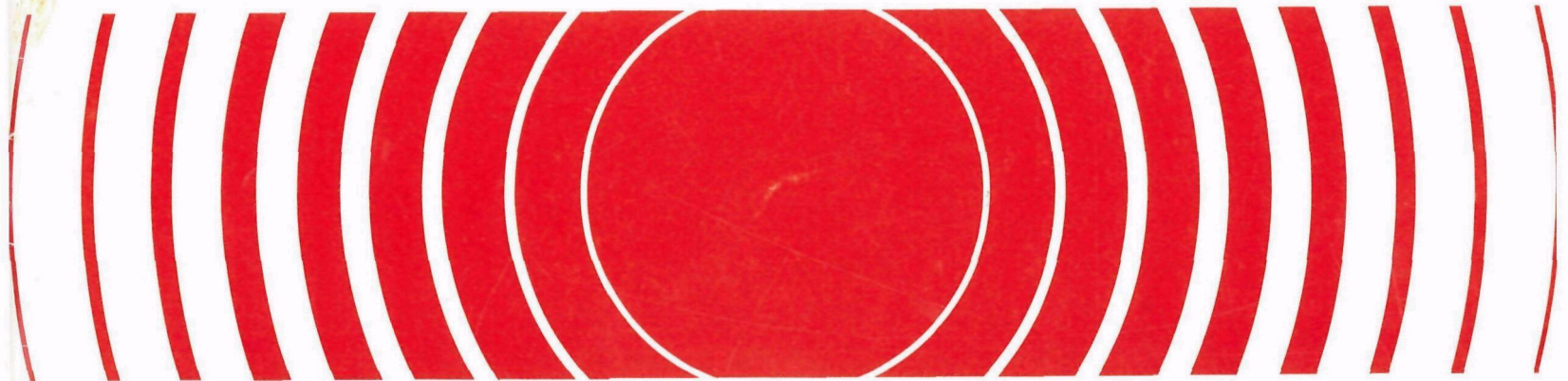




Radiation

# **Development of a Working Set of Waste Package Performance Criteria for the Deepsea Disposal of Low-Level Radioactive Waste**



DEVELOPMENT OF A WORKING SET OF WASTE PACKAGE PERFORMANCE CRITERIA  
FOR DEEPSEA DISPOSAL OF LOW-LEVEL RADIOACTIVE WASTE

BY

P. COLOMBO, M. FUHRMANN, R.M. NEILSON, JR., AND V.L. SAILOR

NUCLEAR WASTE RESEARCH GROUP  
DEPARTMENT OF NUCLEAR ENERGY  
BROOKHAVEN NATIONAL LABORATORY  
ASSOCIATED UNIVERSITIES, INC.  
UPTON, NY 11973

Prepared February 1982

Revised November 1982

This report was prepared as an account of work sponsored by  
the United States Environmental Protection Agency under  
Interagency Agreement No AD-89-F-1-558-0

PROJECT OFFICER  
ROBERT S. DYER  
ANALYSIS AND SUPPORT DIVISION  
OFFICE OF RADIATION PROGRAMS  
U.S. ENVIRONMENTAL PROTECTION AGENCY  
WASHINGTON, D.C. 20460

## DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency, contractor or subcontract thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency, contractor or subcontractor thereof.

# TABLE OF CONTENTS

	<u>Page</u>
FOREWORD.....	v
OBJECTIVE.....	1
1. INTRODUCTION.....	2
1.1 Background.....	2
1.2 Present Ocean Disposal Requirements.....	3
2. GLOSSARY.....	5
3. PRELIMINARY WASTE PACKAGE PERFORMANCE CRITERIA AND SPECIFICATIONS FOR OCEAN DISPOSAL OF LOW-LEVEL RADIOACTIVE WASTES.....	11
3.1 Assumptions.....	11
3.2 Waste Package Performance Criteria.....	12
3.3 Waste Container Performance Criteria.....	13
3.4 Waste Form Performance Criteria.....	14
3.5 Waste Content Criteria.....	17
REFERENCES.....	19
APPENDIX A: LOW-LEVEL RADIOACTIVE WASTE.....	A-1
APPENDIX B: RADIONUCLIDES OF IMPORTANCE.....	B-1
APPENDIX C: DOMESTIC AND INTERNATIONAL REGULATIONS WHICH POTENTIALLY IMPACT THE OCEAN DISPOSAL OF LOW-LEVEL WASTES.....	C-1

## FOREWORD

The Environmental Protection Agency (EPA) was given a Congressional mandate to develop criteria, standards, and regulations governing the ocean disposal of all forms of wastes pursuant to Public Law 92-532, the Marine Protection, Research and Sanctuaries Act of 1972, as amended. In response to this mandate, EPA has initiated a program to develop regulations and criteria to control the ocean disposal of radioactive wastes.

