to 15-year storage period. Limiting the total dose to the waste package and its contents to 10^8 rad may mitigate many of the radiolytic effects discussed below if dose rate effects are unimportant. (See Appendix C

for examples of dose calculations for LLRW packages. See MacKenzie, Lin, and Barletta[10] for further details of the resin loadings.)

A large number of laboratory studies have been carried out on radiation effects in ion-exchange media. In the review by Gangwer, Goldstein, and Pillay[11], it is noted that these studies tend to be concerned more with radiation effects on process parameters such as ion-exchange properties than with the behavior and properties of spent resin radwaste. For example, it appears that soluble, chemically aggressive species as well as combustible gases may be produced as a result of the radiolysis of organic ion-exchange media, but little information is available to relate existing laboratory results to irradiation effects on ion-exchange resin waste under actual storage and disposal conditions. It should also be noted that much of this work was done at radiation doses above 10^8 rad. The above-mentioned limitation on resin loadings, i.e., $<10^8$ rad total accumulated dose, may render at least some of these radiolytic effects unimportant for extended storage unless dose-rate effects are important. More recent work dealing with radiolytic effects in ion-exchange resins has been reviewed by Piciulo.[12]

In some early scoping studies at BNL[13] and at the Georgia Institute of Technology,[14,15] under subcontract to BNL, the effects of radiation (up to 5×10^9 rad) on organic ion-exchange resins were addressed. The properties of a proprietary ion-exchange material, D-mix, said to be typical of resins in the Epicor-II demineralizer system used in the water cleanup operation at Three Mile Island, were investigated as a function of radiation dose by Barletta et al.[16] This work was continued at BNL by Swyler, Dodge, and Dayal,[17,18] who studied organic resins consisting of a polystyrene-divinylbenzene polymer backbone with sulfonic acid (cation exchange) or quaternary ammonium (anion exchange) functional groups. These resins are considered typical of those used in the U.S. commercial nuclear power industry. Cation, anion, and mixed bed resins were all included in the BNL study. The following radiolytic effects of potential significance to the extended storage of LLRW have been emphasized:

- the release of chemically active soluble decomposition products and ions,
- the generation or uptake of combustible or corrosive gases,
- the effect of radiolytic resin decomposition on solidification properties, and
- the effect of radiation on the corrosion of metals (in particular, mild steel) in contact with the resin.

The amount of a chemical change induced by radiation is commonly expressed in terms of the G-value, the number of molecules changed for each 100 eV of radiation energy absorbed.[19] (When considering radiation-induced chemical changes in long-chain polymers as in Section 3.3 below, an alternative definition of G-value has been proposed, i.e., the number of chemical bonds modified per 100 eV absorbed.[20])

3.1.1.2 Radiation-Induced Release of Ions and Liquids; Radiolytic Corrosion

As a result of irradiation of ion-exchange resins, it is found that soluble acidic or basic species are produced and exchangeable counterions are released or leached, effects attributed to radiolytic attack on the resin functional group. For the cation resins, it is found that the pH of the supernate resulting from centrifuging a slurry of the resin in deionized water decreases with increasing radiation dose from 10^7 to 10^9 rad and is not particularly sensitive to the cationic species loading the resin (at least for the following cations: H^+, Na^+, Fe^{2+}, Fe^{3+}, or NH_4^+). For the anion resins, it is noted that at doses approaching 3×10^8 rad the resins lose their ability to retain water so that free liquid is produced. For anion resin in the Cl^- form, the pH of this free liquid and of the centrifugation supernate is neutral to slightly acidic after irradiation. For resins in the OH^- form, free liquid and supernate pH values are found to be more basic than those of the Cl^- form. It is estimated that about 60% of the moisture originally present in the dewatered Cl^- resin has been released as free liquid at 5×10^8 rad. Mixed-bed resins also exhibit free liquid release upon irradiation. The acidity of the free liquid and supernate solutions from mixed bed resins increases with irradiation dose, indicating that the acid species produced by radiolytic decomposition of the cation component are not completely neutralized by the basic species produced by the anion component.(17)

The release of liquid is not merely an aging effect since the free liquid is not reported for the loaded sample tubes set aside as zero-dose control samples. It has been suggested by Swyler et al.(17) that the release of free liquid is related to the radiolytic scission or loss of ion-exchange functional groups, since the moisture content of the resin generally depends on the number of intact functional groups. Alternatively, the decrease in moisture retention has been attributed to changes in resin cross-linking which result from exposure to γ radiation. Swyler et al. estimate a free liquid release of 14 volume percent at 100 Mrad for NaCl form mixed bed resin or "more than 20 times the amount presently considered initially acceptable in a high integrity container at burial."

Experiments on radiation-induced corrosion have been carried out in order to provide a basis for estimating the lifetime of mild steel containers for dewatered resin waste. The general method used is to irradiate resins in contact with metal specimens to various doses and then examine the specimens and determine their weight loss. It is found that corrosion rates depend both on resin functionality and on resin loading. Corrosion weight loss is greatest in the H^+ form cation resin and lowest in the OH^--form anion resin, but test coupons of mild steel immersed in unirradiated H^+-form cation resin also exhibit some loss. (A corrosion rate of about 0.4 mils per year for mild steel immersed in H^+ form cation resin may be obtained from the data.) The corrosive attack on mild steel can be correlated with the radiolytic formation of acidic species in the resins. For a given irradiation dose, corrosion is greater at low dose rates, probably reflecting the longer contact times per unit dose.(18) These results are in accord with those from an earlier study by Marek and Rinker(15), who found that irradiated anion resin samples in

contact with Type 304 stainless steel caused little if any attack on the steel, but that irradiated cation resin samples and their decomposition products caused serious deterioration of the stainless steel, the deterioration generally increasing with increasing radiation dose. The high doses (about 10^8 rad) at which radiation-induced corrosion effects are likely to be a concern should not occur during the extended storage of Class A resins but could accumulate during the extended storage of Class C resins. Therefore, radiation-induced corrosion effects may need to be considered if Class C resins are to be stored in stainless steel HICs for an extended period.

3.1.1.3 Radiolytic Gas Generation and Radiation-Enhanced Oxidation

The principal radiolytic gas generated from these resins when gamma-irradiated in sealed glass tubes is hydrogen. The yield of radiolytic hydrogen from a mixed bed resin may be approximated by the sum of the linearly scaled yields of the individual component resins. This is not true for all radiolytically generated gases. A second major radiolytic gaseous product (except from OH^- form anion resin) is carbon dioxide. Radiolytic methane formation is observed from irradiated anion and mixed-bed resins and trimethylamine formation is observed from irradiated mixed-bed resins. Also, oxygen is rapidly depleted from the atmosphere over irradiated resins. In the resulting anoxic environment, neither radiolytic scission of the cation resin functional group (as measured by the production of sulfite ion) nor radiolytic generation of hydrogen gas from cation resins exhibit strong dependence on the radiation dose rate. The G-values for the radiolytic generation of hydrogen gas from fully swollen sulfonic acid resins are found to range from 0.1 to 0.6.[17]

"Dewatered" ion-exchange resins still contain water, which may be internally absorbed within the resin, adsorbed on the surface of resin particles, or held in the interstices between resin particles.[4] The radiolysis of this remaining water and of the resin itself, as well as the subsequent reactions of the products of radiolysis must all be considered in addressing the overall effects of radiation on "dewatered" ion-exchange resins. The effects of ionizing radiation upon water include the production of a variety of ions, radicals, solvated electrons, and excited species.[21] The yields of these various species may depend on the pH, the presence of scavenging (i.e., reactive) solutes, and, because of their different modes of energy deposition in solution, the linear energy transfer characteristics of the different types of radiation. In general, the major stable products of the radiolysis of pure water are molecular hydrogen (H_2) and hydrogen peroxide (H_2O_2).[21] The overall reaction for the radiolytic decomposition of water may be written

$$2H_2O \rightarrow H_2O_2 + H_2.$$

The intermediate reactions and the various intermediate species are described in the review by Glass[21] and in the monograph by Draganic and Draganic.[22] In summary, for every two moles of hydrogen gas attributed to the radiolysis of water there will indeed be one mole of oxygen, but this radiolytically generated oxygen may not necessarily exist as free oxygen gas

(O_2). Note that H_2O_2 will, under appropriate conditions, decompose to yield free oxygen gas and water by the following reaction

$$2H_2O_2 \rightarrow 2H_2O + O_2.$$

Radiolytic generation of both hydrogen and oxygen was observed from spent inorganic ion-exchanger (zeolite) beds used in the Submerged Demineralizer System (SDS) to remove fission products, principally Cs-137 and Sr-90, from water contaminated as a result of the accident at Three Mile Island Unit 2 (TMI-2).[23] From the gas generation data, it was concluded that:

- The gas generation rate was proportional to the curie loading and was approximately 0.01 cc/Ci-h.
- The gas generation rate showed no sign of decreasing with increasing gas pressure. No approach toward equilibrium was observed.
- Stoichiometric gas mixtures did not immediately evolve in the vessels. The hydrogen/oxygen ratio of the resulting gas mixture was higher than stoichiometric but approaching stoichiometric with time. (Oxygen was being adsorbed or chemically bound.)

(Although the gas generation rates in SDS vessels loaded with more than 15,000 Ci were sufficient to result in flammable gas mixtures by the end of a 14-day test period, the specific activity within the SDS vessels -- each of which has an internal volume of about 10 ft^3 or 270 L -- is much higher than that expected for typical dewatered reactor resins. Note that the NRC Technical Position on Waste Form specifies a total accumulated dose to the resins of 10^8 rad, which is stated to be equivalent to a 10-Ci/ft^3 loading of Cs-137 and Sr-90. The SDS is discussed here as an example of radiolytic generation of both hydrogen and oxygen.) The accumulated gas consisted primarily of hydrogen in one SDS vessel which was found to contain a scrap of plastic, a pipe-end cover. It appeared that in the presence of the radiation field, this organic material had acted as an effective scavenger preventing the formation and/or liberation of radiolytically generated oxygen. This radiolytic oxygen scavenging by organic materials is consistent with observations[24] of radiolytic hydrogen production and oxygen scavenging in the Epicor-II prefilter liners from the TMI-2 cleanup, which contain predominantly organic ion-exchange resins. The liner activities ranged from 166 to 2184 Ci. Each liner contained approximately 30 ft^3 of ion-exchange resins. The following observations were made regarding the radiolytic gas generation from the Epicor-II pre-filter liners:

- All liners showed significant oxygen deficiency, generally less than 0.2% oxygen upon opening.
- Upon opening, two liners were found to be at less than atmospheric pressure, which, it is postulated, was due to insufficient generation of hydrogen and carbon dioxide to replenish the volume of oxygen depleted.

- In addition to H_2, O_2, and N_2, all liners contained CO_2 and trace amounts of CO and CH_4.

- The average hydrogen production rate for the Epicor pre-filters is 5.94×10^{-6} L/Ci-h.

Only one of the 18 Epicor-II pre-filter liners in this study was found when opened to have a hydrogen-to-oxygen ratio less than stoichiometric based on the decomposition of water (i.e., 2 to 1); the gas compositions upon opening the remaining 17 liners were found to be deficient in oxygen. (It should be noted that atmospheric oxygen was very likely present initially in these liners before sealing and that subsequent stoichiometric production of hydrogen and oxygen gases from the radiolysis of water with no accompanying oxygen depletion would result in an excess of oxygen upon opening of the liner.)

Swyler et al.[17] have irradiated ion-exchange resins for 1030 h at 4×10^4 and 1×10^5 rad/h in closed systems. These workers have correlated the radiolytic production of CO_2 with the depletion of the atmospheric oxygen present before irradiation of the resin sample. They postulate that the oxygen could be depleted by a radiation-mediated reaction with the resin to produce CO_2. However, at a higher dose rate or greater dose -- 1.6×10^6 rad/h for 1030 h -- additional CO_2 is generated which is attributed to some unspecified mechanism involving oxygen initially present either in the resin itself or in the incorporated water. Also, CO_2 evolution has been observed as a result of the irradiation of resins in the absence of atmospheric oxygen. In summary, the production of radiolytically-generated carbon dioxide may be attributed to a variety of mechanisms; attack on the resin by oxygen resulting from the radiolysis of water is not the only possible mechanism.

The production of trimethylamine is attributed by Swyler et al.[17] to the radiolytic scission of quaternary ammonium functional group by reactions such as

$$Ar\text{-}CH_2\text{-}N(CH_3)_3{}^+OH^- \rightarrow Ar\text{-}CH_2OH + N(CH_3)_3,$$

where Ar represents an aromatic moiety. No mechanism is postulated by these authors for the production of methane, but it would probably result from either radiolytic scission of the functional group or radiolytic attack on the polymer backbone of the resin.

The above observations of the radiolytic decrease in pH and of oxygen scavenging refer to the irradiation of resins in sealed systems. The data from irradiations carried out in various mixtures of air, helium, and oxygen do not indicate any pronounced variations in hydrogen gas generation from H^+ form resins with atmospheric conditions.[17] Further experiments have been carried out under vented (oxic) conditions -- i.e., with the irradiation chamber open to the atmosphere -- in order to examine the effect of radiolytic oxidation on resin degradation. It is found that the pH of the supernate from samples of resins in the Na^+ or H^+ form irradiated in a vented system is

significantly lower than that from a closed system (e.g., a pH of 1.6 as compared with a pH of 2.1, in both cases for 10 mL of liquid in contact with 2 g of resin irradiated to about 10^9 rad).

These pH values were measured for the deionized water with which the resins were placed in contact in the resin-to-water ratio of 2 g/10 mL. These pH values are not meant to be representative of field conditions but are merely a measure of the extent of radiolytic degradation of the resin. Swyler et al.[17] suggested that the lower pH observed for test systems open to the atmosphere may be attributed to soluble organic acids (e.g., benzylsulfonic acid) produced by the radiolytic scission of the resin backbone. There is also visual evidence for radiation-induced agglomeration. These observations are attributed to an extensive attack on the resin polymer backbone resulting from radiolytic oxidation.[18]

3.1.1.4 Radiation Effects on Resin-Cement Composites

Irradiation of the resins before their incorporation into a cement matrix is found to increase the resistance to cracking when the resulting waste form is immersed in water and to improve the compressive strength of the waste form. For irradiated resin, solidification is possible at increased waste-to-cement ratios.[17] It has also been reported that irradiation of resin/cement composites following solidification can improve resistance to cracking upon immersion in water.[25] However, resin-cement waste forms containing the irradiated resins exhibit an increased leachability for Cs.[17] In other work at BNL, no significant degradation of either the mechanical integrity or of the leachability of composites fabricated from cement and an irradiated proprietary ion-exchange material was found by Barletta et al.[16]

More specifically, as reported in Reference 17, cylindrical resin-cement forms (2 in. diameter x 4-in. high) were cast containing either irradiated (9×10^8 rad at 1.65×10^8 rad/h) or unirradiated sulfonic acid cation resin (Rohm and Haas, Amberlite IRN-77) in the Na^+ form. The cement/water ratio was 2.5. In Table 3.3, the compressive strengths (if applicable) are presented for several resin-cement formulations after 28 days of curing and 28 days of immersion in water. Note that for the two formulations with higher resin/cement ratio, the samples failed by cracking during the 28-day immersion period.

As reported in Reference 17, cement/mixed-bed-resin waste forms using a formulation described only as one "that passed initial acceptability criteria, but did not pass the 2-week water immersion test" were irradiated after two weeks of curing to total doses of 0 (control), 10^4, 10^6, and 10^8 rad at 1.5×10^6 rad/h. No difference in behavior was evident between the control specimens and those irradiated to 10^4 rad; both failed the immersion test. The 10^6 rad specimens exhibited better integrity but also failed the immersion. Only the 10^8 rad specimens passed the immersion test. The retention of integrity after immersion of the specimens irradiated to 10^8 rad was attributed either to increased cross-linking of the resin, thereby reducing its ability

to shrink and swell, or to the reduction of the number of functional exchange sites on the resin, limiting its capacity for ionic sorption. (It is not clear how the latter mechanism could contribute to radiation-induced resistance to immersion.)

Table 3.3

Compression Test Data on Resin-Cement Waste Forms After Immersion in Water[a]

Resin/(Cement + Water)	Compressive Strength (psi) Unirradiated	Irradiated
0.07	not reported	2.7×10^3
0.14	2.1×10^3	2.2×10^3
0.29	failed	1.5×10^3
0.34	failed	1.1×10^3

[a]Information from NUREG/CR-3383, Reference 17.

The proprietary ion-exchange material discussed in Reference 16, called "D-mix," was said to be representative of the first-stage liner (pre-filter) material used in the Epicor-II demineralization system to decontaminate water from the accident at Three Mile Island Unit 2. The test samples consisted of a mixture of D-mix, water, Portland Type I cement, and radium metasilicate in a weight ratio of 350:153:514:51. The samples were allowed to cure in plastic containers for 9 days and then stored uncovered at room temperature for an additional 10 days. The average fracture tensile strength for test samples made with unirradiated D-mix was about 1.0 to 1.4 MPa. For D-mix which had been irradiated to 10^8 or 10^9 rad before incorporation into the cement forms, the average fracture tensile strength was about 2.6 and 3.2 MPa, respectively. The dose rates in this study ranged from 2.9×10^6 to 5.2×10^6 rad/h. D-mix/cement composites irradiated to these doses after solidification exhibited no significant changes in fracture tensile strength.

It appears that irradiation of ion-exchange resins either before or after their incorporation into cement waste forms will improve the resistance to water immersion of the waste forms.

3.1.1.5 Biodegradation Effects in Ion-Exchange Resins

In addition to radiolytic degradation of ion-exchange resins, biodegradation has been considered as a mechanism for changes in properties and behavior of these materials. For example, occasional difficulties have been encountered in attempting to transfer spent ion-exchange resins as a slurry from holding tanks after the resins have been in storage for some time.[26]

Although radiolytic agglomeration of the resins may be a contributing factor, in at least one case mold or bacteria growing in the resin interfered with pumping out the spent resin tank; after the addition of chlorine as a biocide, the resin was reportedly pumped out with ease. Gas buildup in dewatered spent resins, attributed to biodegradative action, has also been reported at one power plant (Duane Arnold); it was found that addition of formaldehyde prevented the gas buildup.[10,12] Biologic activity may have contributed to an exothermic reaction noted in spent ion-exchange resins at Arkansas Nuclear One, which occurred during dewatering after difficulties encountered in transferring the resins from a holding tank.[27] Details regarding the possible biodegradation of spent nuclear reactor ion-exchange resins are reported elsewhere.[27]

3.1.2 Other LWR Process Wastes

Concentrated liquid waste produced in LWRs by the evaporation or crystallization of a wide variety of liquids (Table 3.4) consists of liquids with an elevated suspended and dissolved solids content and also of sludges. Liquids containing sodium sulfate result from demineralizer regeneration primarily in BWRs, but also in PWRs. Borate waste solutions result from the boric acid used in the primary coolant system of PWRs. The typical solids contents for these wastes is 8 to 25 weight percent and 5 to 12 weight percent, respectively, when evaporators are used. For crystallizer/evaporators, these wastes may contain up to 50 weight percent solids, much of it suspended rather than dissolved. Concentrated liquid LLRW is generally incorporated into a solid matrix material for disposal. The activity of concentrated liquid waste averages about 0.03 Ci/ft^3.[3,4]

Filter sludge is produced by precoat filters and consists of filter aids and waste solids (crud) retained by the filter aids, which commonly consist of diatomaceous earth, high-purity cellulose, or powdered ion-exchange resins. These materials are deposited in a thin cake (precoat) on a filter medium such as wire mesh to remove suspended solids (and in the case of the resins, some dissolved ions) from liquids. Diatomaceous earth is composed of particles of silicon dioxide usually smaller than 50 μm and has a dry bulk density of about 0.16 g/cm^3. Cellulose fibers are made from plant material and because of air pockets range in apparent wet density from 0.13 to 0.34 g/cm^3. Both diatomaceous earth and cellulose filter aids can contain 50 weight percent water after "dewatering" and crud loadings of 5 to 10 weight percent. Powdered resins, currently used as filter aids in BWRs, are chemically similar to the bead resins discussed above but consist of particles with linear dimensions of about 0.044 mm, about one tenth that typical for resin beads. Spent powdered resins contain more crud contaminants (>5 weight percent) than bead resins because of their filtration effect. Filter aids are either incorporated into the standard binder materials or dewatered and placed in high-integrity containers. The approximate average activity loadings for PWR and BWR filter sludges has been given as 0.1 and 0.2 Ci/ft^3, respectively.[3,4]

Filter cartridges are disposable filter elements typically constructed of woven or wound fabric (cotton or nylon) or of pleated or matted paper

supported by a steel mesh or basket. Paper filter elements are often impregnated with epoxy and are usually packed for disposal in 55-gallon drums, 3 to 12 cartridges per drum. The average activity loading for spent filter cartridges has been given as approximately 0.3 Ci/ft^3.[3,4]

Table 3.4

Major Sources of Process Wastes in LWRs[a]

BWRs	PWRs
1. Application of Ion-Exchange Resins	
Condensate polishing system	Condensate polishing system
Reactor water cleanup	Chemical and volume control syste
Clean radwaste system	Boron control system
Dirty radwaste system	Spent fuel pool cleanup
Chemical waste system	Steam generator blowdown system
Spent fuel pool cleanup	Miscellaneous waste system
	Chemical waste system
2. Sources of Liquids Concentrated by Evaporation	
Regeneration of resins	Regeneration of resins
General decontamination waste liquids	General decontamination waste liquids
System effluents from:	System effluents from:
Clean radwaste	Liquid radwaste
Dirty radwaste	Chemical radwaste
Chemical radwaste	Laundry waste
Laundry waste	Steam generator blowdown
3. Application of Precoat Filters and Cartridge Filters	
Condensate polishing system	Steam generator blowdown
Reactor water cleanup	Condensate polishing system
Spent-fuel pool cleanup	Boron control system
Equipment and floor drains	Spent-fuel pool cleanup
Chemical waste system	Laundry waste system
Laundry waste system	

[a]Reference 3.

