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SOME ASPECTS OF LOW-LEVEL RADIOACTIVE-WASTE DISPOSAL IN THE U.S.*

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ABSTRACT

This report summarizes the NRC supported Shallow Land Burial research program at Brookhaven National Laboratory and its relationship to the proposed revised ruling on disposal of low level radioactive waste, 10 CFR Part 61. Sections of the proposed regulation, which establish the new low level waste classification system and the performance objective placed on waste form, are described briefly. The report also summarizes the preliminary results obtained from the EPA program in which low level waste drums were retrieved from the Atlantic and Pacific Oceans.

SUMMARY OF THE BROOKHAVEN NATIONAL LABORATORY SHALLOW LAND BURIAL PROGRAM

The central objective of radioactive waste disposal is to isolate the waste because of its radiotoxicity from the public. This isolation period should be sufficiently long to allow for the reduction of radiotoxicity to a point where the waste poses no significant hazard to the public. In the United States (U.S.), low level nuclear wastes (LLW) are currently disposed of by shallow land burial (SLB). Shallow land burial of LLW at government installations was started in the 1940's. Disposal at commercial SLB sites, which are regulated under the authority of the U.S. Nuclear Regulatory Commission (NRC), was initiated in 1962. Since then, several small releases of radioactivity to the environment have occurred. The conditions that lead to these releases were a result of the disposal practices that had been implemented. Though none of the releases were judged as hazardous, they did focus attention on the ability of SLB to provide effective long-term isolation of radioactive waste and, hence, protect the public health and safety.

In 1976, Brookhaven National Laboratory (BNL) began a field and laboratory program, which is sponsored by the NRC. The program works in cooperation with the U.S. Geological Survey, to study existing commercial low level radioactive waste disposal sites. The BNL study has measured the physical properties and the concentrations of radioactive, organic and inorganic chemical components that are contained in water specimens obtained from actual LLW disposal sites. This program has also attempted to describe the major physical and chemical properties of the disposal site that control the migration of radionuclides.

*Work performed under the auspices of the U.S. Nuclear Regulatory Commission.

Early in the program, a broad overview study was performed to survey the water conditions that existed in the LLW disposal trenches and nearby water wells. Water samples were collected from 46 trenches and 5 wells at Maxey Flats, Kentucky; 13 trenches and 3 wells at West Valley, New York; 27 test wells at Sheffield, Illinois; and 6 trenches and 6 test wells at Barnwell, South Carolina. The remaining two commercial disposal sites at Beatty, Nevada and Hanford, Washington, are located in arid regions of the U.S. Therefore, the trenches at these sites are not expected to contain water and these sites are not addressed by the BNL study. The specific conductance, pH, and temperature of the water samples were measured in the field at the time of collection. Dissolved organic carbon (DOC), tritium, gross alpha, gross beta, and gamma radioactivity were measured in the laboratory (BNL).

More detailed radiochemical, inorganic, and organic analyses were performed on selected trench and well waters. The specimens selected for further study were chosen based on extreme and average values of the parameters measured in the survey study. The final selection of trenches for sampling depended upon the availability of water in the trenches, location of new monitoring wells at the disposal sites, and local groundwater flow paths.

A substantial body of data has resulted from these investigations.(1) Many of the conclusions drawn from the study are summarized below.

Trench waters are, in general, complex anoxic chemical systems which require extensive investigation to assess their role in radionculide retention and mobilization.

Water chemistry is often quite variable from trench to trench at a particular disposal site. For example, Table 1 shows the ranges of several properties measured on trench water specimens obtained from the Maxey Flats, Kentucky, disposal site. These variations indicate that the trench contents (the LLW that was emplaced) is variable (accurate records of trench content were not kept) and trench content dominates trench water chemistry. No overall systematic changes in any disposal site trenches were observed during the brief sampling interval (4 years). Although, minor changes in some radionuclide and cation concentrations were observed in several trenches. Tritium was the most abundant of the radionuclides and was found in all the trench waters. Chemically bound as HTO, tritium would be the most mobile radionuclide. ^{90}Sr, $^{238,239,240}Pu$ and ^{137}Cs were found as dissolved species in all trench waters and sorbed on suspended particulates at Maxey Flats and West Valley. Radionuclides measured as dissolved species in trench waters may be compared to the standard for release to an unrestricted area in 10 CFR Part 20. Tritium, ^{90}Sr, ^{137}Cs, and Pu in several trenches at Maxey Flats and West Valley exceeded the standard. Only tritium in two trenches at Barnwell, and ^{137}Cs and ^{90}Sr in one trench at Sheffield exceeded the standard.

