BNL-NUREG--37848

TI86 009436

EXTENDED STORAGE OF LOW-LEVEL RADIOACTIVE WASTE: AN UPDATE*

B. Siskind
Department of Nuclear Energy
Brookhaven National Laboratory
Upton, New York 11973

Received by OSTI
APR 22 1986

ABSTRACT

If a state or regional compact does not have adequate disposal capacity for low-level radioactive waste (LLRW), then extended storage of certain LLRW may be necessary. The Nuclear Regulatory Commission (NRC) has contracted with Brookhaven National Laboratory to address the technical issues of extended storage. 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 circumstances under which extended storage of LLRW would most likely result in problems during or after the extended storage period are considered and possible mitigative measures to minimize these problems are discussed. These potential problem areas include: (1) the degradation of carbon steel and polyethylene containers during storage and the subsequent need for repackaging (resulting in increased occupational exposure), (2) the generation of hazardous gases during storage, and (3) biodegradative processes in LLRW.

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 encouraged the formation of interstate compacts which (subject to approval by Congress) would have been able to refuse LLRW from outside their respective compact areas after January 1, 1986. Congress approved amendments to the Act shortly before adjournment in December, 1985, and the availability of disposal capacity for LLRW now seems assured for some time. However, the amendments to the Act specify timetables for unsited states to demonstrate good-faith efforts to provide disposal capacity for LLRW. The amendments also allow the sited states to limit the quantities of LLRW accepted for disposal and to levy surcharges on the accepted LLRW. Therefore, even though there is no longer an immediate general problem with the availability of LLRW disposal capacity, a state or state compact may find itself without adequate affordable disposal capacity, and 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.

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 are discussed in these introductory comments.

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.(1) In this document the NRC has considered two phases or time scales for extended 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 the uncertainties which still exist 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.

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, focusing on the waste form and container but also considering storage alternatives in order to establish the likely range of storage environments that the wastes would encounter. The dual objectives of this study are (1) to provide practical technical

*Work performed for the U.S. Nuclear Regulatory Commission, under the direction of Mr. Stephen A. Romano, Program Manager (FIN A-3171).

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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. BNL's preliminary findings were presented at this forum last year.[2] In this paper, the major points of BNL's final report to the NRC are summarized.[3]

Storage for Decay

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.[4]

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. 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. For example, estimates of the annual radiation exposure during storage operations have ranged from a high of 35.2 man-rem in a generic evaluation (for a 1000 MWe BWR) by the Atomic Industrial Forum[5] to a low of 4.1 man-rem for a site-specific evaluation (of two 1000 MWe BWR units).[6] These figures should be compared to occupational doses reported at U.S. commercial LWRs in 1981: 1400 and 2300 man-rem per 1000 MWe for BWRs and PWRs, respectively.[7]

CLASSIFICATION OF STORAGE FACILITIES

The various types of LLRW storage facilities, whether existing, under construction, or proposed, have been categorized in a survey of utility plans and actions which was conducted by the Electric Power Research Institute (EPRI)[8] and also in a New York State study of LLRW management practices.[9] The EPRI survey was published in July 1984 and contained information valid as of 1983. The construction status of on-site storage facilities as given in that survey is presented in Table I, from which it may be noted that the storage facilities are classified into three categories, viz., reinforced concrete structures, pre-fab structures (concrete or metal panels) and bunkers. In the New York State study, LLRW storage facilities are grouped into four categories, viz., shielded buildings, shielded storage modules, shielded casks, and unshielded facilities. 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-building-type structures. Each storage facility is in some ways unique, and for the purposes of the present study, a spectrum of storage concepts based on both of the above-mentioned classification schemes will be considered.

TABLE I

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

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

[a]Information from EPRI NP-3617, Reference 8.

The following spectrum of storage facility concepts ranges from shielded structures with temperature and humidity control through those with less environmental control to ones with minimal shielding, as well as minimal environmental control:

- Large engineered structures. These are permanent buildings designed specifically for the extended storage of LLRW. They may be reinforced concrete structures or steel frame buildings with uninsulated metal siding and roofing. They are generally provided with separate shielded areas for the storage of dry active waste and solidified wastes. Typically, some control over the temperature and, sometimes, the humidity is provided, e.g., a heating system to prevent freezing during the winter. Overhead bridge cranes are used for remote handling of the waste packages.

- Shielded storage modules or bunkers. These are permanent concrete structures with removable covers. Waste containers are emplaced or retrieved from above with a crane.

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

- Unshielded pre-fab structures. These are unshielded buildings which provide some degree of weather protection but have no temperature control system. Simple steel frame buildings with uninsulated metal siding and perhaps an overhead crane or hoist but no temperature control would fall into this category. These structures are generally intended for the

storage of low-specific-activity wastes. The waste packages are handled by means of hand dollies, fork-lift trucks, or cranes. These facilities have generally been used for storage for decay rather than extended storage.

- Minimal unshielded facilities. These are simple fenced-in outdoor concrete pads or very simple storage sheds. Little or no environmental protection is provided by these facilities, which were generally intended as holding areas for waste packages awaiting pick-up by a waste broker and not as waste storage facilities.

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. These variables were discussed in the earlier paper presented at WM '85[2] and will not be dealt with again here. The interested reader is referred to that paper and to BNL's final report on this task to the NRC.[3]

It should be noted, however, that 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 greater severity of certain storage environments, waste packages which would be expected to meet the 300-year disposal lifetime criteria may suffer severe performance 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.

PERFORMANCE OF THE LOW-LEVEL RADWASTE PACKAGE DURING STORAGE

In the presentation last year, an overview was given of the properties and behavior of LLRW streams, solidification agents, and container materials. The emphasis was 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 behavior of the waste package materials were also considered.

It must be emphasized that non-radiolytic effects are likely to be the primary concern for the majority of 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,[10] 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 have been estimated as a result of a survey carried out by BNL for the NRC.[11] 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. In this regard, it should be emphasized that the information on waste and waste package characteristics presented here last year[2] and in the final report to the NRC[3] 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.

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.[12] 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.

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. For references, the reader is referred to BNL's final report on this task to the NRC.[3]

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. From actual field data for the atmospheric corrosion of carbon steel containers, 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,[1] 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. For example, at Oak Ridge, a humid site, 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. 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.

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. 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. 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. Special steel alloys have been proposed as container materials for high integrity container designs recently submitted for approval. Further, protective coatings may be used 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.

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; 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. 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.

Radiolytic Gas Generation From Stored Ion-Exchange Resins and Bituminized 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. Radiolytic hydrogen gas production is expected from both bitumens and ion-exchange resins. For example, 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.) Radiolytic gas generation in ion-exchange resins may 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. 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.[13])

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. (The NRC has issued generic Letter 85-14 on use of nuclear reactor sites for the storage of wastes not generated by the utility licensee.) 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.

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 nonradiological 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.

Another problem which may apply to some institutional LLRW as well as to a small subset of fuel-cycle wastes is more of a regulatory issue than a technical issue. Some of these LLRW may be potentially hazardous wastes which, in principle, could be subject to regulation by the Environmental Protection Agency (EPA) as well as by the NRC. Storage of hazardous wastes is addressed in the EPA regulations in terms of the accumulation time for such wastes at the site of waste generation, e.g., in 40 CFR Section 262.34, where limits on the accumulation time are specified. At the time of this writing, the regulation of such mixed wastes remains an unresolved issue.

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The views expressed in this paper are not necessarily those of the U.S. Nuclear Regulatory Commission.

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