There is some information available on the radiolytic properties of cellulosics, such as the cellulose filter aids and the paper or fabric cartridge filter elements. In an early study at ORNL, Bopp and Sisman[28] report G-values for gas generation ranging from 0.5 to 3.0 for a variety of cellulosic materials. The irradiations were carried out by exposing samples of the materials sealed in glass capsules to neutron, β and γ radiation in the maximum-flux region of a graphite reactor. On the basis of comparative

studies made with γ radiation from Co-60 and Au-198, these authors state that qualitatively equivalent and, within a factor of 2, quantitatively equivalent changes are produced in many properties of the irradiated materials for the same energy absorption by both types of radiation. More recently, Bibler[29] reported G-values for the generation of gas from cellulosics exposed to radiation from Cm-244 to be independent of dose rate but to vary with dose, ranging from about 1.9 at 2×10^8 rad to about 0.6 at 4×10^9 rad. During irradiation, oxygen was consumed and hydrogen, carbon dioxide, and carbon monoxide were produced in a ratio of 1.0: 0.7: 0.3. It is not clear, however, whether these results for α radiation, a high linear energy transfer (L T) radiation, may be extended to low LET radiations such as β or γ. In the radiolysis of some organic compounds, only a slight effect of the LET character of the radiation has been observed, but in other cases, effects on the quantities and on the species of the radiolysis products have been noted.[30] Also, the effects of an oxidizing environment (which may be maintained by means of an irradiation chamber kept open to the atmosphere) on the dose rate dependence and on the nature of the radiolysis products is unclear. Finally, the total accumulated dose to filter sludges and cartridge filter elements may be inferred from the activity loadings for these wastes given above. Using the 10^8-rad dose to ion-exchange resins noted above for activity loadings of 10 Ci/ft^3 and assuming linearity of the dose-to-loading relationship, doses of 10^6 to 3×10^6 rad may be calculated for these wastes. Among the other products reported from the radiolysis of cellulosics are various organic acids: formic (64%), glucuronic, 2-ketohexanoic, and 3 unspecified "5-ketohexanoic or uronic acids."[31] The G-value for the overall production from cellulosic materials of organic molecules containing carboxylic acid groups is 3.6, with the G-value for the production of formic acid alone being 2.3. There is a potential for corrosion of container materials such as carbon steel and of binder materials such as cement by these compounds. Also, the radiolytic formation of organic acids may provide a reaction pathway for radiolytic oxygen depletion.

3.2 Binders and Solidification Agents

A solidification agent or binder is a material that when mixed in prescribed proportions with liquid or slurry waste can form a free-standing monolithic product with no free liquid.[32] A monolithic solid waste form may be produced by 1) reaction of the solidification agent with the waste, 2) encapsulation of waste droplets or particles in microscopic cells dispersed through a solid matrix, or 3) coating individual particles of dry waste with an adhesive layer. The main reason for solidifying LLRW has been, until recently, to satisfy regulatory requirements, but the availability of solidification processes (e.g., bituminization) that also reduce the volume of the waste have added economic incentives by reducing disposal costs.[4] Stabilization, i.e., solidification or the use of high-integrity containers, is required only for Class B and Class C wastes. Many nuclear power plants have been solidifying their wastes for some time.

The current state of the art in radwaste solidification technology and commercially available solidification systems are described in a report[4]

published by the Electric Power Research Institute (EPRI). The following solidification agents were specifically listed as acceptable at the three operating commercial burial sites:

- cement (including proprietary and trademarked binders such as Delaware Custom Materiel Media and Envirostone™)
- asphalt or bitumen
- Dow vinyl ester-styrene (a thermosetting organic polymer)
- any solidification agent found suitable by NRC

The NRC is currently reviewing topical reports submitted by vendors on generic versions of these agents. New binder materials are being tested and submitted for approval. Most of these are thermosetting polymers and include polyesters and epoxies. Individual solidification agents are considered under separate headings in this section.

3.2.1 Cement

In the United States cement has been used as a solidification agent for radioactive waste since the inception of the nuclear industry[33] and is still the most common solidification agent used at nuclear utilities.[34] According to a survey sponsored by the Electric Power Research Institute (EPRI) in 1981,[34] 23 of 33 nuclear power stations that were solidifying their waste used cement as the solidification agent. With suitable additives,cement may be used as the solidification agent for a wide range of LLRW, including spent resins and filter aid materials, boric acid waste, and sodium sulfate solutions.[4,35] Cement systems have been favored in part because the water in liquid radwastes is chemically reacted during the solidification process.[36] The physics and chemistry of the solidification process and the effects of the incorporated wastes on the properties of the final solidified waste form are discussed elsewhere.[4,35,37] In the discussion of the properties of cement in this report the emphasis will be on the behavior of cement-solidified LLRW during storage and the effect of extended storage on the properties of the waste after disposal.

Several different kinds of cement have been used for solidifying radwaste. Portland cement, which with various additives is the kind most commonly used for this purpose, is composed mainly of silica (SiO_2), lime (CaO),and alumina (Al_2O_3) with small quantities of other oxides (MgO, Fe_2O_3, SO_3). Other kinds of cement approved for use as LLRW binders by the commercial burial site operators include masonry cement (high-lime cement), Envirostone™ (trade name for a gypsum-based cement), and Delaware Custom Materiel proprietary solidification agent (consisting of sodium silicate, a setting agent such as Portland cement, and, when required, finely divided shale as a cesium scavenger to reduce cesium leachability).[4] Properties of typical cements used for solidification of LLRW are presented in Table 3.5.

Table 3.5

Selected Properties of Cements Used for Solidifying LLRW

Density, g/cm^3		
Portland cement:	bulk, dry[a]	1.5
	as solidification matrix [b,c]	1.5 to 2.5
	maximum theoretical[a]	3.1
Radiation resistance, rad[c,d]		$>10^{10}$
Compressive strength, MPa (psi)[b]		100 (14,500)
Maximum stable temp, °C[c]		300 to 500 (dehydrates)
Thermal conductivity, W/m°C[b]		0.8 to 1.7
Dry waste solids content, weight percent[c]		5 to 55
Volumetric ratio, product/liquid waste[c]		1.5 to 3.0

[a]Reference 4.
[b]Reference 37.
[c]Reference 38.
[d]The terms "irradiation stability" and "radiation resistance" are used in References 37 and 38, respectively, without explicit definition, and seem to refer to retention of the mechanical integrity (as indicated by the compressive strength and of low leachability after irradiation).

Cement as a binder material is expected to show good radiation resistance. Work at both SRL(39) and BNL(16) has shown no indication of deleterious effects on either the mechanical integrity or the leachability of composites of cement and simulated waste, even at doses of 10^9 or 10^{10} rad. In other work at SRL,(40) the production of gas (mostly hydrogen) was observed during the gamma irradiation of sealed canisters of cement containing Fe_2O_3 to simulate Savannah River Plant sludge. The equilibrium pressure of H_2 gas which was produced depended upon the dose rate. Also, oxygen initially present in the canister was consumed. It was hypothesized that atomic hydrogen (radiolytically generated from the water in the cement) reacted with itself to form molecular hydrogen, with hydroxyl radicals (also radiolytically generated) to form water, and with molecular oxygen to form hydrogen peroxide. Another reaction pathway, the catalytic recombination of H_2 and O_2 to form water has been proposed.(41) From the SRL data the equilibrium radiolytic gas pressure in cement is about 10 psi at 10^5 rad/h and, by extrapolation, about 4 psi at 10^4 rad/h.

Extended storage of cement-solidified LLRW will subject the waste form to a dry period of perhaps several years before any possible exposure in the disposal trench to groundwater, a potential leachant. Thus, any effects of extended dry periods on the subsequent leaching properties of cement waste forms are of interest. The extended dry periods are, in effect, extended dry curing times for the cement waste forms. In a study of leaching from hydrofracture grout (a modified cement), Moore et al.[42] found that for leaching of strontium the cumulative fraction released (CFR) decreased by over an order of magnitude as the curing time was increased from 0 to 28 days. For cesium, on the other hand, the CFR decreased by only about 30% as the curing period was increased from 4 to 52 weeks. The two sets of data are not quite comparable, however, because of the different time spans. In more recent work at BNL, Dayal et al.[43] have also found that processes occurring during the unsaturated "dry" period of wet/dry cycling can have a significant effect on the release of strontium but a relatively small effect on the release of cesium. The BNL workers observed that Cs release is governed primarily by the length of the saturated period and that unsaturated periods of up to six days have no significant effect on Cs release. On the other hand, a systematic trend of decreasing Sr release with increasing length of the "dry" period was evident, with more than an order-of-magnitude decrease in the CFR as the length of the dry period was increased from one to six days. This decrease in the leachability of Sr as a result of dry curing is attributed to the ability of the Sr to substitute for Ca in the cement matrix to form relatively insoluble compounds. The effects of a humid vs a dry atmosphere during the curing period on Cs leachability from a hydrofracture grout have been noted by Moore et al.[42] Specimens allowed to dry during curing periods of 28, 56, and 112 days exhibited a release of Cs approximately 100% higher during the first 10 to 15 days of leaching than specimens cured in a humid atmosphere for comparable periods. After this initial period Cs was leached from the two sets of specimens at approximately equal rates.

The hygroscopic swelling of resins incorporated into cement can also result in swelling, cracking, and disintegration of the cement waste form,[8] -- see the discussion in Section 3.1 above on the immersion stability of cement-solidified ion-exchange resins -- but the swelling of cemented wastes may result from properties of the cement matrix itself. As noted by Lea,[44] a set cement undergoes an irreversible decrease in volume on drying for the first time. On each further wetting and drying cycle to which the cement is subjected, a reversible swelling and shrinking cycle may be observed. The cement never recovers its initial volume, however. A quantitative example of such size changes is given by Lea for 1- to 2-mm-thick slabs of a set Portland cement moist cured for 8 months with a water/cement ratio of 0.6; namely, on the first drying from 100% to 10% relative humidity an initial irreversible linear shrinkage of about 0.6% is observed, but during successive wet-dry cycles, the length of the specimen is observed to vary between the following percentages of its original value before its initial irreversible shrinking: 99.4% and 99.6% at relative humidities of 10% and 100%, respectively. In other words, after the first irreversible shrinkage on the initial drying of a set cement specimen, increasing the relative humidity from 10% to

100% results in a 0.2% increase in length of the specimen (since 99.6/99.4 = 1.002) probably due to absorption of water by the cement from the atmosphere. Lea cautions that although the numerical values apply only to the particular system of neat cement measured, the general behavior is typical also of mortars and concretes, as well as cements, although the actual values are considerably lower for mortars and concretes.

In certain extended storage environments (e.g., outdoors or in simple unheated structures), relatively severe temperature fluctuations may have detrimental effects on cement. As noted by Lea[44], in wet cement the water enclosed in the pores of the material will, on freezing, expand and thereby set up severe internal stresses and force the particles of mortar apart. Freezing and thawing will have no significantly damaging effects on any cement not sufficiently saturated with water. (The quantitative meaning of "sufficiently saturated" is very complex because of the different ways water may be incorporated into cement — e.g., water of hydration, pore water, and capillary water -- and will not be discussed here. See Lea,[44] pp. 611 to 621.)

The resistance of concrete to weathering is an important consideration for outdoor storage of concrete containers or casks, particularly in severe climates. (An example of outdoor storage of radwaste in concrete containers is described above in Section 2.2.4.1.) Various accelerated tests, usually involving freeze-thaw cycling, have been devised to simulate weathering action[45] and attempts have been made to extrapolate the durability of constructional concrete from the behavior of cementing agents occurring in natural deposits and in man-made objects.[46] Increasing the resistance of concrete to freeze-thaw cycling by techniques such as air entrainment is discussed in the following paragraph. In addition to temperature fluctuations, phenomena such as acid precipitation, dry deposition of the oxides of sulfur and nitrogen, and particulate emissions may all contribute to the erosion of concrete structures[47] and should be taken into account when estimating the lifetimes of concrete containers for the storage of LLRW, especially if such containers are to be stored outdoors. Quantitative data for the erosion of concrete by atmospheric constituents are not readily available and, in any case, should be obtained for the erosion of the particular composition of concrete used in the storage containers by the potentially corrosive atmospheric constituents expected in the storage environment.

As discussed by Lea,[44] Troxell et al.,[45] and Woods,[48] the freeze-thaw resistance of concrete may be improved by air entrainment, which may be accomplished by means of an appropriate additive, usually a surface-active organic chemical having the property of stabilizing foams. As a result, the quantity of very small bubbles (0.001 to 0.01 in.) in cement is increased. These bubbles do not readily fill with water and thus reduce the degree of water saturation of the cement. The air entrainment additives thus also function as water reduction additives. The bubbles also provide void spaces which tend to relieve any forces which may develop from the growth of ice crystals in the concrete.

It should also be noted that some of the products of waste radiolysis such as formic acid from the radiolysis of cellulosics may be corrosive to cement. The quantitative effect of low-molecular-weight organic acids on cement does not seem to have been documented but acids such as acetic, citric, malic, and lactic, have been found to attack concrete, often having "a marked action" within a few months to a year.[44] When compared with acetic acid in its corrosive effect on cement, formic acid has been described both as acting more slowly[44] and as being more destructive.[49]

3.2.2 Bitumen

The bitumens or asphalts are a class of substances composed primarily of high-molecular-weight hydrocarbons. They have been used for over 20 years in Europe as matrices for the immobilization of radwaste, but the first bituminization unit at a U.S. nuclear power plant (Consumers Power Company's Palisades Facility) began operation in 1982[50] after a relatively long history of investigations of radwaste bituminization at national laboratories (see a 1965 report[51] by workers from ORNL and PNL). Bitumen may be used for both volume reduction and immobilization of wet solid LLRW such as spent deep-bed demineralizer resin beads, powdered demineralizer resins, precoat filter media, and evaporator concentrates, the bitumen serving as a heat transfer medium as well as a matrix for immobilization of the waste.[52] The volume reduction results from the evaporation of water from the wet radwaste during the mixing of the heated liquefied bitumen and the waste. Waste is immobilized as the bitumen cools and solidifies, physically entrapping the waste. Dry active waste may also be immobilized in bitumen, but there is no accompanying volume reduction. Chemical fixation of the radwaste by the bitumen does not occur.[4] Bitumens, in any case, are generally resistant to many chemical reagents,[53] with some important exceptions such as nitric acid, nitrates, and nitrites.[54] The processes used in commercially available bituminization systems are described in various EPRI reports.[4,52,55] It has been noted[56] that although bitumens were the first binders to be used for the volume reduction of LLRW in Western Europe, their use is being curtailed because of concerns regarding potential fire hazards and radiolytic gas generation. The significance of these concerns for the extended storage of LLRW will be discussed later in this section and in Section 4.1.

Two types of bitumen frequently used for immobilization of radwaste are straight-run (or direct) distillation bitumen, which is essentially the residue remaining after the distillation of certain grades of crude petroleum, and air-blown (or oxidized) bitumen, which is a highly colloidal bitumen product formed by blowing air through certain petroleums. Typical data for these bitumens are presented in Table 3.6.

Certain characteristic temperatures important for the bituminization process, such as the softening point, flash point, and ignition point, are given in Table 3.6. These temperatures are also important considerations for the storage of bituminized wastes. Bitumens do not exhibit a sharp solid-to-liquid transition at a definite temperature but gradually become softer and less viscous as the temperature increases. Thus, to compare the melting

properties of bitumens, test methods (for example, ASTM D-36-76[58] and D2348-76[59]) have been developed for determining a softening point temperature under arbitrary but well-defined conditions. Bituminized waste forms retain considerable plasticity, tending to flow even at room temperature and must be stored in containers to maintain their shape. The flash point of a material, the minimum temperature at which the vapors above the material can be ignited by a flame, is different from and lower than the ignition or fire point, the minimum temperature at which the material itself can sustain combustion.[4] Because of the low thermal conductivity of the bitumen (0.15 W/m°C), vaporization of the bitumen is insufficient to allow continued burning of the product.[37] Standard test methods have been described for obtaining values of the flash and fire points.[60] It should be noted that shipments of bitumen obtained from the same source may vary greatly in their physical properties and composition.[57]

Table 3.6

Selected Properties of Bitumen Used for Solidifying LLRW

Density, g/cm^3[a,b,c]	1.0 to 1.9
Radiation resistance, rad[b,c,d]	$\approx 10^9$
Softening point, °C	
typical temperature range[b]	40 to 80
Witco Chemical's Pioneer 221 asphalt[a]	93
straight-run distillation bitumens[e]	34 to 65
oxidized bitumens[e]	70 to 140
Flash point, °C	
typical temperature range[b]	290 to 350
Witco Chemical's Pioneer 221 asphalt[a]	288
straight-run distillation bitumens[e]	230 to 250
Ignition point, °C	
typical temperature range[b]	350 to 450
Witco Chemical's Pioneer 221 asphalt	316
Thermal conductivity, W/m°C[b]	0.15
Dry waste solids content, weight percent[c]	40 to 60
Volumetric ratio, product/liquid waste[c]	0.2 to 1.1

[a]Reference 4.
[b]Reference 37.
[c]Reference 38.
[d]See Table 3.5, note d.
[e]Reference 57.

The effects of ionizing radiation on the properties and behavior of bitumen constitute a major area of concern regarding the use of this material as a matrix for the immobilization of radwaste. Such effects have been reviewed in a recent BNL report[61] and are only briefly surveyed here. No systematic evidence has been found to indicate that leach rates of bituminized waste are significantly affected by irradiation doses of up to 10^8 rad, but for certain bitumen-radwaste formulations doses $<10^8$ rad have affected mechanical properties by causing the material to swell and/or harden. Radiolytic gas generation, however, is probably the most important irradiation effect observed in the behavior of bituminized waste during storage. From studies in the USSR, France, and Germany, it appears that both the dose rate and the presence of oxygen in the bitumen matrix may significantly affect the rate of radiolytic gas generation. In the Russian studies there is evidence of radiolytic oxygen scavenging, manifested by an initial decrease in air pressure in the sample chamber followed by an increase in pressure as radiolytic gas generation becomes the dominant process. This is similar to the radiolytic oxygen scavenging discussed earlier in connection with ion-exchange resins and cement. The Russian findings, which indicate that the gas generation for a given dose varies inversely as the dose rate, are in disagreement with the French results, which indicate that swelling produced at a given dose decreases with decreasing dose rate, but the Russian results have been viewed with some skepticism because of unusually large G-values (approaching 1000) which may be estimated from their data.[61]

Oxidized bitumens, less sensitive than direct-distillation bitumens to ionizing radiation, have been recommended for the immobilization of radwaste with activities greater than 1000 Ci/m^3 (28 Ci/ft^3).[4] Workers at ORNL found that some bitumen samples exhibited a volume increase of up to 36% when irradiated to a dose of 10^8 rad. This swelling was attributed to internal pressurization by radiolytic gas generation.[51] In later work at ORNL, gas bubble formation was observed in bituminized waste forms that had received an internal dose of about 2×10^7 rad.[62] Measurements carried out at BNL on Pioneer 221 asphalts have yielded G-values for the generation of gas (mostly hydrogen) ranging from 0.1 at 10^7 rad to 0.43 at 10^9 rad.[63] Similar measurements in Germany have resulted in G-values for gas generation from bitumen of about 0.2 at a dose of 10^8 rad in one investigation[64] and about 0.4 or 0.5 at doses from 10^6 to 10^8 rad in another study.[65] The gas evolved is predominantly hydrogen (typically >95%); the other minor components have been identified in at least one instance as light hydrocarbons.[65]

Another concern regarding the use of bitumen is its potential for biodegradability. In a study* for the Swedish Nuclear Fuel Supply Corporation of microbial processes likely to occur in bitumen, Roffey and Hjalmarsson note that microbial generation of CO_2 in bitumen may result in pressure buildup and

*R. Roffey and K. Hjalmarsson, "Mircobial Processes in the Final Repository, the Silo Part. Theoretical Approach and Preliminary Experiments on the Biodegradation of Bitumen, Part I. Defense Research Institute, Umea, Sweden, FOA Report C40172-B4, May 1983.

corrosion of metal although, in experiments, they observed only aerobic microbial production of an unidentified gas. On the other hand, it has been concluded from other work that biodegradative processes and irradiation damage may be of potential significance for bitumized waste. In a review of microbial degradation of bitumens by SNL,[53] nearly one hundred different kinds of asphalts have been found to be degraded by microorganisms. For example, microbial oxidation rates ranging from about 5 to 75 g/cm^2 over an eight-week period have been reported for asphalt in the presence of a medium containing moisture, minerals, and free oxygen. Microbial degradation of bitumen may affect nearby materials; for example, the corrosion of metal pipes and cables coated with asphalt has been attributed to biodegradation of the asphalt. Details of a recent experimental study by BNL of biodegradation testing of bitumen are reported elsewhere.[2]

3.2.3 Thermosetting Organic Polymers

Binders consisting of thermosetting organic polymers are relatively new in the management of LLRW. Depending on the particular binder material, solidification is accomplished by catalytic or heat-induced polymerization, as a result of which the molecules of the binder material are linked in an intricate network that physically entraps the waste. Before solidification, the radioactive waste in solution, slurry or powder form is dispersed through the binder material by thorough mixing so that after polymerization discrete droplets or particulates of the waste are physically encapsulated and thus immobilized within small pores uniformly distributed throughout a continuous polymer matrix. As with bitumen binder materials, there is no chemical fixation of the immobilized waste. The thermosetting polymer binders are relatively insensitive to the chemical composition of the waste and are thus compatible with a wide variety of LLRW.[4,33,37,38]

At least five types of thermosetting polymers are commercially available for immobilization of LLRW. A proprietary vinyl ester-styrene is being marketed by Dow, and the product has been tested at BNL.[66,67] In addition, three types of polyester materials and an epoxy solidification agent are available.[4] Some properties of these materials are presented in Table 3.7. The data on flammability, in particular, should be noted, since this class of binders, like the bitumens, consists of organic materials.