Numerous organic compounds were identified in the trench waters at Maxey Flats and West Valley. The organic compounds found in the trench waters consisted of several straight- and branched-chain aliphatic acids, aromatic acids, alcohols, aldehydes, ketones, amines, aromatic hydrocarbons, esters, ethers, and phenols.

Table 1

Ranges of Measurements of Water Samples Taken From Maxey Flats, Kentucky, Disposal Site, Survey Study 1977

Measurement	Range
pH	2.2 - 12.4
Specific conductance (μmho/cm at 25°C)	280 - 2,900
DOC (mg/L)	<1 - 6,000
Gross alpha (nCi/L)	<0.2 - 640
Gross beta (nCi/L)	0.8 - 57,000
Gross gamma (relative cpm)	<10 - 16,000
Tritium (μCi/L)	0.25 - 7,400

Several microbiological experiments on the disposal trench waters have been performed in the BNL-SLB program. These studies have identified aerobic, anaerobic, sulfate reducing, nitrate reducing, and methanogenic bacteria to be present in the disposal trenches. Also, measurable amounts of tritiated and carbon-14 methane have been observed to be produced in actual trench water samples. The presence of active bacteria in the trench leachates suggests that biodegradation may play a significant role in the transformation of organic constituents of the waste. Biodegradation can affect the long-term stability of the wastes and affect the migration of radionuclides from the waste disposal sites.

The ability of disposal sites to retard the migration of radionculides has been addressed by performing radionuclide sorption studies. Experiments have been performed with site-specific soils under a broad range of conditions. The measured distribution coefficients (K_d) have been observed to generally depend upon soil composition, contact time, soil-to-solution ratio, redox conditions, and experimental technique (batch vs column). Under anoxic conditions, the K_d's are generally lower than in oxic conditions. The results also suggest slower kinetics under anoxic conditions. The results of column sorption measurements have been found different by as much as two orders of magnitude from those obtained in batch experiments. The results that have been obtained to date support the use of site-specific materials and experimental conditions which simulate in situ trench environments as closely as possible.

Soil cores, obtained from beneath four actual disposal trenches at the Barnwell disposal site, have been analyzed at BNL for radionculide content. Again, of all the radionuclides, tritium appears in the greatest concentration (10^5 to 10^9 pCi/L). Additionally, tritium was found in substantial concentrations over the entire 3 m length of the soil cores. Cobalt-60 was the next most abundant nuclide, although it was found in only one core. It was

detectable throughout that core, however. More than 90% of the ^{60}Co detected, was situated in the top 30 cm of the core. Gross alpha and beta activities significantly above background, and detectable concentrations of ^{90}Sr and $^{238,239,240}Pu$ were restricted to the uppermost 30 cm of the core.

In summary, several conclusions of this study are:

- Trench waters are, in general, complex anoxic chemical systems, containing many inorganic, organic, and radiochemical components.
- Substantial trench-to-trench compositional variations exist.
- In general, at sites (e.g., Maxey Flats and West Valley) where wastes are in constant contact with water, radionuclide concentrations are approximately one order of magnitude higher than at sites (e.g., Barnwell) where only periodic wetting occurs.
- Reliable measurement of solution-soil radionuclide distribution coefficients requires accurate experimental simulation of disposal site conditions (i.e., redox conditions and solution chemical composition).
- The migration of radionuclides away from the disposal trench has occurred. At the Maxey Flats disposal site fractured flow complicates prediction of future migration.
- Biodegradation of LLW may play a significant role in long-term isolation.

The BNL study and the efforts of others[2] to characterize existing LLW disposal sites have led to the following conclusions:

- Experience at existing LLW disposal sites indicates that safe short-term isolation of LLW has been accomplished by SLB.
- Trench instability is the major cause of the problems that have occurred at existing LLW disposal sites.
- Trench instability has allowed greater infiltration of precipitation which provides the vehicle for radionculide migration.
- In general, trench instability was caused by a combination of disposal site characteristics, disposal practices, and waste characteristics.

Based on our current understanding of the factors that contributed to the problems that have occurred with SLB, more stringent management of LLW is needed to assure long-term isolation of the waste. The NRC has proposed 10 CFR Part 61, "Licensing Requirement for Land Disposal of Radioactive Waste"[3] to accomplish this. The regulation seeks to assure long-term isolation of the waste by establishing performance objectives for (1) the waste

form, (2) the disposal site (including site selection, design, and operations), and (3) the institutional aspects of the disposal system. The next section of this report will briefly summarize the elements of 10 CFR Part 61 that should provide long-term stability of LL waste forms.

SUMMARY OF 10 CFR PART 61, THE PROPOSED REGULATION FOR LAND BURIAL OF LOW-LEVEL RADIOACTIVE WASTE

The U.S. Nuclear Regulatory Commission has had a program under way for several years to develop regulations and other guidance for the management and disposal of LLW. As a part of this program, the Commission announced its intention to:

- Develop technical criteria and standards for the disposal of LLW by shallow land burial and alternative disposal methods.
- Prepare a regulation and supporting Environmental Impact Statement (EIS).
- Coordinate development of technical criteria and standards for shallow land burial and alternative disposal methods with requirements for the classification of waste (define the concentrations and quantities of waste acceptable for disposal by various disposal methods).

As part of developing the regulation the Commission considered:

- The results of program studies and other technical data on LLW management and disposal.
- Licensing experience with current LLW disposal sites and current LLW management techniques.
- Programs by the Environmental Protection Agency (EPA) to develop criteria and standards for LLW management and regulations for disposal of nonradioactive solid and chemically hazardous wastes.
- Recommendations of the Interagency Review Group on Nuclear Waste Management.
- Natural Resources Defense Council (NRDC) Petition for Rulemaking.
- Discussions with industry and public interest groups, State and Federal agencies, and others.
- Recommendations from the State Planning Council.
- U.S. Public Law 96-573, "Low Level Radioactive Waste Policy Act."

It is the purpose of 10 CFR 61 to establish technical criteria and procedures for licensing facilities for the land disposal of radioactive wastes. The rule does not apply to alternative disposal methods such as deep space or ocean disposal. Requirements for ocean disposal are a responsibility of the EPA.

The recently enacted Low Level Radioactive Waste Policy Act sets forth a definition of "low level radioactive waste," i.e., radioactive waste not classified either as high level radioactive waste, transuranic waste, spent nuclear fuel, or uranium mill tailings. Part 61 (10 CFR 61) deals with the disposal of most wastes included in this definition. The waste classification scheme that forms the basis for Part 61 has identified some "low level radioactive wastes" that are not suitable for disposal by the means that Part 61 provides. The term "low level radioactive waste" is not used in Part 61. Reference is made to "waste" and "radioactive wastes" which, within the context of Part 61, refers to those wastes that are acceptable for disposal under the provisions of Part 61.

This proposed regulation includes overall performance objectives expected in any type of land disposal and technical requirements for the disposal of waste near the surface. The technical requirements for disposal are set forth for disposal site characteristics, disposal site design and near surface disposal facility operations, classification and characteristics of wastes, and institutional control and surveillance.

A waste classification scheme has been devised and incorporated into Part 61. The scheme is based on the role that the waste plays in the assurance that the performance objectives of protecting persons from radiation from waste will be met.

The first category identifies those wastes that do not have to meet the stability requirements and that will be segregated at the disposal site. These wastes, called Class A segregated wastes, are defined in § 61.55 in terms of the maximum allowable concentration of certain isotopes and certain minimum requirements on waste form that are necessary for safe handling. The second category is for waste that requires stability, Class B stable waste, and is defined in terms of allowable concentrations of isotopes and requirements for a stable waste form as well as the minimum handling requirements.