Thermal cycling has been found to affect the free liquid observed on the surface of vinyl ester-styrene waste forms. Droplets of a free liquid "sweat" were observed by Piciulo and Chan[67] during a study of the effects of thermal cycle testing on vinyl ester-styrene waste forms. These waste forms consisted of one of four LWR waste streams -- boric acid evaporation bottoms, sodium sulfate evaporation bottoms, bead resins, or powdered resins -- solidified in vinyl ester-styrene. The volume percent of free liquids on the thermally cycled samples was found to range between 0.7 and 1.3. The temperature extremes of the thermal cycle test were -40°C and +60°C. The control samples, which were not thermally cycled, were also found to exhibit free liquid but in smaller amounts, i.e., 0.1 to 0.4 volume percent. The pH of the liquid was found to be somewhat acidic, from 4.5 to 5.5, and thus

potentially corrosive to mild steel. Note that these data are from short-term small-scale testing and their applicability to full-scale waste forms is unclear. For example, if the observed release of free liquid is a near-surface phenomenon — i.e., the liquid originates only from a thin layer at the surface of the waste form — then because of the smaller surface-to-volume ratio of the full-scale waste form the volume percent of free liquid released from a full-scale waste form will be smaller than the volume of free liquid released from a small-scale laboratory waste form.

Table 3.7

Selected Properties of Thermosetting Organic Polymers

Density, g/cm^3[a,b,c]	
polyester	1.0 to 1.9
Dow binder	1.04
Radiation resistance, rad[d]	
polyester[b,c]	10^9
Dow binder[e]	$>10^8$
Compressive strength, MPa (psi)	
polyester[b]	100 (14,500)
Dow binder[f]	11.7 - 29.0 (1700 - 4200)
Maximum stable temp, °C	
polyester[c]	300 to 500 (chars)
Dow binder[f]	538 for ten minutes (chars)
Thermal conductivity, W/m°C	
polyester[b]	0.19
Dry waste solids content, weight percent	
polyester[c]	10 to 50
Volumetric ratio, product/liquid waste	
polyester[c]	1.4 to 1.5

[a]Reference 4.
[b]Reference 37.
[c]Reference 38.
[d]See Table 3.5, note d.
[e]Reference 61.
[f]Reference 66.

The effects of ionizing radiation of the compressive strength and leachability of thermosetting polymer binders have been noted in the literature. In two recent reviews of LLRW binder materials, a value of 10^9 rad is given as the "irradiation stability"[37] or "radiation resistance"[38] of polyester resins. For composites of reactor waste solidified in the vinyl ester-styrene binder material, neither a significant degradation of compressive strength nor an increase in leachability is found up to doses of 10^8 rad[61]. (On the contrary, there is some improvement in these properties as a result of 10^8 rad irradiation.) Only proprietary information seems to exist on radiolytic generation of gas and no information seems to be available on the radiolytic generation, if any, of corrosives from these polymer materials. Radiolytic gas generation has been observed from various low-level wastes (Section 3.1, above), but radiolytically generated products have not been reported for composites of waste solidified in thermosetting organic polymers.

3.3 Waste Container Materials

The LLRW container is primarily a means of handling the waste or waste form during interim storage, transportation, and disposal; it may also serve as the process vessel during solidification. The container is a barrier to radionuclide release (although it is generally given no credit for this after burial unless it is a high-integrity container) and may provide some radiation shielding.[68]

The containers normally used for shipping radioactive waste for disposal are:

- 55-gal carbon steel drums (usually DOT-17C, -17E, or -17H*)
- carbon steel shipping cask liners of various sizes
- wooden crates
- high integrity containers (HICs)

Certain waste generators have used, and continue to use, their own special containers. For example, Brookhaven National Laboratory uses large reinforced concrete vaults with 6- or 12-in. walls. Argonne National Laboratory uses 4- x 5- x 6-ft bins made of 16 gauge mild steel, with the flanged lids gasketed and bolted down.

Except for HICs, the main criterion for use of a particular container is to satisfy DOT regulations. Class B and Class C wastes would thus be contained in either drums or liners, with use of wood restricted to Class A wastes. The large steel liners, used essentially only for waste from nuclear power plants, are marketed by several companies, including Chem-Nuclear

*Department of Transportation (DOT) specifications as given in 49 CFR Part 178.

Systems, Inc., Hittman Nuclear and Development Corp., Nuclear Packaging, Inc., and TTI Engineering. The various liners are described in Appendix C of Reference 2.

An alternative to solidifying Class B and Class C waste streams is the use of a HIC to provide the long-term stability required by 10 CFR Part 61. Use of HICs was initially geared to disposal of dewatered (but not dry) resins from power plants, but is certainly not restricted to that type of waste. A limit of 0.5 percent by volume free liquid is set by 10 CFR Part 61 for all containers except HICs, which are allowed to contain waste with a free liquid content of up to 1% by volume. Since Class B and Class C wastes are required to be stabilized, such waste can be shipped only in HICs, unless it is first solidified. Design criteria for HICs written by NRC and the State of South Carolina,[4] require that the containers have a lifetime of 300 years. So far the only material used in a state-certified HIC is high density polyethylene (HDPE), but container designs recently submitted for approval use either a special steel alloy[69] or fiberglass-reinforced plastic[70] as the container material.* The concept of the HIC and its desirable properties are discussed in Reference 3, and design details of several commercially available HICs are given in Appendix C of Reference 4.

In the following discussions, the properties of carbon steel and high density polyethylene that are important in the context of extended storage of LLRW are considered. These two materials are selected because of their past, present, and likely future use as containers for the storage, shipping, and disposal of LLRW. For carbon steel, its corrosion properties in contact with solidified waste as well as with air are examined. Much of this discussion would be qualitatively similar for the special steel alloys that are being considered for use as HIC materials. For polyethylene, both its potential for corrosion by free liquid in the waste and its susceptibility to radiolytic deterioration are discussed. The implications that these container material properties have for container degradation and container lifetime under the anticipated conditions of extended storage are discussed later in Section 4.1.

3.3.1 Corrosion of Carbon Steel Containers

Corrosion of containers during extended storage is a concern because any loss of container strength and integrity may result in release of waste, loss of shielding, and failure to meet DOT packaging requirements. Corrosion of waste containers may thus necessitate repackaging of the waste for transport and the attendant increase in occupational dose.

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*A reinforced concrete HIC for the disposal of Epicor-II pre-filters from the TMI-2 cleanup has been accepted by the State of Washington. See R. C. Schmitt and H. W. Reno, "Experiences in Development, Qualification, and Use of Concrete High-Integrity Containers in Commercial Disposal of Radioactive Wastes," in Waste Management '85, R. G. Post, Ed., pp. 299-301, 1985.

General accounts of metal corrosion are available in textbooks, e.g., Uhlig,[71] and a brief overview of waste package corrosion considerations for on-site storage of LLRW has been presented by Murray and Guilbeault.[72] The eight major forms of corrosion defined by the National Association of Corrosion Engineers are discussed by these authors, namely, uniform attack, localized attack (pitting and crevice corrosion), galvanic attack, velocity phenomena (erosion corrosion and cavitation erosion), fretting, intergranular attack, dealloying attack, and cracking phenomena (stress-corrosion cracking and corrosion fatigue). Velocity phenomena and fretting, which involve frictional contact and motion, are unlikely to occur during storage, although one velocity phenomenon, erosion corrosion, could be initiated during the filling of radwaste containers by the use of a high-velocity flow, since suspended solids in the liquid may wear away protective oxide layers (or other non-metallic coatings) and substrate metal on the inner surface of the container. Intergranular attack is mentioned as a possible corrosion mode at welds. Dealloying attack of resin containers in which the ion-exchange media are still active is cited as another corrosion concern, particularly for lightly-loaded cation-exchange resins for which the exchange equilibrium may favor extraction of metals from the storage container material. Stress-corrosion cracking is considered a possible failure mode by these authors because of (1) an appropriate chemical environment, e.g., the presence of chloride, hydroxide, and corrosion products in the waste, (2) stress -- residual and welding stresses, in particular, and (3) material, such as stainless steels, which are being considered in some HIC designs. There is, however, no evidence that stressed carbon steels are susceptible to stress-corrosion cracking under these conditions at the temperatures expected during extended storage. Galvanic corrosion was discussed briefly in Section 2.2 above. Investigations of uniform corrosion and pitting of carbon steel by waste and binder materials are discussed below after a brief account of atmospheric corrosion of carbon steel.

3.3.1.1 Atmospheric Corrosion of Carbon Steel

Corrosion of a carbon steel drum or liner used for the extended storage of LLRW may proceed from the inner and/or outer surface of the container wall. External corrosion during extended storage is essentially atmospheric corrosion. The rate of atmospheric corrosion is influenced by several factors, including relative humidity, dust, and corrosive gases.[71] Internal corrosion of the container results from chemical reaction with the waste or waste form or with corrosive liquids, gases, or condensates exuded from the solidified waste.

The presence of moisture is a key factor in the corrosion of iron or steel exposed to the atmosphere. A thin film of condensed moisture on the metal provides the electrolyte necessary for the corrosion process and also allows oxygen to diffuse to the metal surface. Thus, appreciable corrosion of a smooth clean metal surface would be expected only as a result of condensation of water vapor, which occurs at a relative humidity of 100%. The term "relative humidity" is ordinarily considered to be a bulk property of atmospheric air. A film of water can condense on a surface which is cooler than

the bulk air if the humidity of the air is sufficiently high but still less than 100%. The cooler air film next to the water film will be at 100% relative humidity. Because of temperature fluctuations and because of hygroscopic impurities deposited from the atmosphere (i.e., certain dust particles) or originating in the metal itself (e.g., corrosion products), water may condense on the metal surface even at a relative humidity <100%. For example, on a metal surface already covered with corrosion products, surface condensation can take place in the pores of the corrosion product at relative humidity values well below 100%. Rather than enhancing further corrosion, however, rust films formed by atmospheric corrosion are often protective, the corrosion rate decreasing with time until a steady-state corrosion rate is attained. Atmospheric corrosion rates for steels in an industrial atmosphere averaged over a 10-year period are given by Uhlig;[71] the values range from 0.1 to 0.5 mils per year with about half the corrosion occurring during the first year. Experimental values for the critical relative humidity below which atmospheric corrosion is negligible for steel range from about 50% to 70%.[71] If relative humidity is increased above the critical point and subsequently lowered, there is some indication that it must be reduced to 30% or lower for the corrosion rate to once again become inappreciable.[73]

One of the most severe corrosive atmospheric environments is the marine atmospheric environment. From test data taken at Kure Beach, North Carolina,[73] the estimated corrosion rate for carbon steel was 55 mils in 20 years at 80 ft from the beach but fell to 30 mils at 800 ft, where the salt water spray is less frequent. In an earlier report on these corrosion data,[74] it is noted that long-term figures for the corrosion rates of carbon steels at the Kure Beach test site are not reliable because most of these materials corroded away early in the testing. Pit depths were not measured in the Kure Beach specimens, but based on data from other marine locations, pit depths may be as much as two to three times deeper than the average corrosion depth. These enhanced corrosion rates are attributed to the lower relative humidity at which the salts deposited by the oceanic saline spray absorb moisture from the atmosphere.[75] Some of the constituents of sea salt, in particular calcium chloride and magnesium chloride, absorb moisture at relative humidities of 31% and 33%, respectively, which, for corrosion contaminants, are the lowest values of the relative humidity at which moisture absorption occurs. (For example, sodium chloride and potassium chloride absorb water from the air at relative humidities of at least 57% and 80%, respectively.[75])

The presence of dust also influences atmospheric corrosivity. Dust is the chief containment by weight in many atmospheres, ranging from below 2 mg/m^3 in "average" city air to above 1000 mg/m^3 in industrial atmospheres.[71] Industrial atmospheres may contain suspended particulates of substances such as carbon, organic compounds, metal oxides, sulfuric acid, and various salts, which if deposited on a metal surface, may initiate corrosion by forming an electrolyte because of their hygroscopicity.[71]

Gases in the atmosphere also contribute to both external and internal corrosion of the container. Sulfur dioxide, in particular, is a common

corrosion component of urban and industrial atmospheres.[71] The vapor above the LLRW inside the container may also be corrosive, as documented for samples of mild steel and of steel drum walls exposed to the vapor above simulated LLRW solidified in urea-formaldehyde,[68] a binder material no longer acceptable at the operating disposal sites.

The rate of corrosion of drums of transuranic (TRU) waste at the U.S. Department of Energy's Hanford Project, near Richland, WA, has been evaluated by Pacific Northwest Laboratory (PNL).[73] Only corrosion on the outside of the drums, presumably atmospheric corrosion, was discussed in any detail in this study. The TRU waste was stored in 55-gallon mild steel drums "with a desired recovery period of 2.0 years." The PNL authors note that if the estimate of 20 mils uniform atmospheric corrosion in 20 years is accurate, there should be no problems with the structural integrity of the drums since this corrosion depth represents only about one third of their nominal wall thickness. However, it is further noted that at both the Idaho National Engineering Laboratory (INEL) and the Hanford site, increased corrosion was observed on the lids and at points of contact with the ground. Corrosion problems with these drums at INEL were said to have ceased as soon as a commercially available air support weather shield -- basically an air-inflated fabric structure -- was placed into use, presumably because the drums were no longer exposed to rainfall and air-borne particles. The PNL authors also report on the corrosion of similar containers at other DOE facilities. At the Oak Ridge storage site, mild steel drums failed the 20-year lifetime criterion because of atmospheric corrosion and were replaced by more expensive drums of 304 stainless steel. The storage facility for these drums at Oak Ridge is a 40- x 10-ft concrete block structure that is approximately 85% underground. At Savannah River Laboratory, galvanized drums, i.e., drums coated with zinc metal, have been used since 1974 with apparently satisfactory results. The drums, stored at ground level, are covered by plastic sheeting which is in turn covered by 4 ft of earth. Empty drums are stored indoors until filled, and are then placed outside and exposed to the environment for several weeks until they are covered. The drums were examined after one year, and it was estimated that they should last from 15 to 40 years under these conditions. It is unclear, however, whether these estimated drum lifetimes refer to the time needed for complete corrosion of the drum walls, since uniform corrosion rates are not reported for the drums stored at Oak Ridge and Savannah River.

3.3.1.2 Corrosion of Carbon Steel by Waste and Binder Materials

Corrosion of mild steel totally immersed in three kinds of simulated unsolidified LLRW has been investigated by Colombo and Neilson.[68] The three simulated wastes used were a BWR precoat filter cake with powdered resin in slurry form, a BWR chemical regenerative waste and boric acid waste, the last two from forced recirculation evaporators. The corrosion of mild steel in the BWR powdered resin waste and in the BWR chemical regenerative waste is low, <1 mil per year (mpy), but in boric acid waste, it is high, about 3 to 4 mpy. In all three cases, the corrosion is uniform with no localized pitting. The greater corrosivity of the boric acid waste is attributed to its low pH of about 3 to 5. (The pH of BWR powdered resin is about 6.) From the more

recent data of Swyler, Dodge, and Dayal,[18] a corrosion rate of about 0.4 mpy may be derived for mild steel in H^+ form cation resin.

A metallurgical examination of two Epicor-II prefilter liners was conducted by EG&G Idaho, Inc., for the U.S. Department of Energy's Epicor-II Research and Disposition Program. These liners were used during the cleanup of contaminated water from the accident at Three Mile Island Unit 2.[76] Two phenolic coated carbon steel liners, one containing an organic ion-exchange medium and the other an organic ion-exchange medium and an inorganic zeolite and which had held their resin beds for three years, were selected for examination. The exterior phenolic coatings of both liners were found to be in good condition except for minor handling scrapes and some imperfections acquired during fabrication. The interior coatings of both liners exhibited blistering and some spalling, and corrosion was evident where the coating had blistered or spalled. An area of inside surface from which the coating had been removed mechanically (for a conductivity probe) before loading with ion-exchange medium, was encrusted with resin and corrosion products. Both liners had bands of rust on the interior walls at the level corresponding to the top surface of the resin. The coatings in many areas had been applied over corrosion products, which indicates that the base metal was not properly prepared before application of the coatings. It is believed that radiation damage did not contribute to the failure of the coatings since the estimated dose to the coatings on the internal surfaces of both liners was $<10^8$ rad, well below the limiting doses ranging from 2.1×10^9 to 8.3×10^9 rad established for this phenolic coating by tests at Oak Ridge. (The total estimated dose to the coatings for the highest loaded liner from the TMI-2 cleanup is about 4×10^8 rad after 13 years of storage.) A corrosion rate of 5 mpy was estimated from the corrosion of the area of base metal. Using this rate, a lifetime of 50 years was estimated for a liner with a nominal wall thickness of 0.250 in.[76] (In this context "lifetime" evidently means the time needed for complete uniform corrosion of the wall of the container.) The effect, if any, of dose rate on the irradiation of the phenolic coatings does not seem to have been investigated. The dose rates used in the Oak Ridge study were not given in the report on the metallurigical examination of the Epicor-II liners.

Mild steel coupons partially embedded in cement waste forms have also been tested. As expected for a poor electrolyte like cement, corrosion of the steel occurs at such locations as the interface between the air and the waste form or at some defect in the metal.[68] Also, cement provides a highly alkaline environment which protects the steel against corrosion under ordinary circumstances. In addition, the low permeability of well-cured cement with a low water-cement ratio minimizes the penetration of oxygen to steel, chloride ion, and water, all corrosion-inducing agents.[77]

Information on the corrosivity of other binder materials in contact with steel is very limited. A likely cause of such corrosion is chemically aggressive free liquid associated with the solidified waste form, even at 0.5% or 1.0% of the waste volume, the maximum free liquid specified for stabilized wastes or for wastes packaged in a HIC, respectively, by 10 CFR Part 61. As

noted in Section 3.2 above, corrosion of metals in contact with bitumen has been attributed to the effects of biodegradation. It has been stated that vinyl ester-styrene, a thermosetting organic polymer binder material, is non-corrosive to mild steel, but no supporting data are presented.[68] A value of approximately 0.01 mil/day (4 mpy) is reported for the corrosion rate of mild steel embedded in waste forms consisting of a chelating decontamination chemical, Dow Chemical Company's NS-1, solidified in Dow vinyl ester-styrene binder in a 1.5 waste/binder ratio.[78] Also, as noted above in Section 3.2, droplets of a potentially corrosive (pH = 4.5 to 5.5) free liquid "sweat" have been observed on waste forms consisting of various simulated LWR waste streams solidified in vinyl ester-styrene binder.[67] Estimates of mild steel waste container lifetimes under anticipated storage conditions based on the limited data available are given in Section 4.1 below.

Reduction of the relative humidity by heating the air or removing some of its moisture (e.g., by passing the air over hygroscopic drying agents) is a well-known method for mitigating corrosion of metals. Lowering the relative humidity to 50% is often sufficient, although the presence of unusually hygroscopic dust or other surface impurities may necessitate even lower values.[71]

Metallic, inorganic, and organic coatings have all been used to mitigate the corrosion of metals, whether from atmospheric constituents or from contained corrosive materials. A 1-mil-thick zinc coating was found to last for about 11 years or longer in rural or suburban locations and for about 8 years when exposed to marine atmospheres, but only for about 4 years in industrial atmospheres. Vitreous enamel coatings are used mostly on steel and protect the base metal against many corrosive environments although these coatings are susceptible to mechanical damage and cracking by thermal shock.[71] Several kinds of organic coatings have been developed to protect metals from corrosion, e.g., phenol-formaldehyde formulations, silicone resins, vinyl resins, and epoxy resins. The importance of proper preparation of the base metal before application of these protective coatings has been noted both for corrosion of metals in general[71] and for the specific case of corrosion of phenolic-coated carbon steel by ion-exchange resins.[76] In the latter case, there were indications that blistering and spalling of the phenolic coating inside the steel container had been caused by penetration of moisture through the coating where it had been applied over corrosion products that had not been removed from the surface of the metal. Further information on the use of coatings for the mitigation of corrosion may be found in standard texts on corrosion and corrosion control, e.g., Uhlig and Revie,[71] and the references cited therein.

3.3.2 Corrosion of Polyethylene

HICs have been used extensively for disposal of dewatered ion-exchange resins from nuclear power stations. Free liquid in a HIC may be a vehicle for corrosion from within if there are corrosive constituents in the liquid. Ion-exchange resins and other dewatered or solidified waste may release liquid from incomplete dewatering, solidification, or other processes (e.g., thermal

cycling -- see discussion in Section 3.2.3). The TP states that the maximum allowable free liquid in a HIC should be less than 1% of the waste volume. Piciulo[12] attempted to characterize ion-exchange resin wastes from nuclear power plants. It was concluded, however, that the characteristics of drainable liquids in a HIC filled with dewatered resins vary considerably because of the large variations in water quality and resin wastes among plants. Upper and lower limits for certain quantities can, in principle, be identified, however, on the basis of experience. A pH range of 4 to 11 may be expected. Similar large variations in the pH may be expected from other dewatered wastes to be disposed of in HICs.

Corrosion of polyethylene means chemical attack which may solvate the polyethylene matrix and/or result in the breaking of chemical bonds. Polyethylene is also subject to stress-corrosion cracking in certain environments. Solvation is typically accompanied by swelling and softening and, taken to the extreme, results in dissolution of the polyethylene. Actual dissolution of polyethylene occurs only at elevated temperatures, above approximately 70°C (160°F), in organic solvents such as toluene, xylene, petroleum ether, lubricating oils, turpentine, and degreasers such as trichloroethylene. At temperatures below approximately 50°C (120°F) polyethylene is insoluble but may swell, soften, and suffer a limited amount of leaching due to solvent attack.[61] The maximum temperature to which the container would be exposed during storage is likely to be about 60°C (e.g., as a result of insolation of a closed uncooled storage building).

The breaking of chemical bonds in polyethylene is generally the result of attack by oxygen or chemical oxidants. Antioxidants are frequently added to polyethylene and other plastics to retard this process.[79] A very slow "autocatalytic" oxidation of high-density polyethylene (HDPE) in air at room temperature has been reported. This room temperature autocatalytic oxidation would result in a weight loss of an HDPE container not exceeding 10% in 300 years.[61] It is unclear whether the effect of antioxidants is included in this autocatalytic oxidation process. The rate of oxidation of most polymers is usually very small at ambient temperatures and in the absence of radiation because the rate of production of initiating free radicals is small. The free radical chain reaction for oxidative degradation may be initiated photochemically, thermally, mechanically, or by ionizing radiation such as γ rays.[80] In principle, antioxidants would not be required for polyethylene containers which are shielded from direct sunlight and kept at ambient temperatures and which are neither subjected to mechanical strain nor exposed to ionizing radiation.

Oxidizing acids (e.g., nitric, sulfuric) and other chemical oxidants (e.g., hydrogen peroxide, sodium hypochlorite) also attack polyethylene. At room temperature, attack from most chemical oxidants and oxidizing acids is reportedly not significant for moderately concentrated solutions (e.g., 30% nitric acid, 30% hydrogen peroxide, and 70% sulfuric acid reportedly had no effect on HDPE at room temperature during a three-month test period). However, these reagents at higher concentrations do attack polyethylene at room temperature. Additionally, for chemicals capable of attacking polyethylene,

a temperature increase generally leads to attack at smaller concentrations. It should be noted that these observations of the effects of chemical oxidants on HDPE are in the absence of radiation.

Polyethylene is subject to stress corrosion cracking in certain environments. Such attack could lead to failure of a HIC by a breach of containment at stresses lower than its ultimate strength. In general, chemicals which can induce stress-corrosion cracking in polyethylene include alcohols, halogenated solvents, oils, soaps, and detergents.[61]

3.3.3 Radiation Effects on Polyethylene

Exposure of polymers to ionizing radiation can result in a deterioration of their mechanical properties. For example, the elasticity can deteriorate, the polymer material becoming hard, brittle and susceptible to cracking under relatively minor mechanical stress. In a few cases, such materials may become soft and tacky. The major cause of such changes in the macroscopic mechanical properties results from scission of the polymer chains and/or from cross-linking between chains. Oxygen typically plays a major role in the degradation when polymeric materials are irradiated in air. In addition, the effects of dose rates can be important in the presence of oxygen.[81]

In an earlier BNL survey[61] of irradiation studies on polyethylene in the context of HICs, it was found that irradiation in anoxic conditions promotes cross-linking in polyethylene rather than degradation. Hydrogen gas is generated in the cross-linking process. The G-value for the production of hydrogen gas from polyethylene during irradiation has been given by Charlesby[82] as 3.7 based on measurements conducted at doses of about 10^7 rad. In irradiated samples, the tensile yield point and tensile creep modulus are generally increased, while the ultimate tensile elongation (or ductility) is decreased. In particular, these effects have been measured for cross-linked HDPE of the type used in high integrity containers.[83,84] Presumably these effects represent increased cross-linking. It is also reported that irradiation improves environmental stress cracking resistance of this material.[85]

In cases where radiolytic oxidation of HDPE is important, the effect of irradiation may depend upon dose rate as well as total dose. Oxidation degrades the polymer and, for a given dose of irradiation, the degradation may be more extensive at lower dose rates.[61] The "premature" mechanical failure of polyethylene electrical insulation in reactor environments has been attributed to radiolytic oxidation effects at low dose rates. These effects were not evident at the higher dose rates used to accelerate testing in the laboratory.[86] As another example of such dose-rate effects, polyethylene and polyvinyl chloride cable insulation have been found to suffer more damage at low dose rates (1-10 krad/h) than at high dose rates (1-10 Mrad/h) at the same total dose (typically 10^7 to 10^8 rad).[87]

Dougherty and co-workers at BNL have specifically investigated irradiation effects on the bulk tensile and flexural properties of HDPE at different radiation dose rates.[84] In these experiments, samples were irradiated in contact with soil and ion-exchange resin. These experiments do not indicate any substantial contribution of the environment to irradiation effects on the bulk tensile properties of 1/8-in. and 1/2-in. thick HDPE samples. Dougherty finds, however, that there is a radiation dose region above which irradiated samples abruptly break in tensile testing, rather than neck and elongate, which is suggestive of a ductile-to-brittle transition, although the samples yield before rupture in all cases. The dose to produce this tensile "necking-to-cracking" transition decreases with irradiation dose rate.

Dougherty and co-workers find that, when plotted as a function of dose vs dose rate, the transition from necking to breaking appears linear on a log-log scale. The relationship obtained from these plots for Chemplex 5701, a high-density, high-molecular-weight non-cross-linked polyethylene, is

$$D_N = 550000\ (R)^{0.32}, \tag{3.1}$$

and for non-HIC Marlex CL-100 material, it is

$$D_N = 77000\ (R)^{0.48}, \tag{3.2}$$

where D_N is the dose (rad) up to which necking predominates at a dose of R(rad/h). Marlex CL-100 is a high-density, highly cross-linked polyethylene. The relationship for the Marlex HIC material is presumed to be the same as that for the non-HIC Marlex material in Eq. 3.2.

Estimates of D_N (i.e., the dose below which necking will predominate in irradiated material) and the time to reach D_N for several dose rates using these equations have been made by Dougherty et al. and are presented in Table 3.8. The values of D_N and the time to D_N at the lower three dose rates in Table 3.8 represent extrapolations from the data obtained primarily at dose rates between 2 and 100 krad/h. The tabulated dose rates were chosen to bracket estimated initial dose rates for highly loaded IX resin waste, as described below.

As noted by Dougherty and co-workers, for wastes whose activity is dominated by isotopes with half lives on the order of 30 years (e.g., Cs-137), such that the total accumulated dose would be 10^8 rad, the dose rate to which the container may be exposed upon loading would be approximately 250 rad/h. The accumulated dose, based on this loading, would be approximately 2.3 Mrad after one year. Similarly, for wastes whose activity is dominated by isotopes with half lives of 5 years (e.g., Co-60) and loaded such that the total accumulated dose would be 10^8 rad, the dose rate to which the container would be exposed when filled is approximately 1500 rad/h. In this case, one year after loading, the accumulated dose would be approximately 13 Mrad. It should be noted that these estimates of anticipated dose rates and doses are probably conservatively high since they neglect container geometry and self-shielding by the resin wastes. Using these dose rates as a benchmark for expected field

conditions, however, leads one to conclude that the transition could occur within 2 months to a year. Although uncertainties are associated with this estimate, it would appear that the consequences of such embrittlement during storage should be considered in the design of HICs made from HDPE. In any case, the transition appears to be dose-rate dependent and occurs at lower total doses for lower dose rates over the ranges of doses and dose rates investigated.

Table 3.8

Estimates of the Dose and Time-to-Dose for the Necking to Breaking-Without-Necking Transition for Marlex CL-100 and Chemplex 5701[a]

Material	R(rad/h)	D_N(Mrad)[b]	Time to D_N (Days)
Marlex CL-100	2000	3.0	63
	1000	2.1	88
	500	1.5	125
	100	0.7	292
Chemplex 5701	2000	6.3	130
	1000	5.0	209
	500	4.0	335
	100	2.4	1000

[a]From Dougherty et al.(84)
[b]Calculated from $D_N = 77000\ (R)^{0.48}$ and $D_N = 550000(R)^{0.32}$ for Marlex CL-100 and Chemplex 5701, respectively.

Crack initiation was observed by Dougherty et al. only on the inner oxidized surface of the rotationally molded Marlex CL-100 HDPE specimens. Arora and Dayal(83) report that for unirradiated Marlex CL-100 specimens from which the oxide layer had been removed by polishing with sandpaper prior to tensile strength testing, elongation at break is observed to increase in a "dramatic" manner from about 22% to about 900%. It was suggested that the low extensibility of the oxidized inner-surface layer significantly contributes to the relatively brittle behavior of the specimens with an intact oxide layer. It is unclear whether radiolytic changes in the bulk material may enhance the propagation of the surface-initiated cracking into the bulk.

At low temperatures (≈10°C), there appears to be little evidence for a significant radiolytic oxidation effect on the bulk properties of HDPE. Irradiation in air at 60-63°C, however, results in a loss of strength, as well as

decrease in elongation at yield and at break. The loss of strength appears to result from embrittlement which begins at the surface and which progresses into the bulk material as the irradiation in air is continued. This is attributed to radiation-induced oxidation.[84] Radiolytic oxidation has been observed in HDPE[83] at low temperatures, but it seems to be largely confined to a surface layer which is small compared to sample thickness. However, although such surface layers may not affect bulk properties such as yield stress, oxide films might play a role in the promotion of cracking behavior. According to both Dougherty[84] and Arora et al.,[83] commercial samples of rotationally molded HDPE contain an oxide layer on the inner surface, and cracking or rupture in tensile tests is always initiated at this surface. Presumably, oxide layers formed by radiolytic oxidation might also affect either the initiation or slow propagation of cracks. It must be recalled, however, that the radiation dose will be greatest, and perhaps largely localized, at the inner surface of a HIC. For a sealed container containing ion-exchange resins, the atmosphere inside the container may quickly become anoxic (see Section 3.1 above).[17] The customary addition to commercially available polyethylene of antioxidants, the nature and quantity of which are generally proprietary, does not allow the data from a particular set of laboratory tests to be generally applicable to all polyethylene material. Also, whether radiation-enhanced oxidation is a factor in the presence of chemical oxidants other than oxygen remains an open question.

Tensile testing indicates that while certain mechanical properties (creep modulus, yield strength) are improved by irradiation, an increasing loss of ductility or a tendency toward embrittlement also occurs. This loss of ductility may affect other properties such as impact resistance, which are not measured in the tensile test. Taking the area under the tensile stress-strain curve as a measure of impact resistance suggests that impact resistance may be significantly decreased in irradiated samples. However, the correspondence between tensile data and impact resistance is difficult to quantify.

3.4 Overview of Properties of Wastes, Binders, and Container Materials

In Sections 3.1 to 3.3 above, the properties and behavior of low-level radioactive waste streams, solidification agents, and container materials have been 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, no general accounts of properties and behavior are available. Therefore, the characterization of waste streams in this report deals only with fuel-cycle wastes. An overview of the properties and behavior of the wastes, binders, and solidification agents is presented below. It should be noted that many of the radiation effects are based on data obtained from laboratory tests at high doses ($>10^8$ rad), which were used to accelerate the tests.

Ion-Exchange Resin Wastes

- 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. According to the guidance given to LLRW generators in the NRC Technical Position on Waste Form (TP), 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 TP is exceeded. It should be noted that the following radiation processes may be affected both quantitatively and qualitatively by the the partial pressure of oxygen and by the dose rate.

  - Irradiation of ion-exchange resins may produce and/or release chemically active substances that can adversely affect the binder and container materials.

  - Radiolytic generation of gases has been observed. The predominant gas, hydrogen, 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 has been reported as resulting from the irradiation of ion-exchange resins.

  - Irradiation of ion-exchange resins before their solidification in cement has been reported to improve the compressive strength and immersion resistance of the resulting waste forms. (Such effects may be of significance if the spent resins are stored for some time in an unsolidified form and then solidified without repackaging.)

- Agglomeration of and gas buildup in unsolidified ion-exchange resins during storage has been attributed to biodegradation.

Other LWR Wastes

- The radiolytic generation of gases (predominantly hydrogen) and of corrosive substances has been observed in cellulosic materials.

- Because much of this waste consists of organic materials (e.g., cellulose) biodegradation is likely if the wastes have not been self-sterilized or treated with a biocide. 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.

- 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.

- Freeze-thaw cycling can damage cements which contain sufficient amounts of freezable water.

- Certain of the products of waste radiolysis, such as low-molecular weight organic acids, have been found to attack cement.

Binder Materials: Bitumens

- Radiolytic generation of gas has been observed in bitumens. 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, whether in the gas phase or incorporated into the solid matrix of oxidized bitumens.

- Biodegradation of bitumens has been observed. There is some evidence that corrosive substances may be produced as a result of biodegradation of bitumens.

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.

- A small amount (<0.4 volume percent) of a somewhat acidic (pH=5) free liquid has been observed on the surface of small-scale waste forms consisting of simulated LWR waste streams solidified in vinyl ester-styrene. Thermal cycling of these waste forms increases the amount of this free liquid (up to 1.3 volume percent).

Container Materials: Carbon Steel

- The following generic types of corrosion are 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).

- Corrosion by the atmosphere, generally 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,corrosion commonly known

as "rust". Ten-year average rates of 0.1 to 0.5 mils per year (mpy) are reported for the atmospheric corrosion of steels in an industrial atmosphere, with about half the corrosion occurring during the first year.

- 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. (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. Corrosion of metals in bitumen has been attributed to biodegradation. A corrosion rate of about 4 mpy is reported for mild steel embedded in waste forms consisting of a chelating decontamination reagent solidified in vinyl ester-styrene.

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 rather than degradation. Irradiation in air produces radiolytic oxidation at the surface. This oxidized zone is believed to penetrate gradually 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 may be temperature dependent.

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4. EVALUATION OF THE EFFECTS OF STORAGE CONDITIONS

4.1 Potential Problem Areas

Potential problem areas for the extended storage of LLRW are considered in this section. It is assumed in the following that the waste is not to be repackaged for shipment, but is to be shipped from the extended storage facility and disposed of in the same containers used for storage. These two assumptions are in accord with the design guidance given by the NRC for temporary on-site storage of LLRW.* Under these circumstances, the waste container would have to meet the requirements for packaging and transportation of radioactive materials as set forth in 49 CFR Part 173 Subpart I and 10 CFR Part 71. In addition, the waste and/or container would have to meet the requirements for disposal set forth in 10 CFR Part 61, in particular, Sections 61.55 and 61.56. A further corollary of these assumptions is that liquid waste will not be stored for extended periods unless it can be processed in the storage container to a form suitable for disposal without repackaging. The areas of concern about extended storage of LLRW may be grouped into two categories:

1) performance of the waste, waste form, and/or container material during storage, and

2) effects of extended storage that are important after the storage period.

The areas of concern in these two categories are discussed in Sections 4.1.1 and 4.1.2, below. The discussion is based on the information in Sections 3.1, 3.2, and 3.3.

4.1.1 Performance During Storage

In Sections 3.1 to 3.3 of the report it is noted that evidence for radiolytic gas generation has been observed for several waste, binder, and container materials, particularly at high accumulated doses ($>10^8$ rad). During an irradiation in a sealed system an initial pressure drop, attributed to radiolytic oxygen scavenging, is often observed. As the irradiation proceeds, this pressure drop is then followed by an increase in pressure due to radiolytic generation of gas, mostly hydrogen. Such a sequence of events has been observed during the irradiation of ion-exchange resins, cement, and bitumen and may be inferred for polyethylene from other observations. In addition, the G-values for gas generation, i.e., the number of molecules of a particular species produced per 100 eV of absorbed radiant energy, exhibit an apparent dose-rate dependence. "Apparent" is used because the rate-limiting step seems to be a thermally activated process.

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*In Appendix 11.4-A to the Standard Review Plan for the Review of Safety Analysis Reports for Nuclear Plants, LWR Edition, NUREG-0800, July 1981.

(Two examples of such thermally activated processes which can be significant at room temperature are diffusion of the reactant and/or product species through the waste and/or binder matrix and cross-linking of a polymer material with radiolytic production of hydrogen as a by-product. The temperature dependence of the diffusion of hydrogen gas through binder materials does not seem to be documented, but the available data on the radiolytic generation of hydrogen gas from bitumen indicate that this gas is released from bitumen samples at room temperature. Radiolytic gas generation from bitumen is discussed in Section 3.2.2. Whether the release of hydrogen gas involves thermally activated diffusional processes does not appear to have been investigated. The temperature dependence of the G-value for radiation-induced cross-linking of a polyethylene material is given by Charlesby[1], who reports that specimens subjected to gamma irradiation showed an increase in G-value by a factor of about 4 over the temperature range -196°C to +73°C. On the other hand, Dougherty et al.,[2] investigating the γ-irradiation of high-density polyethylene, found evidence for radiolytic oxidative degradation, which involves the thermally activated diffusion of oxygen through the polyethylene, at 60 to 63°C but not at 10 to 11°C.)

The amount of radiolytically generated gas has been calculated for several of the materials discussed in the report (see Appendix C). For example, for a G-value of 0.4, which is believed to be reasonable for bitumen, and an internal dose of 5×10^7 rad delivered over a period of 30 years, a 55-gallon drum of bitumen has been estimated to generate a daily average of 10 cm^3 of gas, consisting predominantly (>95 volume percent) of hydrogen with some light hydrocarbons.[3] The radiolytic generation of gas could present problems for the binder material, the container, and the storage facility.

If the rate of generation of gas in a waste form matrix exceeds the rate of diffusion of the gas out of the matrix, the monolithic integrity of the waste form could be compromised by internal pressurization within the waste form matrix. Materials such as bitumen, which retain some plasticity, are likely to swell as a result of such pressurization, in accord with the observations of radiation-induced swelling of this material discussed in Section 3.2 of the report. Gas bubble formation in certain irradiated bitumens has also been observed. More rigid materials could fracture as a result of internal gas generation if the pressure is not relieved through loss of gas by diffusion or by some other mechanism.

On a larger scale, the radiolytic generation of gas could result in pressurization of a sealed storage container. If the generation of gas within the container is greater than the diffusion of gas through the wall or gasket of the container, then pressurization and, possibly, failure of the container could occur. Also, escape of radiolytically generated hydrogen and other flammable gases (e.g., hydrocarbons) can create a hazard if allowed to accumulate because of poor ventilation in storage areas.

Radiolytic generation of potentially corrosive substances is also of concern. Such substances are produced by the radiolytic degradation of waste and binder materials. For example, as discussed in Section 3.1,

the radiolytic oxidation of ion-exchange resins is reported to produce acidic liquids and the radiolytic degradation of cellulosics is said to generate an array of organic acids. The production of such substances can result in the failure of the container during storage. Failure of carbon steel shipping containers (e.g., DOT 17-H 55-gallon drums) may occur by one or more of the several corrosion processes discussed in Section 3.3 above. The container wall may be penetrated by pitting corrosion. There may be a gradual loss of structural strength as the container wall is weakened by processes such as uniform corrosion or selective leaching. Stress-corrosion cracking is also a possible failure mode for steel in contact with certain LLRW free liquids (e.g., those containing chlorides and hydroxides)[3] and could result in sudden failure of stored stacked drums.

As discussed in Section 3.3 above, little quantitative information is available on the corrosion of carbon steel in contact with solidified LLRW. From the corrosion measurements of 55-gallon drums made of mild steel and containing solidified chelating decontamination agent (DOW Chemical's NS-1 in vinyl ester-styrene binder), a container lifetime of five years has been extrapolated.[4] From similar measurements at BNL of mild steel coupons embedded in NS-1 solidified in vinyl ester-styrene,[5] it was concluded that a lifetime of about 25 years could be expected for an 18-gauge 55-gallon drum with a nominal wall thickness of 50 mils.[4] These solidified decontamination wastes are not typical of routine power reactor wastes, but their corrosive properties are indicative of the potential for such corrosivity in other solidified LLRW. Corrosion measurements of mild steel in contact with various unsolidified LLRW simulations were also discussed in Section 3.3. These corrosion data and, assuming uniform corrosion attack, the resulting lifetimes estimated for 18-gauge 55-gallon steel drums containing these wastes are given in Table 4.1. Note that these data are for corrosion in the absence of radiation. If pitting corrosion occurs, then the drum wall will be penetrated before complete uniform corrosion of the container wall on which the container lifetimes are based. The data in Table 4.1 are relevant to Class A wastes, which are not heavily loaded with radionuclides and which may be packaged for disposal -- and, thus, very likely for storage -- in steel drums in dewatered form without solidification. Except for some observations by Colombo and Neilson of the corrosion of mild steel at the interface between cement-solidified waste and air,[6] the corrosion of steel by LLRW solidified in binder materials does not seem to have been quantitatively addressed.

Corrosion of steel in contact with unsolidified Class A wastes could be enhanced by radiation from outside the waste package if Class A waste packages are stored adjacent to unshielded Class B or Class C waste packages containing γ-emitters. As discussed in Section 3.3, irradiation of ion-exchange resins in contact with mild steel enhances corrosion. From these penetration data for the corrosion of mild steel in resins during irradiation,[7] the largest average penetration, 1.4×10^{-2} cm, is found for fully swollen H^+ resins irradiated to 10^9 rad over a period of (about 6 mils) 600 hours. If pitting is taken into account, the penetration could be as large as 9×10^{-2} cm (about 35 mils). These data are considered conservative because of the large total dose and because unsolidified resins heavily loaded with radionuclides

(Class B or Class C) would presumably not be stored in mild steel drums if the same drums are to be used for disposal. However, even if the container wall is not penetrated by pitting, structural strength will gradually be lost as a result of significant corrosion of the container wall.