There are concentrations of certain isotopes that will require protection against inadvertent intrusion after institutional controls have lapsed. These concentrations have been determined by analysis of the exposure to humans from the postulated intrusion of an individual after the 100-year period of institutional control. Any waste with concentrations of these isotopes that would cause an exposure greater than 500 millirem must be protected from intrusion by deeper burial or some other barrier. Wastes requiring such protection are identified as Class C intruder wastes.

The waste classification section also places upper limits on concentrations of isotopes in any class of waste. Wastes containing higher concentrations are generally excluded from near surface disposal. Part 61 provides for special consideration by the Commission of proposed disposal methods on a case-by-case basis for wastes that exceed these values.

For most of the alpha emitting transuranic nuclides, the maximum allowable concentrations were calculated to be in the range of 10 nCi/g currently imposed by disposal facilities. A value of 350 nCi/g was established for plutonium-241, since this concentration of short lived beta emitting isotope decays to a 10 nCi/g concentration of americium-241, a longer lived alpha emitter. At present, wastes containing transuranic nuclides in concentrations greater than 10 nCi/g are not being generated in significant volumes in the U.S.

Wastes that would be exempt from Part 61 would be considered on a case-by-case basis. Such exemptions have been made for certain levels of tritium and carbon-14 in liquid scintillation and animal carcass waste.

In addition to 10 CFR Part 61 the NRC is preparing a Branch Technical Position which describes overall procedures acceptable to the Regulatory staff which may be used by licensees to determine the presence and concentrations of radionuclides listed in Table 1 of 10 CFR Part 61, which is reproduced here as Table 2, and thereby classifying waste for near surface disposal.

Each shipment of radioactive waste by a waste generator to a licensed collector, processor, or operator of a land disposal facility must ensure that a shipment manifest does accompany the waste. The manifest must include information on waste characteristics including the type of waste (e.g., solidified resins, trash), the waste volume and mass, the radionuclide identity and concentration, the total radioactivity, the chemical form*, the solidification agent (if any), and the waste class. As part of this, the total quantity of the particular radionuclides listed on Table 1 of § 61.55 must be listed**. These radionuclides are used to determine the classification status of radioactive waste.

To classify waste for disposal and fill out shipment manifests, a licensee must make two basic determinations: (1) whether the waste is acceptable for near surface disposal, and (2) if acceptable for near surface disposal, whether the waste is classified as Class A, Class B, or Class C waste.

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*This should be the principal chemical form that the radioactivity is contained in -- e.g., calcium fluoride, toluene, etc. As part of this, the presence of hazardous chemicals as defined by EPA should be noted.

**Radionuclides not specifically listed in Table 2 and forming a significant part of the total radioactivity should also be listed for purposes of waste handling and transportation safety. Radionuclides not specifically listed in Table 2 and not forming a significant part of the total radioactivity (particularly those having half-lives <5 years) need not be listed.

Table 2

Isotope	Column 1[a]	Column 2[b]	Column 3[c]
Any with half-life <5 years.....	700	70,000	Theoretical maximum specific activity
H-3.....	40	10[c]	Theoretical maximum specific activity[d]
C-14.....	0.8	0.8	0.8[d]
Ni-59.....	2.2	2.2	2.2
Co-60.....	700	70,000	Theoretical maximum specific activity
Ni-63.....	3.5	70	70
Nb-94.....	0.002	0.002	0.002
Sr-90.....	0.04	150	700
Tc-99.....	0.3	0.3	0.3[d]
I-129.....	0.008	0.008	0.008[d]
Cs-135.....	84	84	84
Cs-137.....	1.0	44	4600
Enriched uranium.....	0.04	0.04	0.04
Natural or depleted uranium.....	0.05	0.05	0.05
Alpha emitting transuranic isotopes..	-----	------	10 nCi/g
Pu-241.....	-----	------	350 nCi/g

[a]Maximum concentration for Class A segregated waste. Above this, it is Class B stable waste $\mu Ci/cm^3$.
[b]Concentrations above which some wastes become Class C intruder waste $\mu Ci/cm^3$.
[c]Maximum concentration for any waste class $\mu Ci/cm^3$.
[d]Near surface disposal facilities will be limited to a specified quantity for the disposal site. The quantity will be determined at the time the license is issued and will be governed largely by the characteristics of the site. Therefore, the total activity of these isotopes in each package of waste must be shown on the shipping manifest (see Section 20.311 of 10 CFR).