Table 4.1

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

Simulated 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. Wastes are unsolidified.
[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.(6)
[d]Measured corrosion rate from Swyler, Dodge, and Dayal.(7)

Such failure is of more concern for the carbon steel shipping containers than for high-density polyethylene (HDPE) high-integrity containers (HICs), since, according to manufacturer data sheets, HDPE is considered to have satisfactory resistance to dilute aqueous solutions of several low-molecular weight organic acids (e.g., acetic, formic, lactic, and oxalic). This is in accord with the expectations for these containers after disposal. Carbon steel shipping containers are generally not given any credit towards stability in the disposal trench, but the design goal for HICs is a minimum lifetime of 300 years. From data on the fluids released by ion-exchange resins, these fluids do not appear to contain substances detrimental to HDPE under ordinary conditions (i.e., in the absence of a radiation field). The stability of HDPE to these fluids in the presence of a radiation field and its stability when exposed to the temperature fluctuations possible in certain modes of storage remain open questions. A radiation-enhanced creep rate has been observed in HDPE at loads greater than half the nominal yield strength and could pose problems if HDPE HICs are stacked too high during storage.

Atmospheric corrosion may be a problem for carbon steel waste containers during storage. Industrial atmospheres, in particular, often contain components that enhance the corrosion of metals exposed to them, but even in such atmospheres, a lifetime of several hundred years may be estimated (from the atmospheric corrosion rates in Section 3.3 above) for a 17-H 55-gallon carbon

steel drum with a wall thickness of 50 mils. However, atmospheric corrosion rates an order of magnitude greater are possible under certain conditions, e.g., marine atmospheres. As discussed in Section 3.3, the rate of corrosion of carbon steel in a marine atmosphere may depend on the distance from the beach and can be rapid enough to cause complete corrosion of a 50-mil drum wall within 20 years. (The drum integrity would be compromised before complete corrosion occurs.)

Biodegradative processes are potential concerns during the storage of LLRW for some of the same reasons as radiolytic processes. Gas production from ion-exchange resins in HICs and corrosion of metals in contact with bitumen have been attributed to biodegradation. Biodegradative gas production can result in pressurization of sealed containers, but no flammable or explosive gases have been reported as resulting from biodegradation in LLRW. Non-fuel-cycle wastes containing animal carcasses are subject to biodegradative processes, which may result in gas generation and the production and release of corrosive liquids. Systematic data in this area are lacking, however. Bagging of such wastes may limit the effects of corrosive liquids from these wastes on metal containers.

4.1.2 Effects Important After Storage

Corrosion of the shipping container during extended storage may compromise the ability of the container to meet certain of the requirements for radwaste shipping containers specified by the Department of Transportation in 49 CFR Part 173 Subpart I and by the NRC in 10 CFR Part 71. In particular, weakening of the container structure due to corrosion during extended storage may result in the container's failing the free drop, compression, and penetration tests specified by DOT for Type A packages in Section 173.465 of 49 CFR Part 173. Thus, even if the waste containers do not fail during extended storage, their contents may need repackaging before shipment from the extended storage facility.

Agglomeration of ion-exchange resins has been attributed to both radiolytic and biodegradative processes. As noted above in Section 3.1, such agglomeration has been observed in ion-exchange resins irradiated under vented (oxic) conditions. On the other hand, agglomeration of spent ion-exchange resins that had been stored in a holding tank has been attributed to biodegradation. Whatever the cause, such agglomeration of untreated spent resins may interfere with subsequent processing of the spent resins in the storage container to a form suitable for disposal.

The leaching properties of cement waste forms which had been stored in air -- the storage time, in effect, functioning as a curing period -- have been found to be affected by the length of the storage period as well as by the relative humidity of the atmosphere during that period. Storage periods ranging from a day to a year were investigated in several studies. The longer storage periods prior to leaching result in less leaching of strontium, and storage in a humid atmosphere decreases the initial leaching of cesium. It is not clear what effect a fluctuating humidity would have on subsequent

waste-form leaching. A fluctuating humidity would conservatively simulate the atmospheric moisture conditions for storage facilities with no controlled humidity. The effect of a fluctuating humidity before leaching may only delay the completion of curing, but BNL also notes that the effect on the leachability of various radionuclide cations may not be that simple. For example, cations such as strontium, which can chemically substitute into the cement matrix, may exhibit decreasing cumulative release with increasing periods of dry curing time, while cations such as cesium which diffuse through the cement matrix without being incorporated, may exhibit an initial "pulse" of increased release but not a change in the total cumulative release.

The temperature fluctuations in certain storage environments (in particular, outdoors) may have detrimental effects on the monolithic integrity of solidified waste forms. Cement, in particular, is subject to freeze-thaw degradation caused by expansion if the water enclosed in the pores of the material should freeze. Such degradation could adversely affect the disposal properties of the waste form. Vinyl ester-styrene exhibits an enhanced free-liquid production as a result of thermal cycling. Since the produced liquid is slightly acidic, corrosion of the storage container is a possibility.

Corrosion of binder materials and, in particular, of cement by radiolysis products is another possible area of concern. If significant degradation should occur during storage, the transportation and disposal properties of the waste forms could be compromised. Radiolytically induced corrosion of waste-binder formulations may be dismissed as a concern for extended storage in those instances where it has been concluded that the formulation is resistant to such degradation for the 300-year stability period specified for disposal in the NRC Technical Position on Waste Form.

The HDPE HIC material is subject to at least three radiation effects. First, radiolytic cross-linking of the polymer material accompanied by some generation of hydrogen gas results in a loss of ductility with a consequent embrittlement of the material. Second, radiolytic oxidative degradation of the material results in loss of strength. The third effect, a radiation enhanced creep rate, has already been mentioned above in connection with behavior of HDPE HICs during storage. At least the first two of these effects exhibit an apparent dose-rate dependence, presumably because a thermally activated rate-limiting step is part of the mechanism in each effect. In accord with this hypothesis, loss of strength resulting from irradiation of HDPE in air at 60 to 63°C was attributed to radiation-induced oxidation, but no evidence of oxidative degradation was found in HDPE irradiated at 10 to 11°C. Whatever the details of the mechanisms of these irradiation effects, as a result of any embrittlement and loss of strength of HDPE material during the extended storage of HICs fabricated from such material, the HIC may no longer meet the free drop and lifting load criteria in the Technical Position and, therefore, repackaging of the waste for shipment may be necessary.

A summary of the areas of concern for the extended storage of low-level radioactive waste are provided in Table 4.2.

Table 4.2

Potential Problem Areas for Extended Storage of LLRW

Area of Concern	Potential Problem
1. Performance of waste, binder, and/or container during storage.	
a. Radiolytic gas generation - from waste - from binder - from container material	a. - Pressurization, causing damage to waste form and/or container - Flammability hazard
b. Internal corrosion of shipping container during storage	b. Failure of container by corrosion during storage: - Penetration of container wall by pitting - Gradual loss of structural strength leading to collapse of stacked containers during storage
c. Atmospheric corrosion - relevant chiefly to steel containers	c. Failure of container during storage
d. Radiation-enhanced creep of HDPE	d. Collapse of stacked containers during storage
e. Biodegradation (i) gas production (ii) corrosion	e.(i) Pressurization (ii) Failure of container during storage
2. Properties of waste, binder, and/or container after storage.	
a. Corrosion of shipping container during storage	a. Container may not meet DOT requirements for shipping of radwaste. Repackaging of waste for shipment may be necessary
b. Radiolytic cross-linking and radiolytic oxidative degradation of HDPE	b. Failure of HIC to pass free drop and lifting load requirements. Repackaging of the waste for shipment may be necessary.
c. Agglomeration of spent ion exchange resins (biodegradative and/or radiolytic)	c. Interference with subsequent transfer and processing of resins
d. Effect of storage period on leaching properties, mechanical strength, and immersion resistance of waste forms, in particular, cement binders	d. Change in these properties (improvement or degradation)
e. Temperature fluctuations (freeze-thaw cycling)	e. Loss of monolithic physical integrity of cement waste forms

4.1.3 Time Scales for Extended Storage: Storage Phases

Since an accurate prediction of a maximum length of storage time before disposal is not currently possible, it is convenient to consider several ranges of possible storage period durations or, more briefly, storage phases. Phase I storage, for 1 to 5 years, is addressed in Generic Letter 81-38 where

it is termed "temporary" waste storage. Phase II covers a longer period of 5 to 15 years, which is not addressed in the generic letter. If required, Phase III would be for more than 15 years. The technical considerations for safety and surviellance could be significantly different for differing time periods, depending largely on time frames for particular waste package degradation modes, which proceed at rates depending on the characteristics of the waste, the binder material, and the container material, as well as the characteristics of the storage environment. Unfortunately, quantitative information on degradation rates of low-level waste package components is generally not available except for some rather specific waste package systems, e.g., rates of corrosion of carbon steel drum material by resin waste.

In general, the guidance presented in Generic Letter 81-38 regarding the low-level waste package will still be applicable to Phase II and Phase III storage periods if these should prove to be necessary. It will still be necessary to ensure that container materials are compatible with the waste forms and with environmental conditions external to the containers, that gas generation from the waste is addressed, and that provisions are made for additional reprocessing or repackaging if container failure should occur. For those specific waste package systems for which quantitative information on degradation rates is available, the length of time the packages may be stored without significant degradation may be estimated. Such information would be useful in selecting surveillance methods and in determining the frequency of surveillance for detecting the likely degradation mode (if any) for a particular storage situation, e.g., ultrasonic detection of internal corrosion of the container by the waste, visual inspection for external corrosion by the atmosphere, and the monitoring of gaseous releases, either combustible or radioactive.

Unfortunately, quantitative information on the rates of degradation of waste container materials by wastes or waste forms is rather sparse. Corrosion rates for carbon steel drum material in contact with unsolidified nonradioactive wastes of various kinds were summarized in Table 4.1, and drum lifetimes (assuming complete uniform corrosion of the drum wall) were inferred from the data. Thus, storage of unsolidified boric acid waste in carbon steel drums could be a concern for Phase II storage, but storage of unsolidified BWR chemical regenerative waste, BWR powdered resin waste, and low-activity (Class A) cation resins in carbon steel drums would probably not be a concern unless Phase III storage were to be implemented. Some quantitative information has been reported on the rate of corrosion of carbon steel by a particular solidified decontamination agent (Dow NS-1 in Dow vinyl ester-styrene binder); from the data (as discussed in Section 4.1.1 above), corrosion by this particular waste-binder system could be a concern for either Phase I or Phase II storage in carbon steel drums. Corrosion of carbon steel by other waste-binder materials does not seem to have been investigated in a systematic, quantitative manner except for some initial work by Colombo and Neilson,[6] who observed localized corrosion of the steel at the interface between cement waste forms and air. (Such a localized ring of internal corrosion could compromise the ability of the drum to withstand lifting loads and other stresses during handling after storage and thus repackaging could be required.) There are

also indications from tests of the corrosion of 304 stainless steel by irradiated cation resins (discussed in Section 3.1 above) that spent ion-exchange resins with Class C or possibly Class B radionuclide loadings could (perhaps as a result of radiolytic decomposition products) cause localized corrosion of stainless steel containers. In any case, the sparseness of the data on corrosion of steels by wastes and waste forms makes prediction of waste container performance lifetimes difficult. Because corrosion of the container by its contents, at least in the initial stages, cannot be monitored by visual inspection, ultrasonic measurements and other nondestructive examination techniques will be necessary for surveillance of stored waste packages which may be subject to corrosion.

Quantitative data on the rates of corrosion of carbon steel drum materials by the atmosphere and its constituents seem more readily available, but here, too, the uncertainties are large. Atmospheric corrosion during Phase I storage would not seem to be a problem if even minimal measures are taken, e.g., covering the stored containers with an air-inflated fabric weather shield. Only certain extremely aggressive atmospheres, e.g., marine atmospheres and some industrial atmospheres, are likely to cause damaging external corrosion of carbon steel drums during Phase I storage. Also, relatively simple visual surveillance may be used to monitor atmospheric corrosion of waste containers.

Biodegradative processes have been implicated in the generation of gases in and "spontaneous" heating of spent ion-exchange resins.(8) The time scales of the actual reported incidents indicate that such phenomena would be of concern during or after Phase I storage.

Radiolytic generation of flammable or explosive gases has been observed in laboratory tests from a variety of organic waste, binder, and container materials. For spent ion-exchange resins, the yield of radiolytically generated hydrogen gas calculated from the laboratory data is in reasonable accord with the yield measured in the field, i.e., from the Epicor II resins used in the Three Mile Island cleanup. (See the discussion in Section 3.1.1.) Radiolytic gas generation is likely to be a problem only for the highest-activity wastes, in particular, Class C wastes containing organic materials or water,* and then only during Phase I and, to a lesser extent, Phase II storage, when the activity of the waste is highest. Monitoring for flammable or explosive gas mixtures would be advisable for Class C wastes during Phase I and Phase II storage.

Radiolytic embrittlement of polyethylene HICs is discussed in Section 3.3, where it is noted that if the laboratory data for the ductile-to-brittle transition is extrapolated to field conditions, radiolytic embrittlement may occur in a year or less for highly loaded wastes (i.e., wastes which would receive a total accumulated dose of 10^8 rad). Such embrittlement could result in failure of the container to meet free drop and

*Even "dewatered" wastes may contain bound water.

lifting load requirements for transport and handling after storage. Repackaging of the waste for shipment may then be necessary. The time frame for such embrittlement based on the discussion in Section 3.3 is Phase I storage.

4.2 References

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4. Memorandum from R. E. Barletta, BNL, to R. E. Davis, "Review of CECO Corrosion Test Results," March 11, 1981, Attachment to Monthly Letter Report to NRC, FIN A-3159, April 1981.

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5. SUMMARY AND CONCLUSIONS

LLRW storage needs that may result from the unavailability of disposal capacity is a relatively new radwaste management concern in the United States. Most nuclear power plants were not designed with on-site LLRW storage capacity of extended duration, since 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. The areas of concern regarding the extended storage of LLRW need to be ascertained, and appropriate actions recommended to resolve these concerns must be determined.

5.1 Summary of 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, 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.

The potential storage time is a variable significantly affected 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 affecting the availability of disposal sites for LLRW are likely to be major considerations in determining the length of time for which storage of LLRW may be needed. For this report, 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 HVAC systems in this design. A minimum temperature of 50°F (10°C) is explicitly mentioned by one utility for its LLRW storage facility. An upper bound for the temperature of 80°F (27°C) is conservatively estimated and sharp variations even within this temperature range would not be expected. Since there was 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%. Temperatures 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). At the other extreme, the wastes in a simple fenced-in concrete storage pad will be exposed to outdoor temperatures and humidities, which in some locations may range over a year from below -40°F (-40°C) to above 104°F (+40°C) and from 0% to 100%, respectively. For such outdoor storage there is, of course, a significant potential for wetting of the container by rain or, in locations near bodies of salt water, by salt spray, which is very corrosive towards carbon steel.

For α and β radiation it is a good approximation that 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 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 this estimate that all the drums in the stacked array contain the same concentrations of γ emitters.)

In certain respects, the storage environment can be more severe than the disposal environment. According to guidance provided by the NRC to waste generators, under the expected disposal conditions Class B and C waste forms should maintain gross physical properties and identity over a 300-year period and high integrity containers should be designed to maintain their structural integrity over such a period. Yet, because of the variability of certain storage environments, waste packages which would be expected to meet the 300-year disposal lifetime criterion may suffer degradation over a much shorter extended storage period. Among the ways in which a storage environment can be more severe than a disposal environment are temperature fluctuations (in unheated facilities in areas with cold winters) and corrosive atmospheres (e.g., industrial and marine atmospheres, as well as acid deposition). Also, no subsequent handling of the waste package after disposal is anticipated. Stored waste packages, on the other hand, need to maintain sufficient integrity to prevent dispersal of the waste during storage, transport, and handling up to and including emplacement for disposal. Loss of waste package integrity prior to disposal will require repackaging of the waste.

The environmental variables discussed above are summarized in Table 5.1 in a largely qualitative manner for four categories of storage concepts. These four categories are large engineered structures, storage modules, shielded casks, and unshielded facilities. These categories of storage facilities are described in Section 2.2.5. 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 heating, ventilation and air-conditioning (HVAC) system.

Table 5.1

Summary of Extended Storage Environments

	Large Engineered Structures	Storage Modules	Shielded Casks	Unshielded Facilities
Temperature (°C)	Controlled 10 to 27	Uncontrolled, near ambient[a] -40 to +60	Uncontrolled, near ambient[a] -40 to +60	Uncontrolled, near ambient[a] -40 to +60
Relative Humidity	Controlled <50%	0% to 100%	0% to 100%	0% to 100%
Potential for Wetting	Negligible	Negligible to moderate	Negligible to moderate	Negligible to very large[b]
Radiation Field From Adjacent Packages	Potentially significant. Factor of up to 3[c]	Potentially significant[c]	Slight to potentially significant[d]	Potentially significant[c,e]

[a]Insolation may increase temperature to greater than ambient.
[b]Negligible in prefabricated structures. Very large in uncovered storage pad.
[c]Conservative ratio of dose to infinite medium (approximating stacked drums) to dose to 55-gallon drum for γ-radiation. Assumes multiple packages in storage structures.
[d]Slight if only one waste package per cask. Potentially significant if cask contains several packages.
[e]Not significant if such facilities are used only for storage of low-specific activity waste.

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, the containers may be without protection 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.

5.2 Summary of Waste and Waste Package Characteristics

In Sections 3.1, 3.2, and 3.3 above, the properties and behavior of low-level radioactive waste streams, solidification agents, and container materials have been reviewed, with emphasis on those characteristics important

for addressing the effects of extended storage on the waste and waste package. Because of the variations in non-fuel-cycle wastes, no general accounts of properties and behavior are available. Therefore, the characterization of waste streams in this report deals only with fuel-cycle wastes. An overview of the properties and behavior of the wastes, binders, and solidification agents is presented below. It must be emphasized that non-radiolytic effects are likely to be the primary concern for the majority of LLRW packages. Also, much of the testing from which the following information is drawn, particularly for phenomena involving radiation, were carried out under worst-case (or even beyond realistic worst-case) conditions in order to accelerate testing or for the sake of conservatism. For example, many of the radiation effects are based on data obtained from laboratory tests which were accelerated by the use of high doses ($>10^8$ rad) over a period of weeks or months.

5.2.1 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. According to the guidance given to LLRW generators in the NRC Technical Position (TP) on Waste Form, 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 TP is exceeded. It should be noted that the following radiation processes may be affected both quantitatively and qualitatively by the partial pressure of oxygen and by the dose rate.

  - Irradiation of ion-exchange resins may produce and/or 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 before their solidification in cement has been reported to improve the compressive strength and immersion resistance of the resulting waste forms.

- Biodegradation has been identified as a possible cause of agglomeration of and gas buildup in unsolidified ion-exchange resins during storage.

5.2.2 Other LWR Wastes

- The radiolytic generation of gases (predominantly hydrogen) and of corrosive substances has been observed in cellulosic materials.

- Because much of this waste consists of organic materials (e.g., cellulose) biodegradation is likely if the wastes have not been self-sterilized or treated with a biocide. 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.

5.2.3 Binder Materials: Cement

- Radiolytic gas generation observed in waste forms consisting of low-level waste solidified in cement has been attributed to radiolysis of water in the cement. Once again, the gas is predominantly hydrogen.

- 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.

- Freeze-thaw cycling can damage cements containing sufficient freezable water, particularly if mitigative measures have not been taken (e.g., air entrainment).

- Certain of the products of waste radiolysis, such as low-molecular weight organic acids, have been found to attack cement.

5.2.4 Binder Materials: Bitumens

- Radiolytic generation of gas has been observed in bitumens. 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, whether in the gas phase or incorporated into the solid matrix of oxidized bitumens.

- Biodegradation of bitumens has been observed. There is some evidence that corrosive substances may be produced as a result of biodegradation of bitumens.

5.2.5 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 waste forms increases the amount of this free liquid (up to 1.3 volume percent). Similar data on full-scale waste forms do not seem to be available.

5.2.6 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).

- 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, corrosion 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 the corrosion occurring during the first year.

- Corrosion of carbon steel containers result from 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. (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. Corrosion of metals in bitumen has been attributed to biodegradation. A corrosion rate of about 0.01 mil/day (4 mpy) is reported for mild steel embedded in waste forms consisting of a chelating decontamination reagent solidified in vinyl ester-styrene.

5.2.7 Container Materials: Polyethylene

- High density polyethylene (HDPE) 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 rather than degradation. Irradiation in air produces, in addition, radiolytic oxidation at the surface. This oxidized zone is believed to penetrate gradually 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 these radiolytic processes. Furthermore, the rate-limiting steps of irradiation-induced cross-linking and of radiolytic oxidation are thought to be activated processes, so that the rate will be temperature dependent. It is not clear, however, whether radiolytic oxidation of HDPE is an important degradation mode for containers stored at ordinary temperatures.

5.3 The Effects of Extended Storage on Waste Package Performance

Only a few of the data available are directly relevant to the performance of low-level waste packages during storage and subsequent handling (e.g., radiolytic gas generation data from the Epicor-II prefilter resins at TMI-2, atmospheric corrosion of steel containers of transuranic wastes) and thus their performance for the most part must be inferred from data on the characteristics of the storage environments and the properties of the waste package components. From the various data, the following problems, and the specific circumstances under which they may be expected to arise, are identified:

- external corrosion of steel containers stored outdoors,
- internal corrosion of steel containers,
- radiation-induced embrittlement of stored polyethylene containers, and
- radiolytic gas generation from stored ion-exchange resins and bituminized wastes.

Since the storage of non-fuel-cycle wastes at nuclear reactor LLRW storage facilities has been proposed the following concern about the storage of certain non-fuel-cycle wastes is also identified:

- biodegradation of institutional wastes.

In the following sections, those problems are discussed, mitigative measures are considered, and where applicable, NRC guidance in these matters is noted.

5.3.1 External Corrosion of Steel Containers Stored Outdoors

If steel containers of radwaste, especially carbon steel drums or liners commonly used for Class A and stabilized wastes, are stored outdoors, then the exposed surfaces of these containers will be subject to atmospheric corrosion. In principal, facilities such as simple fenced-in concrete pads are to be used only as holding areas prior to shipment for disposal, but in the event that disposal capacity should become temporarily and unexpectedly unavailable, such facilities may become de facto storage areas. As noted in Section 3.3.1, actual field data for the atmospheric corrosion of carbon steel containers are available. From these data, it has been concluded that uniform atmospheric

corrosion should not be a problem for the structural integrity of carbon steel drums since the estimated quantity of uniform corrosion over period of one to two decades represents only a fraction of the nominal 50- to 60-mil wall thickness of a typical 55-gallon carbon steel drum. However, non-uniform modes of corrosion, e.g., pitting corrosion and enhanced corrosion at welds, seams, and areas of moisture accumulation, may result in localized deterioration of the container and release of the contents of the drum or liner. For example, at the Idaho National Engineering Laboratory (INEL) and at Hanford, both low-humidity sites, carbon steel drums corroded mainly on the lids and at points of contact with the ground. Also, rusty 55-gallon drums received at the Richland disposal site had generally been in storage for at least six months.* Such corroded containers may not have sufficient structural integrity to withstand handling after storage and may not meet the disposal site acceptance criteria. Repackaging of the wastes, which will likely result in additional occupational exposure, may become necessary.

In Generic Letter 81-38 (reproduced as Appendix A), Section III(b), the NRC has provided guidance with regard to atmospheric corrosion of radwaste containers during storage. The effects of atmospheric corrosion upon steel containers may be mitigated by the selection of a more corrosion-resistant alloy as the container material or by use of protective coatings. (As noted in Section 3.3.1, the former option was chosen at Oak Ridge, a humid site, where mild steel drums were replaced by stainless steel drums.) It is further stated in Generic Letter 81-38 [in paragraph III(d)4] that steps should be taken to prevent corrosion of the containers by the weather and by accumulation of water.

An air support weather shield was used effectively at INEL, a dry site, to reduce corrosion of carbon steel drums (see Section 3.3.1). At more humid sites, condensation of moisture under such a simple structure may enhance corrosion and thus a simple storage shed may be more effective in limiting external corrosion of the containers. A large engineered storage facility with controlled temperature and humidity conditions can provide a relatively non-corrosive external environment for the waste containers, but such a facility is expensive. The degree of protection which a storage facility should provide will depend on the severity of the climate; while a simple air support weather shield may provide adequate protection against corrosion of carbon steel drums in a mild, dry climate, more elaborate facilities with some degree of temperature and humidity control may be necessary in humid climates with extreme temperatures and corrosive atmospheres (e.g., industrial or coastal areas). Monitoring of the stored containers in any of these facilities may be accomplished by visual inspection either directly or remotely, with due regard for minimization of occupational exposure. A program of at least quarterly visual inspection is specified in Generic Letter 81-38.

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*Mentioned by Ms. Nancy Kirner of the Washington State Department of Health and Social Services during her presentation at the Short Course on Radioactive Waste Management, sponsored by the American Society of Mechanical Engineers, Alexandria, VA, May 6-10, 1985.

5.3.2 Internal Corrosion of Steel Containers

Internal corrosion of the container material by the contents of the container is another possible mode of degradation of container performance during extended storage. As discussed in Section 3.3.1, there is relatively little quantitative information on the corrosion of carbon steel in contact with LLRW. Using available data and assuming uniform corrosion, the time for complete corrosion of an 18-gauge 55-gallon carbon steel drum was estimated to be one or two decades for unsolidified boric acid wastes and for a decontamination agent solidified in vinyl ester-styrene. Pitting corrosion may result in even earlier penetration of the drum wall. However, even if the container wall is not penetrated by pitting a gradual loss of structural strength will occur before complete corrosion of the container wall. As noted in Section 3.3.1, localized corrosion of carbon steel at the interface between the cement-solidified radwaste and the air has also been observed. Containers which have been corroded by interaction with their radwaste contents may not have sufficient structural integrity to withstand handling after storage and may not meet the disposal site acceptance criteria. In addition, there is the potential for release of the contents. Repackaging of the wastes will likely result in additional occupation exposure.

In Generic Letter 81-38, Section III(b), the NRC has provided guidance with regard to radwaste container corrosion caused by incompatability between the container materials and the wastes or waste forms. In accord with this guidance, the effects of corrosion of the steel container materials by the waste may be mitigated by the selection of a more corrosion-resistant alloy. As noted above in Section 3.3, special steel alloys have been proposed as container materials for high integrity container designs recently submitted for approval. Further, in Section 3.3.1, there are discussion of the use of protective coatings to mitigate corrosion of the container by the waste (in accord with guidance given in Section V(d)2 of the Generic Letter).

Corrosion-resistant materials such as stainless steels may be used to store most LLRW with a relatively high degree of assurance against corrosion of the waste container during storage. Selection of a container material will depend upon the corrosivity of the contents and on the anticipated length of the storage period. For example, carbon steel drums probably have sufficient resistance to corrosion by dry contaminated material such as paper or trash so that they may be used to store these materials for several years, neglecting external corrosion, but may not have adequate corrosion resistance for use in extended storage of dewatered (Class A) ion-exchange resin wastes or some solidified radwastes.

Monitoring of the stored containers for internal corrosion is more difficult than monitoring for external corrosion. Internal corrosion will not be detectable by visual inspection until the container has failed, either by penetration or by loss of structural integrity. Nondestructive examination techniques (e.g., ultrasonic probes) are available for detecting corrosion on

internal surfaces, but implementation of such techniques may result in an increase in occupational exposure.*

5.3.3 Radiation-Induced Embrittlement of Stored Polyethylene Containers

High-integrity containers (HICs) fabricated from high density polyethylene (HDPE) and containing high activity wastes may be subject to radiation-induced changes in properties during extended storage. Dose rate as well as the dose delivered to the HIC material can be important in determining the nature, magnitude, and rate of occurrence of such changes. Radiation-induced gas generation, oxidative degradation, and cross-linking have been observed in polyethylene materials (see Section 3.3.3); embrittlement resulting from the radiation-induced cross-linking is of concern for extended storage. Unfortunately, estimates of the time to reach the ductile-to-brittle transition at realistic dose rates, expected to be between 250 to 1500 rad/h, were obtained by extrapolation of data at higher dose rates, primarily between 2 and 100 krad/h. It was concluded that embrittlement of the HDPE material could occur within a year.(2) The container may then not withstand handling after storage and may no longer meet the acceptance criteria for HICs at a disposal site. Repackaging of the wastes may become necessary and will likely result in additional occupational exposure.

Although no explicit guidance is given by NRC in Generic Letter 81-38 with regard to changes in the properties of polymeric materials, the effects of radiation and aging should be considered in the design of and selection of materials for HICs. Alternatively, the waste could be stored in an on-site holding tank, if practicable, and not transferred to a HDPE HIC until immediately before shipment for burial.

5.3.4 Radiolytic Gas Generation From Stored Ion-Exchange Resins and Bitumized Wastes

Radiolytic generation of gases from ion-exchange resins has been observed both during irradiations in the laboratory and from heavily loaded spent resins in the field. On the basis of laboratory data, similar gas generation may be expected from heavily loaded bituminized wastes. It was noted in Section 4.1.1 that radiolytic hydrogen gas production is expected from both bitumens and ion-exchange resins. For example, it may be concluded from the calculations in Appendix C that a 55-gallon container of bituminized waste could, in principle, generate more than its own volume of gas in five years and result in pressurization of a gas-tight container. If the generated gas is released from the container into a confined unventilated storage area, the accumulated hydrogen gas could eventually exceed its lower flammability limit in air (9.5 volume percent at 25°C and 1 atm.)(3) On the basis of the discussion in Section 3.1.1, radiolytic gas generation in ion-exchange resins may

*A discussion of ultrasonic probes and other nondestructive examination techniques which might be adapted for use in monitoring internal corrosion of containers in extended storage facilities is given by S. D. Strauss.(1)

be accompanied by free liquid production. Breach of a container from pressurization or corrosive free liquids could necessitate further processing and repackaging of the wastes with the concomitant additional occupational exposure.

In Generic Letter 81-38, Section III(b), the NRC has provided guidance with regard to radiolytic and other kinds of gas generation from stored waste containers. In addition to this guidance, i.e., special vent designs to relieve container pressurization and one-year maximum storage times, adequate ventilation of the storage area may be necessary to prevent flammable or explosive gas accumulations. On the basis of the calculations of radiolytic gas generation in Appendix C, significant gas accumulations could, in principle, occur within one year. It is therefore recommended that if only limited disposal capacity is available, the highest activity waste be shipped for disposal first. (The NRC has recently included requirements regarding the generation of combustible gas mixtures in NRC Certificates of Compliance for transport packages. These conditions typically limit hydrogen generation to 5% by volume of the secondary container gas void during twice the expected shipment time.[4])

5.3.5 Biodegradation of Institutional Wastes

Since storage of non-fuel-cycle wastes at nuclear power reactor sites has been proposed, a few brief comments on the biodegradation of institutional wastes will be given here. (Generic Letter 85-14 on use of nuclear reactor sites for the storage of wastes not generated by the utility licensee is reproduced as Appendix D.) The institutional wastes subject to biodegradation during storage are biological wastes such as animal carcasses, animal bedding and excreta, and labeled culture media. Since such wastes may contain pathogenic organisms, biodegradative generation of gases and liquids can lead to pressurization and corrosion of containers and to dispersal of pathogens.

The gases and liquids produced from biological radwastes during storage as well as their rates and quantities of generation will depend on the microbes present, the nature of biological wastes, and the environmental conditions such as pH, temperature, moisture, and partial pressure of oxygen, i.e., aerobic vs anaerobic conditions. A discussion of biodegradative gas generation after disposal is given at Gause et al.[5] and the references cited therein.

Because of the uncertainties regarding biodegradation, attention should be given to packaging specifications for storage of biological pathogenic or infectious radwastes. Packaging for the disposal of such wastes has been considered, e.g., the NRC requires (in 10 CFR Section 61.56) that waste containing hazardous, biological, pathogenic, or infectious material must be treated to reduce to the maximum extent practicable the potential hazard from the non-radiological materials. Further, the site licensees for the LLRW disposal facilities have packaging criteria for the disposal of radioactive biological

wastes. If practicable, such wastes should either be stored for radioactive decay in refrigerated facilities to retard biodegradative processes or should be incinerated.

5.4 Reference

1. S. D. Strauss, "Nondestructive Examination," Power 51-510, (1983).

2. D. R. Dougherty et al., Brookhaven National Laboratory, "An Evaluation of the Effects of Gamma Irradiation on the Mechanical Properties of High Density Polyethylene," NUREG/CR-3898, BNL-NUREG-51802, December 1984.

3. J. A. Dean, Lange's Handbook of Chemistry, Twelfth Edition, p. 11-16, McGraw Hill, New York, 1979.

4. U. S. Nuclear Regulatory Commission, Office of Inspection and Enforcement, "Clarification of Conditions for Waste Shipments Subject to Hydrogen Gas Generation," IE Information Notice No. 84-72, September 10, 1984.

5. E. P. Gause et al., Brookhaven National Laboratory, "Alternative Containers for Low-level Wastes Containing Large Amounts of Tritium," NUREG/CR-3973, BNL-NUREG-51814, November 1984.

APPENDIX A

STORAGE OF LOW-LEVEL WASTE AT POWER REACTOR SITES (GENERIC LETTER 81-38)

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

November 10, 1981

TO ALL HOLDERS OF AND APPLICANTS FOR OPERATING LICENSES AND CONSTRUCTION PERMITS

SUBJECT: STORAGE OF LOW-LEVEL RADIOACTIVE WASTES AT POWER REACTOR SITES
(Generic Letter 81-38)

Gentlemen:

As a result of a reduction in waste disposal availability in the United States, many nuclear power reactor licensees are taking or are planning to take steps to provide for additional onsite storage of low-level radioactive wastes generated onsite. These steps range from storing packaged wastes in unused space to construction of new facilities for volume reduction and extended storage. The NRC has been considering the variety of plans which are underway and how they should be reviewed and approved.

Actions on waste storage can influence the development and implementation of final disposal plans by states, acting individually or on a regional basis, to establish additional disposal capacity. Some states have indicated to NRC that utilization of disposal services by nuclear power plant licensees is essential if disposal sites are to be developed by states or regional compacts. Thus, it is important that the NRC not take deliberate action that would hinder the establishment of additional disposal capacity by the states and yet, consistent with NRC regulatory safety requirements, permit necessary operational flexibility by its licensees. It is with these points in mind that the following guidance is provided.

For proposed increases in storage capacity for low-level waste generated by normal reactor operation and maintenance at power reactor sites, the safety of the proposal must be evaluated by the licensee under the provisions of 10 CFR 50.59. If (1) your existing license conditions or technical specifications do not prohibit increased storage, (2) no unreviewed safety question exists, and (3) the proposed increased storage capacity does not exceed the generated waste projected for five years, the licensee may provide the added capacity, document the 50.59 evaluation and report it to the Commission annually or as specified in the license.

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Radiological safety guidance has been developed by the staff for the design and operation of interim contingency low-level waste storage facilities. Necessary design features and administrative controls will be dictated by such factors as the waste form, concentrations of radioactive material in individual waste containers, total amount of radioactivity to be stored, and retrievability of waste. A copy of the guidance document is enclosed with this letter. This guidance shall be used in the design, construction and operation of your storage facility. In addition, the NRC will judge the adequacy of your 50.59 evaluation based on your compliance with the guidance. Please note also that IE Circular No. 80-19, dated August 22, 1980, provides information on preparing 50.59 evaluations for changes to radioactive waste treatment systems.

If you determine that an unreviewed safety question exists, authority for use should be requested through application to the Office of Nuclear Material Safety and Safeguards (NMSS) pursuant to 10 CFR 30, accompanied by an environmental evaluation that considers the incremental impact as related to reactor operations. Such application for a separate Part 30 license is for the administrative convenience of the Commission and is not intended to be substantively different than an application for amendment of the facility operating license. Application for use should also be accompanied by a showing that the storage provisions will not impact on the safety of reactor operations and will not foreclose alternatives for disposal of the wastes.

NMSS will notice the receipt of application in the Federal Register, offer an opportunity for public hearing if significant public interest is demonstrated, and will perform an environmental assessment to determine if the proposed activity will significantly affect the quality of the environment. Facility construction prior to the staff's determination would be carried out at the licensee's risk. Any license issued will be for a standard five-year term, renewable if continued need is demonstrated and if safety of continued storage is established. NRC licensing jurisdiction will be retained in Agreement States in accordance with 10 CFR 150.15(a)(1) for storage of low-level waste generated and stored onsite. Indemnity coverage will be provided under and in accordance with your existing indemnity agreement with the Commission.

If it is determined that the storage provisions could impact on the safety of reactor operations or an existing license condition or technical specification limit on the amount of waste storage, a change in the conditions of the reactor facility license may be necessary.

The provisions for added capacity should be used only for interim contingency storage, and low-level wastes should continue to be shipped to disposal sites to the extent practicable. The "Low Level Radioactive Waste Policy Act" of 1980 gives primary responsibility for the disposal of low-level waste to the states. Some states have initiated disposal plans, and we believe it is important that power reactor licensees, as major waste generators, work with and provide technical assistance and other support to assist individual states or regions in developing new disposal sites. You are encouraged to take an active role in the development of additional disposal sites.

Some licensees are considering the installation of major volume reduction processes, e.g., incineration, dehydration, or crystallization to substantially reduce the volume of waste for disposal. You are encouraged to examine the costs and benefits of such processes for your operations. However, notwithstanding the use of volume reduction, you are also encouraged to take an active role in the development of additional disposal sites.

For proposed increases in storage capacity for more than five years (long-term), the application and review procedures will be pursuant to 10 CFR 30 with consideration of container integrity and retrievability, volume reduction, influence on state planning for disposal, and implications of de facto onsite disposal. Any long-term license issued will be for a five-year, renewable term.

If you have any questions about these matters, please let us know.

Sincerely,

William J. Dircks
Executive Director
for Operations

Enclosure:
Guidance Document

Enclosure

RADIOLOGICAL SAFETY GUIDANCE FOR ONSITE CONTINGENCY STORAGE CAPACITY

I. Introduction

The objective of this technical position is to provide guidance to licensees considering additional onsite low level radioactive waste storage capabilities. While it may be prudent and/or necessary to establish additional onsite storage capability, waste should not be placed in contingency storage if the ability to dispose of waste at a licensed disposal site exists. The shipping of waste at the earliest practicable time minimizes the need for eventual waste reprocessing due to possibly changing burial ground requirements, reduces occupational and non-occupational exposures and potential accident consequences, and in the event of burial ground closure, maximizes the amount of storage space available for use.

The duration of the intended storage, the type and form of waste, and the amount of radioactive material present will dictate the safeguards and the level of complexity required to assure public health and safety, and minimal risk to operating personnel. The longer the intended storage period, the greater the degree of controls that will be required for radiation protection and accident prevention. For purposes of this document, the duration of temporary waste storage is to be up to five (5) years. The magnitude of the onsite storage safety hazard is predicated on the type of waste being stored, the amount of radionuclides present, and how readily they might be transported into the environment. In general, it is preferable to store radioactive material in solid form. Under some circumstances, however, temporary storage in a liquid form may be desirable or required. The specific design and operation of any storage facility will be significantly influenced by the various waste forms, consequently, this document addresses wet waste, solidified wet waste and dry low level radioactive waste.

Guidance similar to that provided in this enclosure has been incorporated in NUREG-0800, NRC/NRR Standard Review Plan, July 1981, as Appendix 11.4-A to SRP 11.4, Solid Waste Management Systems.

II. General Information

Prior to any implementation of additional onsite storage, substantial safety review and environmental assessments should be conducted to assure adequate public health and safety and minimal environmental impact. The acceptance criteria and performance objectives of any proposed storage facility, or area, will need to meet minimal requirements in areas of design, operations, safety considerations and policy

considerations. For purposes of this technical position, the major emphasis will be on safety considerations in the storing, handling and eventual disposition of the radioactive waste. Design and operational acceptability will be based on minimal requirements which are defined in existing SRPs, Regulatory Guides, and industry standards for proper management of radioactive waste. Considerations for waste minimization and volume reduction will also have to be incorporated into an overall site waste management plan and the onsite storage alternative. Additional waste management considerations for ALARA, decontamination, and decommissioning of the temporary storage facility, including disposal, should be performed as early as possible because future requirements for waste forms may make stored wastes unacceptable for final disposition.

Facility design and operation should assure that radiological consequences of design basis events (fire, tornado, seismic event, flood) should not exceed a small fraction (10%) of 10 CFR Part 100, i.e., no more than a few rem whole body dose.

The added capacity would typically extend storage to accommodate no more than an amount of waste generated during a nominal five-year period. In addition, waste should not be stored for a duration that exceeds five-years. Storage of waste in excess of the quantities and duration described herein requires Part 30 licensing approval. The design capacity (ft^3, Ci) should be determined from historical waste generation rates for the station, considering both volume minimization/reduction programs and the need for surge capacity due to operations which may generate unusually large amounts of waste.

The five-year period is sufficient to allow licensees to design and construct additional volume reduction facilities (incinerators, etc.), as necessary, and then process wastes that may have been stored during construction. Regional state compacts to create additional low-level waste disposal sites should also be established within the next five years.

III. Generally Applicable Guidance

(a) The quantity of radioactive material allowed and the shielding configurations will be dictated by the dose rate criteria for both the site boundary and unrestricted areas onsite. The 40 CFR 190 limits will restrict the annual dose from direct radiation and effluent releases from all sources of uranium fuel cycle and 10 CFR Part 20.105 limits the exposure rates in unrestricted areas. Offsite doses from onsite storage must be sufficiently low to account for other uranium fuel cycle sources (e.g., an additional dose of $\leq$ 1 mrem/year is

not likely to cause the limits of 40 CFR 190 to be exceeded). Onsite dose limits associated with temporary storage will be controlled per 10 CFR Part 20 including the ALARA principal of 10 CFR 20.1.

(b) Compatibility of the container materials with the waste forms and with environmental conditions external to the containers is necessary to prevent significant container corrosion. Container selection should be based on data which demonstrates minimal corrosion from the anticipated internal and external environment for a period well in excess of the planned storage duration. Container integrity after the period of storage should be sufficient to allow handling during transportation and disposal without container breach.

Gas generation from organic materials in waste containers can also lead to container breach and potentially flammable/explosive conditions. To minimize the number of potential problems, the waste form gas generation rates from radiolysis, biodegradation, or chemical reaction should be evaluated with respect to container breach and the creation of flammable/explosive conditions. Unless storage containers are equipped with special vent designs which allow depressurization and do not permit the migration of radioactive materials, resins highly loaded with radioactive material, such as BWR reactor water cleanup system resins, should not be stored for a period in excess of approximately one year.

A program of at least periodic (quarterly) visual inspection of container integrity (swelling, corrosion products, breach) should be performed. Inspection can be accomplished by use of TV monitors; by walk-throughs if storage facility layout, shielding, and the container storage array permit; or by selecting waste containers that are representative of the types of waste and containers stored in the facility and placing them in a location specifically designed for inspection purposes. All inspection procedures developed should minimize occupational exposure. The use of high integrity containers (300 year lifetime design) would permit an inspection program of reduced scope.

(c) If possible, the preferred location of the additional storage facility is inside the plant protected area. If adequate space in the protected area is not available, the storage facility should be placed on the plant site and both a physical security program (fence, locked and alarmed gates/doors, periodic patrols) and a restricted area for radiation protection purposes should be established. The facility should not be placed in a location that

requires transportation of the waste over public roads unless no other feasible alternatives exist. Any transportation over public roads must be conducted in accordance with NRC and DOT regulations.