For isotopes contained in metals, metal alloys, or permanently fixed on metal as contamination, the values above may be increased by a factor of ten, except natural or depleted uranium which can be the natural specific activity.

For isotopes not listed above, use the values for Sr-90 for beta emitting isotopes with little or no gamma radiation; the values for Cs-137 for beta emitting isotopes with significant gamma radiation; and the values for U-235 for alpha emitting isotopes other than radium.

Wastes containing chelating agents in concentrations greater than 0.1% are not permitted except as specifically approved by the Commission.

For mixtures of the above isotopes, the sum of ratios of an isotope concentration in waste to the concentration in the above table shall not exceed one for any waste class.

Concentrations may be averaged over volume of the package. For a 55-gallon drum multiply the concentration levels by 200,000 to determine allowable total activity.

Until establishment and adoption of other values or criteria, the values in this table (or greater concentrations as may be approved by the Commission in particular cases) shall be used in categorizing waste for near surface disposal.

Waste is determined to be generally unacceptable for near surface disposal if it contains any of the radionuclides listed in Column 3 of Table 2 of Section 61.55 in concentrations exceeding the limits established for the radionuclides. If determined to be acceptable for near surface disposal, waste is determined to be Class A, Class B, or Class C based upon the list of radionuclide concentration limits set forth in Columns 1 and 2 of Table 2. (Note that the concentration limits in Table 2 may be subject to change.)

All licensees must carry out a compliance program to assure proper classification of waste. There are four basic programs which may be potentially used either individually or in combination by licensees:

- materials accountability
- classification by source
- gross radioactivity measurements, or
- direct measurement of individual radionuclides.

The Branch Technical Position will outline instances and conditions whereby each basic program or combinations thereof would be acceptable to Regulatory staff to demonstrate compliance with the waste classification requirement.

SUMMARY OF BNL-EPA OCEAN DUMPING PROGRAM

From 1946 through 1970 the United States disposed of low level radioactive waste materials either by shallow land burial at government owned sites, or by ocean dumping at Atomic Energy Commission (AEC) approved sites. Because the materials were regarded primarily as garbage, precise records were not kept of the disposal operations. Materials for land burial were packaged in a wide variety of containers, while materials for ocean disposal were encased primarily in concrete-filled steel drums.

Existing records of the ocean dumping activities consist primarily of licenses issued by the AEC contractors and of logs indicating the approximate locations of disposal sites. In most of the cases the records do indicate the nature of the materials, the quantities, the estimated radioactivity, and the approximate coordinates of the dumping location. They do not indicate the specific isotopic content. Neither the content of the containers nor the precise dumping locations can be verified.

The environmental survey work of the ocean nuclear waste dumpsites by EPA was initiated under the mandate of the Marine Protection, Research and Sanctuaries Act (PL 92-532). In 1974 EPA sent a survey team to the Farallon Islands dumpsites to confirm the feasibility of locating the containerized waste packages and to determine if any measurable amounts of radioactivity had been released into the dumpsites. Using an unmanned submersible vehicle several canisters were located. Underwater photographs were taken to assess the condition of the drums, and sediment samples were collected in proximity to both intact and imploded drums. Low levels of plutonium-238, -239, and -240 were detected.

In 1977, two additional surveys of the Farallon Island sites were undertaken; the first to provide estimates of biologic activity and diversity and to take samples of the water column and the ocean bottom, and the second to measure ocean currents and attempt the recovery of one of the waste canisters.

In the Atlantic Ocean a similar series of investigations was undertaken at the 2800 and 3800 meter sites. A preliminary sampling team went out to the 2800 meter site in May, 1974. In 1975 three dives in a submersible vehicle were made at the 2800 meter site to locate and document the condition of drums and to take a variety of sediment, water, and biological samples. In 1976 a waste canister was recovered from the 2800 meter site for detailed laboratory analysis of canister corrosion and physical integrity, and in 1978, a waste drum was recovered for analysis from the 3800 meter site.

The 80-gallon radioactive waste package which was retrieved from the Atlantic Ocean 2800 meter depth disposal site was transported to Brookhaven National Laboratory where container corrosion and matrix leach rate and degradation studies were conducted.

The following summary of the work and the conclusions resulting from the study were taken from the draft report, "Analysis and Evaluation of a Radioactive Waste Package Retrieved from the Atlantic 2800 Meter Disposal Site," P. Colombo, R. M. Nielson, Jr., and M. W. Kendig:

> "The retrieved waste package was an 80-gallon mild steel drum with no lid which contained a cement concrete matrix. Markings on the concrete surface indicated that it had been disposed in 1961. Within the concrete matrix, a sealed steel vessel was found which contained some liquid and 3 wound filter assemblies. Although this vessel had a major indentation running along its length resulting from the pressure differential during descent in disposal, it had not leaked."

Weight loss measurements, compressive strength measurements, and visual inspection indicated that the concrete matrix had not degraded appreciably. In summarizing the study, the authors noted a conservative estimate indicated that it would require a minimum of 100 years in this environment before the waste form would lose its integrity due to cement phase dissolution. Radiochemical analysis indicated the presence of cesium-137, cesium-134, and cobalt-60 in both the concrete matrix and the inner vessel. Based on the measured cesium-137 distribution in concrete cores, a bulk leach rate of 2.4×10^{-3} g/(cm^2-day) was calculated. This corresponds to an average fractional activity loss rate of 3.7×10^{-2} per year.

Corrosion rates for general attack on the upper portion of the 80-gallon mild steel drum were 0.0013-0.0019 in/yr (0.032-0.049 mm/yr) assuming a constant rate with no induction period. A lower limit for the rate of local pitting corrosion of 0.0026 in/yr (0.067 mm/yr) was determined. Using the range of general attack rates, an 18 gauge (nominal 0.0476 in thickness) mild steel drum would require 25-37 years in this disposal environment before integrity loss due to corrosion.

The major conclusions determined from this study as listed in the above draft report are:

- Little dissolution of the concrete waste form in the ocean environment occurred as evidenced by a maximum waste package weight loss of approximately 5%. Water loss through evaporation during curing of the cement and dissolution of calcium hydroxide in disposal environment are believed to be the primary mechanisms responsible for the apparent weight loss. A conservative estimate that assumes a constant 0.33%/yr weight loss due solely to cement phase dissolution predicts that it would require a minimum of 100 years in this environment before the concrete waste form would lose its integrity.

- The measured compressive strength of the concrete waste form is in the range expected for comparable concrete formulations. This indicates the absence of appreciable sulfate attack which is also supported by the observation that negligible surface deterioration of the waste form has occurred.

- The concrete waste form contained Cs-137, Cs-134, and Co-60. Based on the Cs-137 distribution in the waste form, a bulk leach rate for this radionuclide of 2.4×10^{-3} g/(cm^2-day) was calculated. This corresponds to an average fractional activity loss rate of 3.7×10^{-2} per year.

- While the inner container which enclosed 3 wound filter elements imploded due to the pressure differential during descent, water analysis indicated that the container did not leak and hence radionuclides were contained.

- Corrosion rates for general attack on the upper portion of the steel drum (assuming a constant rate with no induction period) were 0.0013-0.0019 in/yr (0.032-0.049 mm/yr). A lower limit for the rate of local pitting corrosion of 0.0026 in/yr (0.067 mm/yr) was determined. Using these rates of general attack, an 18 gauge (nominal 0.0476 in thickness) mild steel drum would require 25-37 years before integrity loss due to corrosion failure occurs.

- Variations in the corrosion attack between the upper and lower portions of the drum are ascribed to differences in surface finishes on the respective portions of the drum. While the coating on the lower portion of the drum successfully inhibited the initiation of general attack, instances of severe local attack leading to pitting and perforation adjacent to the drum chimes were observed.

- The waste container limits seawater exposure and movement through the waste form in disposal. As a result, the rate of activity loss through leaching and the rate of cement phase dissolution were decreased.

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