(d) For low level dry waste and solidified waste storage:

1. Potential release pathways of all radionuclides present in the solidified waste form shall be monitored as per 10 CFR 50, Appendix A. Surveillance programs shall incorporate adequate methods for detecting failure of container integrity and measuring releases to the environment. For outside storage, periodic direct radiation and surface contamination monitoring shall be conducted to insure that levels are below limits specified in 10 CFR 20.202, 20.205, and 49 CFR 173.397. All containers should be decontaminated to these levels or below before storage.

2. Provisions should be incorporated for collecting liquid drainage including provisions for sampling all collected liquids. Routing of the collected liquids should be to radwaste systems if contamination is detected or to normal discharge pathways if the water ingress is from external sources and remains uncontaminated.

3. Waste stored in outside areas should be held securely by installed hold down systems. The hold down system should secure all containers during severe environmental conditions up to and including the design basis event for this waste storage facility.

4. Container integrity should be assured against corrosion from the external environment; external weather protection should be included where necessary and practical. Storage containers should be raised off storage pads where water accumulation can be expected to cause external corrosion and possible degradation of container integrity.

5. Total curie limits should be established based on the design of the storage area and the safety features provided.

6. Inventory records of waste types, contents, dates of storage, shipment, etc., should be maintained.

IV. <u>Wet Radioactive Waste Storage</u>

(a) Wet radioactive waste will be defined as any liquid or liquid/solid slurry. For storage considerations, wet waste is further defined

as any waste which contains free liquid in amounts which exceed the requirements for burial as established by the burial ground licensing authority.

(b) The facility supporting structure and tanks should be designed to prevent uncontrolled releases of radioactive materials due to spillage or accident conditions.

(c) The following design objectives and criteria are applicable for wet radioactive waste storage facilities:

1. Structures that house liquid radwaste storage tanks should be designed to seismic criteria as defined in Standard Review Plan (Section 11.2). Foundations and walls shall also be designed and fabricated to contain the liquid inventory which might be released during a container/tank failure.

2. All tanks or containers should be designed to withstand the corrosive nature of the wet waste stored. The duration of storage under which the corrosive conditions exist shall also be considered in the design.

3. All storage structures should have curbs or elevated thresholds with floor drains and sumps to safely collect wet waste assuming the failure of all tanks or containers. Provisions should be incorporated to remove spilled wet waste to the radwaste treatment systems.

4. All tanks and containers shall have provisions to monitor liquid levels and to alarm potential overflow conditions.

5. All potential release pathways of radionuclides (e.g., evolved gases, breach of container, etc.) shall be controlled, if feasible, and monitored as per 10 CFR 50, Appendix A (General Design Criteria 60 and 64). Surveillance programs should incorporate adequate methods for monitoring breach of container integrity or accidental releases.

6. All temporarily stored wet waste will require additional reprocessing prior to shipment offsite; therefore, provisions should be established to integrate the required treatment with the waste processing and solidification systems. The interface and associated systems should be designed and tested in accordance with the codes and standards described in Standard Review Plan Section 11.

V. Solidified Radioactive Waste Storage

(a) Solidified radwaste for storage purposes shall be defined as that waste which meets burial site solidified waste criteria. For purposes of this document, resins or filter sludges dewatered to the above criteria will be defined under this waste classification/criteria.

(b) Any storage plans should address container protection as well as any reprocessing requirements for eventual shipment and burial.

(c) Casks, tanks, and liners containing solidified radioactive waste should be designed with good engineering judgment to preclude or reduce the probability of occurrence of uncontrolled releases of radioactive materials due to handling, transportation or storage. Accident mitigation and control for design basis events (e.g., fire, flooding, tornadoes, etc.) must be evaluated and protected against unless otherwise justified.

(d) The following design objectives and criteria are applicable for solidified waste storage containers and facilities:

1. All solidified radwaste should be located in restricted areas where effective material control and accountability can be maintained. While structures are not required to meet seismic criteria, protection should be afforded to insure the radioactivity is contained safely by use of good engineering judgment, such as the use of curbs and drains to contain spills of dewatered resins or sludges.

2. If liquids exist which are corrosive, proven provisions should be made to protect the container (i.e., special liners or coatings) and/or neutralize the excess liquids. If deemed appropriate and necessary, highly non-corrosive materials (e.g., stainless steel) should be used. Potential corrosion between the solid waste forms and the container should also be considered. In the case of dewatered resins, highly corrosive acids and bases can be generated which will significantly reduce the longevity of the container. The Process Control Program (PCP) should implement steps to assure the above does not occur; provisions on container material selection and precoating should be made to insure that container breach does not occur during temporary storage periods.

3. Provision should be made for additional reprocessing or repackaging due to container failure and/or, as required for

final transporting and burial as per DOT and burial site criteria. Contamination isolation and decontamination capabilities should be developed. When significant handling and personnel exposure can be anticipated, ALARA methodology should be incorporated as per Regulatory Guides 8.8 and 8.10.

4. Procedures should be developed and implemented for early detection, prevention and mitigation of accidents (e.g., fires). Storage areas and facility designs should incorporate good engineering features and capabilities for contingencies so as to handle accidents and provide safeguard systems such as fire detectors and suppression systems, (e.g., smoke detector and sprinklers). Personnel training and administrative procedures should be estabished to insure both control of radioactive materials and minimum personnel exposures. Fire suppression devices may not be necessary if combustible materials are minimal in the area.

V. Low Level Dry Waste Storage

(a) Low level dry waste is classified as contaminated material (e.g., paper, trash, air filters) which contains radioactive material dispersed in small concentrations throughout large volumes of inert material and contains no free water. Generally, this consists of dry material such as rags, clothing, paper and small equipment (i.e., tools and instruments) which cannot be easily decontaminated.

(b) Licensees should implement controls to segregate and minimize the generation of low level dry waste to lessen the impact on waste storage. Integration of Volume Reduction (VR) hardware should be considered to minimize the need for additional waste storage facilities.

(c) The following design objectives and criteria are applicable for low level dry waste storage containers and facilities.

1. All dry or compacted radwaste should be located in restricted areas where effective material control and accountability can be maintained. While structures are not required to meet seismic criteria, protection should be afforded to insure the radioactivity is contained safely by use of good engineering judgment.

2. The waste container should be designed to insure radioactive material containment during normal and abnormal occurrences. The waste container materials should not support combustion. The packaged material should not cause fires through spontaneous chemical reactions, retained heat, etc.

3. Containers should generally comply with the criteria of 10 CFR 71 and 49 CFR 170 to minimize the need for repackaging for shipment.

4. Increased container handling and personnel exposure can be anticipated, consequently, all ALARA methodology should be incorporated per Regulatory Guides 8.8 and 8.10.

APPENDIX B

NRC GUIDANCE ON STORAGE FOR DECAY

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

June 25, 1980

TO ALL MEDICAL AND ACADEMIC LICENSEES

There are a number of steps licensees engaged in nuclear medicine practice and biomedical research can take under NRC rules to substantially reduce, and in some cases eliminate, the need to send radioactive waste to commercial low-level waste disposal facilities. By taking advantage of these alternatives and following good waste management practices, licensees can often reduce the risk of having their programs impacted through further curtailment of commercial waste disposal facilities. Some of the more important steps that can be taken are to:

1. Segregate radioactive waste from non-radioactive waste to reduce unnecessary volume. This simply requires a little time and discipline in the laboratory.

2. Hold waste with short-lived radionuclides in storage for decay to background levels, then dispose of it in the ordinary trash. This procedure requires a license amendment. (See Enclosure 1 for information to be submitted with the amendment request).

3. Release certain materials into the sanitary sewage system in accordance with 10 CFR Part 20.303. No license amendment is required but 10 CFR Part 20.303 should be carefully reviewed to stay within limits.

Judicious use of these three steps can substantially reduce the volume of waste shipped to burial grounds. Some nuclear medicine laboratories using only short-lived radionuclides can eliminate waste shipments.

Waste from biomedical research is generally somewhat more difficult to manage. Two of the most common problems are disposal of liquid scintillation counting waste (LSCW) and animal carcasses. The most frequently used radioisotopes in both are tritium and carbon-14. LSCW presents a particularly troublesome problem due to the flammability and toxicity of the solvents. Disposal of LSCW has been given special consideration by NRC. The staff has investigated alternatives to managing these wastes and the results have been published in NUREG-0656.

Consideration should be given to disposal by incineration for LSCW and laboratory animals containing small amounts of tritium and carbon-14. This method requires a license amendment; 10 CFR Part 20.305 contains the provisions for incineration. Enclosure 2 identifies the information to be submitted with an amendment request for incineration.

There are other provisions in the regulations that cover waste disposal. We have mentioned only the few that are most easily and commonly used. Other regulatory provisions include:

1. Disposal by burial in soil in accordance with 10 CFR 20.304 (A proposed rule change is under consideration to delete this provision. It will likely be replaced by a provision which requires specific approval by license amendment for burial).

2. Release as effluents to unrestricted areas pursuant to 10 CFR Part 20.106. In keeping with the ALARA concept, this method should normally be used only for releases incident to the procedures involved.

We suggest that you review and consider alternatives to commerical land burial for the management of your low-level radioactive waste. Implementation of some of these alternatives may require an amendment to your license. Amendment requests should be submitted to the Material Licensing Branch through the use of normal channels. Specific licensing questions concerning NUREG-0656 should be directed to the Material Licensing Branch (301) 427-4232. Copies of the NUREG-0656 may be obtained from the Division of Technical Information and Document Control, U.S. Nuclear Regulatory Commission, Washington, D.C. 20555.

Sincerely,

Wandy L. Miller

Wandy L. Miller, Chief
Material Licensing Branch
Division of Fuel Cycle and
Material Safety

Enclosures:

1. Information to be submitted When Requesting Amendment to Dispose of Radioactive Waste by Decay-In-Storage.

2. Information Required for Commission Approval of Treatment or Disposal by Incineration.

Enclosure 1

Information to be Submitted When Requesting Amendment to Dispose of Radioactive Waste by Decay-In-Storage Method

This is in reference to your request for information concerning authorization to dispose of radioactive waste via decay-in-storage. In order to approve such an amendment request, we need the following information:

1. Please submit a diagram of the area where the waste will be decayed-in-storage. Show the type, location, and thickness of shielding that you will have available in this area on your diagram. Your storage area should be large enough to handle an accumulation of used Tc-99m generators as well as other solid waste.

   Identify adjacent unrestricted areas located across the walls from the storage area and show that adequate steps have been taken to assure that radiation levels do not exceed the limits specified in 10 CFR 20.105 (enclosed).

2. Describe your security measures for the decay-in-storage area.

3. Confirm that radiation levels in this area will be surveyed and recorded at least weekly.

4. Describe your procedures for monitoring the waste to assure that it has decayed to background levels prior to disposal. As a minimum, your description should include these points:

   a. Monitor the waste in a low background area.

   b. Monitor with a low level GM type survey meter as appropriate for contamination surveys. Use the most sensitive scale.

   c. Remove all shielding prior to monitoring.

   d. Maintain records of these surveys as required under 10 CFR 20.

5. Note that decay-in-storage may not be a practical method of disposal for Tc-99m generators. These generators may contain long-lived radioisotopic contaminants. If you intend to dispose of generators by this method, you should include procedures for segregating the generator columns so that they may be monitored separately.

Be certain to submit your amendment request in duplicate. Unless your institution is fee exempt, your request should be accompanied by the appropriate amendment fee. Refer to 10 CFR 170.

Enclosure 2

INFORMATION REQUIRED FOR COMMISSION APPROVAL OF TREATMENT OR DISPOSAL BY INCINERATION

Revised October 3, 1979

1. State specifically the isotopes you wish to incinerate. For each isotope listed, you should submit calculations demonstrating that air concentrations of the effluents at the stack are in accordance with the requirements of Section 20.106 of 10 CFR Part 20.

2. Submit the characteristics of the incinerator such as height of the stack, height of and distance to buildings in the surrounding areas, rated airflow of the incinerator in cubic feet per hour or similar units and its proximity to any air intake ducts.

3. The gaseous effluent from the incinerator stack should not exceed the limits specified for air in Appendix B, Table II, 10 CFR Part 20, when averaged over a twenty-four (24) hour period.

4. In order to be in compliance with the ALARA philosophy stated in Section 20.1(c) of 10 CFR Part 20, the gaseous effluent from the incinerator stack should be a fraction (approximately 10%) of the limits specified for air in Appendix B, Table II, 10 CFR Part 20, when averaged over a one year period.

5. Describe the method of measurement or estimation of the concentration of radioactive material appearing in ash residue.

6. Describe the procedures for handling and disposing of ash from the incinerator.

7. Describe procedures to be followed to prevent overexposure of personnel during all phases of the operation, including instruction given to personnel handling the combustibles and the ash.

8. Submit evidence that all State and local regulations concerning incineration of radioactive material have been met by your institution.

9. State the maximum number of burns to be performed in any one week and the maximum number of burns per year.

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

SEP 12 1980

TO ALL INDUSTRIAL LICENSEES

There are a number of steps licensees engaged in industrial uses of licensed material can take under NRC rules to substantially reduce, and in some cases eliminate, the need to send radioactive waste to commercial low-level waste disposal facilities. By taking advantage of these alternatives and following good waste management practices, licensees can often reduce the risk of having their programs impacted through further curtailment of commercial waste disposal facilities. Some of the more important steps that can be taken are to:

1. Segregate radioactive waste from non-radioactive waste to reduce unnecessary volume. This simply requires a little time and discipline in use of radioactive materials at a licensee's facility.

2. Compact radioactive waste to the extent practicable to reduce volume.

3. Hold waste with short-lived radionuclides in storage for decay to background levels, then dispose of it in the ordinary trash. This procedure requires a license amendment unless your license already contains provision for this method. (See Enclosure 1 for information to be submitted with the amendment request).

4. Release certain materials into the sanitary sewage system in accordance with 10 CFR Part 20.203. No license amendment is required but 10 CFR Part 20.203 should be carefully reviewed to stay within limits.

5. Industrial licensees who use relatively long-lived sealed sources in general may only dispose of their sources by transfer to an authorized recipient. However, even in cases where only sealed sources are used, if the half-life of the source is less than 90 days, considerations should be given to holding the source in storage until it decays to background.

Judicious use of these five steps can substantially reduce the volume of waste shipped to burial grounds. Some industrial licensees using only short-lived radionuclides can eliminate waste shipments.

Waste from industrial licensees involved in biomedical research or similar activity is generally somewhat more difficult to manage. Two of the most common problems are disposal of liquid scintillation counting waste (LSCW) and animal carcasses. The most frequently used radioisotopes in both are tritium and carbon-14. LSCW presents a particularly troublesome problem due to the flammability and toxicity of the solvents. Disposal of LSCW has been given special consideration by NRC. The staff has investigated alternatives to managing these wastes and the results have been published in NUREG-0656.

Consideration should be given to disposal by incineration for LSCW and laboratory animals containing small amounts of tritium and carbon-14. This method requires a license amendment; 10 CFR Part 20.305 contains the provisions for incineration. Enclosure 2 identifies the information to be submitted with an amendment request for incineration.

There are other provisions in the regulations that cover waste disposal. We have mentioned only the few that are most easily and commonly used. Other regulatory provisions include:

1. Disposal by burial in soil in accordance with 10 CFR 20.304 (A proposed rule change is under consideration to delete this provision. It will likely be replaced by a provision which requires specific approval by license amendment for burial).

2. Release as effluents to unrestricted areas pursuant to 10 CFR Part 20.106. In keeping with the ALARA concept, this method should normally be used only for releases incident to the procedures involved.

We suggest that you review and consider alternatives to commercial land burial for the management of your low-level radioactive waste. Implementation of some of these alternatives such as compaction, hold for decay and incineration may require an amendment to your license. Amendment requests should be submitted to the Material Licensing Branch through the use of normal channels. If you have any questions concerning what actions require an amendment to your license or any other questions concerning NUREG-0656, they should be directed to the Material Licensing Branch (301) 427-4228. Copies of the NUREG-0656 may be obtained from the Division of Technical Information and Document Control, U.S. Nuclear Regulatory Commission, Washington, D.C. 20555.

Sincerely,

William H. Miller Jr.

for Vandy L. Miller, Chief
Material Licensing Branch
Division of Fuel Cycle and
Material Safety

Enclosures:
1. Information to be submitted When Requesting Amendment to Dispose of Radioactive Waste by Decay-In-Storage.

2. Information Required for Commission Approval of Treatment or Disposal by Incineration.

Enclosure 1

Information to be Submitted When Requesting Amendment to Dispose of Radioactive Waste by Decay-In-Storage Method

This is in reference to your request for information concerning authorization to dispose of radioactive waste via decay-in-storage. In order to approve such an amendment request, we need the following information:

1. Please submit a diagram of the area where the waste will be decayed-in-storage. Show the type, location, and thickness of shielding that you will have available in this area of your diagram.

   Identify adjacent unrestricted areas located across the walls from the storage area and show that adequate steps have been taken to assure that radiation levels do not exceed the limits specified in 10 CFR 20.105 (enclosed).

2. Describe your security measures for the decay-in-storage area.

3. Confirm that radiation levels in this area will be surveyed and recorded at least weekly.

4. Describe your procedures for monitoring the waste to assure that it has decayed to background levels prior to disposal. As a minimum, your description should include these points:

   a. Monitor the waste in a low background area.

   b. Monitor with a low level GM type survey meter as appropriate for contamination surveys. Use the most sensitive scale.

   c. Remove all shielding prior to monitoring.

   d. Maintain records of these surveys as required under 10 CFR 20.

Be certain to submit your amendment request in duplicate. Unless your activity is fee exempt, your request should be accompanied by the appropriate amendment fee. Refer to 10 CFR 170.

Enclosure 2

INFORMATION REQUIRED FOR COMMISSION APPROVAL OF TREATMENT OR DISPOSAL BY INCINERATION

Revised October 3, 1979

1. State specifically the isotopes you wish to incinerate. For each isotope listed, you should submit calculations demonstrating that air concentrations of the effluents at the stack are in accordance with the requirements of Section 20.106 of 10 CFR Part 20.

2. Submit the characteristics of the incinerator such as height of the stack, height of and distance to buildings in the surrounding areas, rated airflow of the incinerator in cubic feet per hour or similar units and its proximity to any air intake ducts.

3. The gaseous effluent from the incinerator stack should not exceed the limits specified for air in Appendix B, Table II, 10 CFR Part 20, when averaged over a twenty-hour (24) hour period.

4. In order to be in compliance with the ALARA philosophy stated in Section 20.1(c) of 10 CFR Part 20, the gaseous effluent from the incinerator stack should be a fraction (approximately 10%) of the limits specified for air in Appendix B, Table II, 10 CFR Part 20, when averaged over a one year period.

5. Describe the method of measurement or estimation of the concentration of radioactive material appearing in ash residue.

6. Describe the procedures for handling and disposing of ash from the incinerator.

7. Describe procedures to be followed to prevent overexposure of personnel during all phases of the operation, including instruction given to personnel handling the combustibles and the ash.

8. State the maximum number of burns to be performed in any one week and the maximum number of burns per year.

9. Submit confirmation that you have complied with all state and local regulations concerning incineration.

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

JUN 04 1981

TO ALL MEDICAL LICENSEES:

On June 23, 1980 all medical and academic licensees were sent a letter describing steps that they could take to substantially reduce or eliminate radioactive waste sent to commercial low-level waste disposal facilities. One of these steps was to hold radioactive waste in storage for decay to background levels before disposal in ordinary trash. For those licensees who do not have decay-in-storage as a method for disposal of radioactive waste in their NRC license, this requires a license amendment.

In order to ease the burden of applying for an amendment to your license for decay-in-storage of radioactive waste, we have decided that we will place a condition on all medical and academic licenses which states:

"The licensee is authorized to hold radioactive material with a physical half-life of less than 65 days for decay-in-storage before disposal in ordinary trash provided:

a. Effected radioactive waste shall be held for decay a minimum of ten (10) half-lives.

b. Prior to disposal as normal waste, radioactive waste shall be monitored to determine that its radioactivity cannot be distinguished from background with typical low-level laboratory survey instruments. All radiation labels will be removed or obliterated.

c. Generator columns shall be segregated so that they may be monitored separately to ensure decay to background levels prior to disposal."

There are two ways that the above condition can be incorporated into your license:

1. Without your prior request, we will automatically place this condition on all medical byproduct material licenses as they are issued in response to new or renewal applications and amendment requests; or

2. If you desire to have this condition placed on your present license right away, you should submit a request for amendment referencing this document. This type of amendment request will be fee exempt.

You are reminded of the requirements contained in 10 CFR 20.105 and 10 CFR 20.207, which address established limits for radiation levels in unrestricted areas and storing or securing radioative material respectively.

Yandy L. Miller, Chief
Material Licensing Branch
Division of Fuel Cycle and
Material Safety, NMSS

APPENDIX C

DOSE AND RADIOLYSIS CALCULATIONS FOR EXTENDED STORAGE OF LLRW

In Section 4.1 of this report radiolytic gas generation from LLRW during storage has been identified as a possible problem. As a result of such gas generation, pressurization of gas-tight containers may occur. Furthermore, since much of the radiolytically-generated gas may be hydrogen and light hydrocarbons, an explosion or flammability hazard may be created under certain storage conditions.[1] In addition, a potential for acceleration of container corrosion by radiolytic acid generation was also discussed in Section 4.1; the possibility of such corrosion after disposal has been noted[2] and should be considered for extended storage. In this appendix, the absorbed dose for each of several radionuclides is calculated for activity loadings at the numerical limits specified in 10 CFR Section 61.55. The absorbed dose is then used to calculate the resulting radiolysis products for several waste materials.

The calculations of the absorbed dose resulting from the radioactive decay of the radionuclides in a 55-gallon drum waste package follows the method outlined by McFarland[3] and described in more detail by Swyler, Barletta, and Davis.[4] The expressions for the total absorbed beta and gamma doses due to the complete decay (i.e., at time $t = \infty$) of a radionuclide may be written as

$$D^{\beta}(\infty) = 2.65 \times 10^4 \; t_{1/2} \; \overline{E}_{\beta} C \qquad (C.1)$$

$$D^{\gamma}(\infty) = 1.77 \times 10^3 \; t_{1/2} \; \Gamma C \qquad (C.2)$$

respectively, where $\overline{E}_{\beta}$ is the average beta energy in MeV, Γ is the gamma ray constant in rad-cm^2/mCi-h, C is the radionuclide activity concentration in Ci/m^3 ($=10^{-3}$ mCi/cm^3 = μCi/cm^3), and $t_{1/2}$ is the half-life in years. For the $D^{\gamma}(\infty)$ calculation, the value for $\overline{g}$, the average geometrical factor for gamma ray absorption, was taken as 140 cm and is included in the numerical constant in Equation C.2. The value for $\overline{g}$ was interpolated from a table of such values in a standard work on radiation dosimetry.[5] The radionuclide is assumed to be uniformly distributed throughout the volume of a 55-gallon drum with a 29-cm radius and a 86-cm height.[6] Tissue equivalency for γ-ray absorption, unit mass density of the waste material in the drum, and a near-unity value for the rad/roentgen ratio[7] are also assumed. The last assumption allows use of the published values of Γ, which are generally tabulated in units of R-cm^3/mCi-h.[8] As noted in connection with similar calculations by MacKenzie, Lin, and Barletta,[9] estimation of the value of $\overline{g}$ involves the greatest uncertainty in the calculation of the gamma dose. The absorbed dose at any time t may be found from

$$D^{\beta}(t) = D^{\beta}(\infty) \; [1 - \exp(-0.693t/t_{1/2})] \qquad (C.3)$$

and

$$D^{\gamma}(t) = D^{\gamma}(\infty)\ [1 - \exp(-0.693t/t_{1/2})] \qquad (C.4)$$

for beta and gamma radiation, respectively. These are essentially the same equations used by Swyler, Barletta, and Davis[4] and by Dougherty and Adams[10] expressed in terms of $t_{1/2}$, half-life, rather than decay constant, with ln2 approximated by 0.693. Values for D(∞) and D(t = 5 years) for selected nuclides at their 10 CFR Part 61 activity limits are presented in Table A.1 of this Appendix. For Cs-137 and Sr-90/Y-90, the five-year cumulative dose is in excess of the maximum cumulative dose of 10^8 rad for ion-exchange resins and high-integrity containers, which is specified in the TP as guidance for waste generators. Note that over a 5-year period Co-60, Cs-137, and the Sr-90/Y-90 couple impart the largest absorbed doses for any of the tabulated radionuclides at the specified activity limits. (The assumption that LLRW will contain any given radionuclide at its upper limit is probably quite conservative for most situations, although prior to the effective date of 10 CFR Part 61 occasional waste packages with radionuclide loadings in excess of present Class C loadings were shipped[2,11].)

A crude and conservative calculation of the dose absorbed by a waste package from its radioactive contents may be performed by assuming that all radiation emitted by radionuclides within the waste package is absorbed within the waste package. This approximation is implicitly assumed in the β-dose calculations in this Appendix and is a fairly good approximation for β-rays, whose maximum range--the thickness of absorbing medium required to stop the most energetic of the β-rays under consideration--is generally a few centimeters at most (e.g., 1.2 cm for a maximum β-ray energy of 2.5 MeV).[12] On the other hand, a considerable fraction of the γ-ray energy will not be absorbed within the package in which the γ-rays originate. The average geometrical factor $\overline{g}$, also discussed in this appendix, has been derived to take account of this. A calculation of the absorbed dose assuming no loss of γ-radiation from the package is still of some interest, however, because it provides an upper bound for the absorbed γ-ray dose and does not depend on either geometry or assumptions of tissue equivalency for γ-ray absorption. Such a calculation also provides an approximation to the effect of the γ-radiation field of adjacent waste packages on the package in question. The effect of intervening air spaces and container walls on the γ radiation from adjacent packages is neglected in this approximation and, in effect, an infinite absorbing medium with an activity concentration equal to that of the waste is assumed.

For example, the energy per disintegration of Co-60 (obtained by adding the γ-ray energy and average β energy) totals 2.6 MeV. The initial dose rate D(0) to a unit density medium containing this isotope at an initial activity concentration of 700 Ci/m^3 (the upper limit specified in 10 CFR Part 61) may be found as follows:

$$\dot{D}(0) = \frac{2.6 \text{ MeV/dis} \times 700 \text{ Ci/m}^3 \times 3.7 \times 10^{10} \text{ dis/Ci-s}}{6.24 \times 10^{13} \text{ eV/rad-g}}$$

$$= 1.08 \text{ rad/s}$$

Essentially, all that has been done is to multiply the energy per disintegration by the number of disintegrations per second. $D(\infty)$, the total absorbed dose due to complete decay of the isotope may be found by integrating the product of $D(0)$ and an exponential decay factor over time from $t=o$ to $t=\infty$. Omitting the details, the result is

$$D(\infty) = 4.55 \times 10^7 \, t^{1/2} \, \dot{D}(0)$$

when $t_{1/2}$ is the half-life in years. For Co-60 at an activity density of 700 Ci/m^3, $t_{1/2}$ = 5.26y and thus $D(\infty) = 2.6 \times 10^8$ rad. In the calculations for $D^{\beta}(\infty)$ and $D^{\gamma}(\infty)$ above, where the γ-ray losses from a 55-gallon drum are accounted for by means of the geometric factor $\bar{g}$, the value of $D(\infty)$ $[= D^{\beta}(\infty) + D^{\gamma}(\infty)]$ is calculated as 9.5×10^7 rad, about a factor of three lower. It should be noted that about 96% of the Co-60 radiation is emitted as γ-rays, for which the dose is geometry-dependent. A similar calculation for Cs-137 yields $D(\infty) = 3.2 \times 10^9$ rad for the complete absorption of γ-rays as compared to 1.5×10^9 rad for the 55-gallon drum. See Table C.2.

The yields of chemical species produced via radiolysis and appearing in homogeneous distribution in the medium or material under consideration are known as G-values and are defined in terms of the number of altered species produced (e.g., radicals, molecules, or functional groups formed or decomposed) per 100 eV of absorbed radiant energy.(13) Except for chain reaction processes such as polymerization, G-values between 1 and 5 are most common.(14) An absorbed dose of 1 rad (=100 erg/g = 6.24×10^{13} eV/g) will thus produce $6.24 \times 10^{11} \cdot G$ altered species per gram. On a gram-molecular-weight (or molar) basis, an absorbed dose of D rad will result in the production of $1.03 \times 10^{-12} \cdot G \cdot D$ moles/g of altered species. The yield of radiolytically generated species due to an absorbed dose D in a 55-gallon drum of radioactive waste (assuming unit density for the waste, i.e., 2.1×10^5 g of waste) is about $2.16 \times 10^{-7} \cdot G \cdot D$ moles. Differences in yield between various types of radiation have not been considered, but fast electron (i.e., beta) and gamma radiations are considered to have similar effects in aqueous systems.(13) The radiolytic gas generation from plastic, cellulosics, and bitumen and the acid generation from cellulosics in Table C.3 have been calculated for the three nuclides (Co-60, Cs-137, Sr-90/Y-90) which were found to impart the largest 5-year doses at their 10 CFR Part 61 limits. Apparent dose and dose-rate dependences of the G-values are not considered in the present calculations but are discussed in Section 4.1 of the main text. The gas generation over a 5-year period can be on the order of 1000 L or an average of about 1/2 L of gas per day. It must be stressed that these caluclations are for worst-case conditions with respect to activity concentrations, which are at the maximum values specified in 10 CFR Part 61 Section 55. Only a small percentage of the waste volume, probably less than 5%, will contain radionuclide concentrations even approaching these values.

Table C.1

Dose Calculations for Selected Radionuclides at Their 10 CFR Part 61 Limits in a 55-Gallon Drum

Nuclide	$t_{1/2}$ yr	Upper Limit Activity Concentration[a] C_{max} 10^{-3} mCi/cm^3	$E_\beta(8)$ MeV	$D_\beta(\infty)$ rad	D_β(5 yr) rad	rad-cm^2 / mCi-h	$D_\gamma(\infty)$ rad	D_γ(5 yr) rad	D_{total} (5 yr) rad	D_{total} (10 yr) rad	D_{total} (15 yr) rad
C-14	5720	8	0.49	5.9×10^8	3.6×10^5	----	----	----	3.6×10^5	7.2×10^5	1.1×10^6
C-14(am)[b]	5720	80	0.49	5.9×10^9	3.6×10^6	----	----	----	3.6×10^6	7.2×10^6	1.1×10^7
Ni-59 (am)[b]	1×10^5	220	----	----	----	5.5	2.1×10^{11}	7.4×10^6	7.4×10^6	1.5×10^7	2.2×10^7
Nb-94 (am)[b]	2×10^4	0.2	0.156	1.7×10^7	2.9×10^3	7.5	5.3×10^7	9.2×10^3	1.2×10^4	2.4×10^4	3.6×10^4
Tc-99 (am)[b]	2.1×10^5	3	0.085	1.4×10^9	2.3×10^4	----	----	----	2.3×10^4	4.6×10^4	6.9×10^4
I-129	1.7×10^7	0.08	0.040	1.4×10^9	2.9×10^2	0.05	1.2×10^8	2.4×10^1	3.1×10^2	6.2×10^2	9.3×10^2
H-3	12.3	>40[a]	0.005	6.5×10^4	1.6×10^4	----	----	----	1.6×10^4	2.8×10^4	3.7×10^4
Co-60	5.26	>700[a]	0.094	9.2×10^6	4.4×10^6	13.2	8.6×10^7	4.1×10^7	4.5×10^7	7.0×10^7	8.2×10^7
Ni-63	92	700	0.017	2.9×10^7	1.1×10^6	----	----	----	1.1×10^6	2.1×10^6	3.1×10^6
Ni-63 (am)[b]	92	7000	0.017	2.9×10^8	1.1×10^7	----	----	----	1.1×10^7	2.1×10^7	3.1×10^7
Sr-90	28.1	7000	0.200	1.1×10^9	1.2×10^8	----	----	----	6.8×10^8	1.3×10^9	1.9×10^9
(Y-90)	c		0.931	4.9×10^9	5.6×10^8	----	----	----			
Cs-137	30	4600	0.195	7.1×10^8	7.8×10^7	3.3	8.1×10^8	8.8×10^7	1.6×10^8	3.1×10^8	4.4×10^8

[a] C_{max} is the upper limit for the 10 CFR Part 61 Class C concentrations except for Co-60 and H-3, for which only a Class A upper limit is given in 10 CFR Part 61.
[b] (am) - in activated metal.
[c] Y-90 is a decay daughter of Sr-90 with a 64-h half-life and is in secular equilibrium with its parent.

Table C.2

Effect of Assumption of Complete γ-Ray Absorption on Dose Calculation

Radionuclide	Activity Concentration C Ci/m³	Average Beta Energy(8) $\overline{E\beta}$ MeV	Gamma Energy(8) Eγ MeV	Energy per Decay to Stable Product MeV	Initial Dose Rate D(0) rad/s		Absorbed Dose D(∞) 10^8 rad	
					Complete Absorption	55-gallon drum	Complete Absorption	55-gallon drum[b]
Co-60	700	0.094	1.173(100%) 1.332(100%)	2.6	1.1	0.40	2.6	0.95
Cs-137	4600	0.195	0.662	0.86	2.3	1.1	32	15
Sr-90[c]	7000	0.200	----	1.1	4.6	4.6	59	59
(Y-90)[c]		0.931	----					

[a]These concentrations are the largest values for the respective isotopes as specified in 10 CFR Part 61 Section 55. Only a very small portion (<5%) of LLRW is likely to have concentrations even approaching these values.
[b]From Table C.1.
[c]Included for comparison purpose only. Sr-90 and its Y-90 daughter are pure beta emitters.

Table C.3

Five-Year Generation of Radiolysis Products for Selected Waste Media in 55-Gallon Drums

Radionuclide	D^{total} (5-yr)[a] (from Table C.1) (rad)	Gas Generation[b] Plastics (G=3.7)	Gas Generation[b] Cellulosics (G=0.63)	Gas Generation[b] Bitumen (G=0.4)	Acid Generation Cellulosics (G=3.6)
Co-60	4.5×10^7	36 moles 8.1×10^2 L	6.1 moles 1.4×10^2 L	4 moles 9×10^1 L	35 moles
Sr-90 (Y-90)	6.8×10^8	5.4×10^2 moles 1.2×10^4 L	93 moles 2.1×10^3 L	6×10^1 moles 1×10^3 L	5.3×10^2 moles
Cs-137	1.6×10^8	1.3×10^2 moles 3.0×10^3 L	23 moles 5.1×10^2 L	1×10^1 moles 3×10^2 L	1.3×10^2 moles

[a]From Table C.1.
[b]Gas volumes at STP. G-values from References 1 and 2.

Alternatively, the yields of radiolytically-generated chemical species can be expressed in terms of gram-molecular-weight of product per unit activity per unit time or, for gases, in terms of L/Ci-h assuming ideal gas behavior. The utility of this alternative representation as a radiolytic product generation rate is the relative ease of comparison with empirical data. For example, the expressions in the previous paragraph for yield of radiolytic species may be converted to yield rate expressions by replacing D, the absorbed dose, with D(0), the initial absorbed dose rate. After multiplying by 22.4 L/mole and 3600 s/h, the yield rate of a radiolytically-generated ideal gas is found to be $8.31 \times 10^{-2} \cdot G \cdot D(0)$ (in units of L/h). To obtain the yield rate per unit activity for gases generated by radiolysis of materials containing one of the three isotopes addressed in Table C.2, the respective value of D(0) and the G-value for the material of interest should be substituted into the expression for the yield rate, which should then be divided by the respective value of activity concentration from Table C.2. As noted in Section 3.1 of the main text, the G-values for the radiolytic generation of hydrogen gas from fully-swollen sulfonic acid resins are found to range from 0.1 to 0.6[15] Using these G-values and the values of D(0) (for a 55-gallon drum) and C, the activity concentration, from Table C.2, the rates per unit activity of the radiolytic hydrogen gas generation from such resins containing Co-60, Cs-137, or Sr-90 have been calculated and are presented in Table C.4. For comparison, the radiolytic generation of hydrogen gas from the ion-exchange resins in the Epicor-II pre-filters used in the cleanup of contaminated water at Three Mile Island Unit 2 has been reported to proceed at a rate of 5.9×10^{-6} L/Ci-h.[16]

Table C.4

Radiolytic Hydrogen Gas Generation Rate per Unit Activity for Sulfonic Acid Resins Containing Co-60, Cs-137, or Sr-90

Radionuclide	Hydrogen Gas Generation Rate L/Ci-h	
	G = 0.1	G = 0.6
Co-60	4.8×10^{-6}	2.9×10^{-5}
Cs-137	2.0×10^{-6}	1.2×10^{-5}
Sr-90	5.5×10^{-6}	3.3×10^{-5}

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APPENDIX D

COMMERCIAL STORAGE AT POWER REACTORS OF LLRW NOT GENERATED BY THE UTILITY (GENERIC LETTER 85-14)

UNITED STATES
NUCLEAR REGULATORY COMMISSION
WASHINGTON, D. C. 20555

August 1, 1985

TO ALL LICENSEES

SUBJECT: COMMERCIAL STORAGE AT POWER REACTOR SITES OF LOW-LEVEL RADIOACTIVE WASTE NOT GENERATED BY THE UTILITY (Generic Letter 85-14)

Gentlemen:

The Low-Level Radioactive Waste Policy Act of 1980 (P. L. 96-573) assigned to the states the responsibility to provide for disposal of commercial low-level radioactive waste (LLW) generated within each state. The Act envisioned that all states would be capable of providing for disposal of commercial LLW generated within their borders by 1986. Based on the current status of state efforts and the substantial time required to establish new disposal facilities, no new sites will be available for at least several years. Due to the uncertainty of this situation and statements made by some officials of states within which currently operating disposal sites are located, it appears possible that access to the existing sites may be restricted.

While some licensees have taken steps to temporarily store LLW generated at their sites to alleviate any impact that limiting of access to disposal capacity may have on licensed operations, provisions for storing LLW should be used only for interim contingency purposes. It is the policy of the NRC that licensees should continue to ship waste for disposal at existing sites to the maximum extent practicable.

In anticipation of possible curtailment of access to existing disposal facilities, interest is being expressed in some states in commercial storage of LLW generated within the states. While the NRC recognizes that storage may appear desirable in states which have not resolved their low-level waste disposal problems, commercial storage facilities, however, should not become de facto disposal sites. NRC will require for commercial storage under its jurisdiction that, in addition to safe siting and operation, commitments and assurances be made for eventual disposition of all waste stored at commercial storage locations. This includes provisions for repackaging (if necessary), transportation and disposal of the waste, as well as decommissioning of the facilities.

Some of the concepts for commercial storage involve using nuclear power reactor sites as commercial storage locations for LLW not generated by the utility licensee. As a matter of policy, the NRC is opposed to any activity at a nuclear reactor site which is not generally supportive of activities authorized by the operating license or construction permit and which may divert the attention of licensee management from its primary task of safe operation or construction of the power reactor. Accordingly, interim storage of LLW within the exclusion area of a reactor site, as defined in 10 CFR 100.3(a), will be subject to NRC jurisdiction regardless of whether or not the reactor is located in an Agreement State, pursuant to the regulatory policy expressed in 10 CFR 150.15(a)(1). Within Agreement States, for locations outside the exclusion areas, the licensing authority is in the Agreement State.

In order for NRC to consider any proposal for commercial storage at a reactor site, including commercial storage in existing low-level waste storage facilities, the NRC must be convinced that no significant environmental impact will result and that the commercial storage activities will be consistent with and not compromise safe operation of the licensee's activities, including diverting reactor management attention from the continued safety of reactor operations. A Part 30 license is required for the low-level waste storage and a Part 50 license amendment may also be required. The application must include:

By the utility

- A determination by the utility licensee that the proposed low-level waste commercial storage activities do not involve a safety or environmental question, and that safe operation of the reactor will not be affected. In making this determination, the licensee shall consider:

  - Direct impacts of the commercial storage operation on reactor operations during normal and accident conditions;
  - Diversion of utility management and personnel attention from safe reactor operation;
  - Combined effects of onsite and offsite dose during normal and accident conditions;
  - Influence on effectiveness of reactor emergency plans;
  - Influence on effectiveness of reactor security plans;
  - Financial liability provisions, including impact on indemnity coverage; and
  - Environmental impact of the storage facility, including potential interaction with the generating station.

By the applicant (the utility or another person)

- Information relating to the safety of the commercial storage operation;

- Information relating to the environmental impact of the storage operation in sufficient detail to allow staff to establish the need for preparation of an Environmental Impact Statement;

- Financial assurance to provide for the commercial storage operation and decommisioning including any necessary repackaging, transportation and disposal of the waste; and

- Written agreement from the jurisdiction responsible for ultimate disposal, the State, that provisions are sufficient to assure ultimate disposal of the stored waste.

The Office of Nuclear Reactor Regulation (NRR) will conduct an environmental review and review the application to determine whether the low-level waste commercial storage activities on a reactor site impact the safe operation of the reactor. Following NRR review, the licensing authority for commercial storage on a reactor site under NRC jurisdiction (all locations in non-Agreement States and locations within reactor exclusion areas in Agreement States) is the Office of Nuclear Material Safety and Safeguards. The NRC will assess

environmental impact and will issue an Environmental Impact Statement, if appropriate, in accordance with provisions of 10 CFR 51.20, 51.21 and 51.25. As part of the procedures, the NRC will provide notice in the FEDERAL REGISTER of receipt and availability of any application received for commercial storage activities. The public notice will also indicate the staff's intent regarding preparation of an environmental assessment and its circulation for public review and comment. An Environmental Impact Statement will most likely be needed based on the environmental assessment.

Because the NRC has not yet received or reviewed an application for a centralized commercial low-level waste storage facility intended to store large amounts of LLW for five or more years, the NRC may consider applying the criteria described above to such commercial storage facilities whether they be on a reactor site or not.

Interim storage of utility licensee-generated LLW will continue to be considered according to the provisions stated in Generic Letter 81-38, dated November 10, 1981.

For additional information, please contact Frank Miraglia, Office of Nuclear Reactor Regulation, U. S. Nuclear Regulatory Commission, Washington, D.C. 20555 [Telephone: (301) 492-7980] or Richard Cunningham, Office of Nuclear Material Safety and Safeguards, U. S. Nuclear Regulatory Commission, Washington, D.C. 20555 [Telephone: (301) 427-4485].

Sincerely,

William J. Dircks
Executive Director
for Operations

NRC FORM 335 (2-84) NRCM 1102, 3201, 3202

U.S. NUCLEAR REGULATORY COMMISSION

BIBLIOGRAPHIC DATA SHEET

SEE INSTRUCTIONS ON THE REVERSE.

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12. SUPPLEMENTARY NOTES

13. ABSTRACT (200 words or less)

If a state or state compact does not have adequate disposal capacity for low-level radioactive waste (LLRW), then extended storage of certain LLRW may be necessary. Extended storage of LLRW is considered in order to determine for the Nuclear Regulatory Commission areas of concern and actions recommended to resolve these concerns. The focus is on the properties and performance 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 are grouped into two categories:

1. Performance 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 embrittlement of high-density polyethylene and the weakening of steel containers resulting from corrosion).

A discussion is given of additional information and actions required to address these concerns.

14. DOCUMENT ANALYSIS – a. KEYWORDS/DESCRIPTORS

low-level radioactive waste, extended storage, storage environments, cement, bitumen, ion-exchange resins, corrosion, gas generation, polyethylene, radiation effects

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