The EPA ocean dumping regulations and criteria were issued in the Federal Register on January 11, 1977. These regulations require that high-level radioactive wastes be prohibited from ocean dumping, and all other radioactive materials be contained to prevent their direct dispersion or dilution in ocean waters. Furthermore, these containerized radioactive wastes must radiodecay to environmentally innocuous levels within the life expectancy of the container(s) and/or the inert matrix.

The United States Congress has had a continuing interest in the question of ocean dumping of low-level waste and, in consequence, has recently approved Public Law 97-424. This Act amended PL 92-532 to include provisions to specifically consider the structural aspects of each container when evaluating any permit for the ocean disposal of radioactive waste.

For the past few years the EPA has been considering the question of suitable packaging of radioactive waste for sea disposal, both by evaluating the fate of radioactive waste packages dumped at formerly-used United States ocean dumpsites, and considering how past packaging designs might be improved. Although the EPA has not received a request for a permit to dispose of low-level radioactive waste in the ocean, it is incumbent on the Agency to develop the knowledge necessary to evaluate permit requests.

In order to determine whether any particular containment system or waste packaging system is adequate, it is necessary to establish a set of performance criteria or guidelines against which to evaluate a particular packaging system. These performance criteria must present requirements for the behavior of the waste in combination with its immobilization agent and outer container in a deepsea environment. It is expected that some of the criteria given in this report may be modified or amended, or new criteria added as more technical information becomes available.

The following report has been prepared by the Nuclear Waste Research Group of the Brookhaven National Laboratory and contains recommendations to the Environmental Protection Agency on packaging performance guidelines and criteria. As such, these recommendations have not been adopted by the Agency as necessary or sufficient to meet the Agency ocean dumping regulations.

The Environmental Protection Agency requests agencies and individuals to provide the Agency with any comments or suggestions pertinent to improving the document and the recommendations contained therein. Such comments or suggestions should be submitted to Mr. David E. Janes, Director, Analysis and Support Division, Office of Radiation Programs (ANR-461), Washington, D.C. 20460.

Glen L. Sjoblom, Director  
Office of Radiation Programs

## OBJECTIVE

The purpose of this document is to provide a working set of waste package performance criteria for consideration by the U.S. Environmental Protection Agency (EPA) pursuant to its responsibilities in accordance with the Marine Protection, Research and Sanctuaries Act of 1972 (PL 92-532). It is assumed that these criteria may be modified as more technical information becomes available.

The criteria and specifications are oriented toward the development of waste packages (waste form and container) which can provide continued isolation to assure the health and safety of the biosphere. These performance criteria, as proposed, rely on a combination of engineered and natural barriers. The development of these criteria is a first step toward evaluating packaging technologies and the subsequent need and extent to which waste packages must be developed or improved for ocean dumping.

## 1. INTRODUCTION

### 1.1 Background.

Sea disposal of low-level radioactive waste began in the United States in 1946, and was placed under the licensing authority of the Atomic Energy Commission (AEC). In 1962, the first commercial shallow-land disposal site was licensed in Beatty, Nevada. As land disposal operations expanded, ocean dumping was sharply reduced. The practice stopped completely in 1970 upon the recommendations of the Council on Environmental Quality in a Report to the President [1].

Most of the waste dumped at sea was packaged in second-hand or reconditioned fifty-five gallon drums filled with cement so that the average package density was sufficiently greater than sea water to assure sinking. It was assumed that all the contents would eventually be released since the packages were not designed or required to remain intact for sustained periods of time after descent to the ocean bottom [2].

In 1965, the Nuclear Energy Agency (NEA) of the Organization for Economic Cooperation and Development (OECD), in collaboration with a number of interested countries, undertook a series of studies of the practicability of joint sea dumping operations for low-level radioactive wastes. These studies led to the formulation of a number of conditions relating to the selection of suitable dumping areas, the design of waste containers and the selection of ships suitable for dumping operations. Procedures were also adopted for conducting and supervising these operations under satisfactory conditions and to prevent unacceptable radioactive contamination.

On the basis of the results of these studies, NEA sponsored the first international sea dumping operation for radioactive waste in 1967. Five OECD/NEA Member Countries (Belgium, France, the Federal Republic of Germany, the Netherlands, and the United Kingdom) participated in this first sea disposal operation under international supervision. During the

next internationally supervised sea disposal operation in 1969, Italy, Sweden and Switzerland also participated while the Federal Republic of Germany abstained. Since 1971, however, only Belgium, the Netherlands, Switzerland and the United Kingdom have used the sea disposal option. Recently, there has been renewed interest in ocean disposal, both in this country and abroad, as a waste management alternative to land burial. It is currently under review in several European countries and in Japan as a viable method for the disposal of other than high-level wastes.

Although the United States is not presently engaged in ocean dumping of radioactive waste, it has ratified and is a Contracting Party to the London Dumping Convention (an international agreement to control sources of marine pollution, including the dumping of radioactive materials, in international waters) [3]. Also, the United States is a Member Nation of the International Atomic Energy Agency (IAEA) and a Participating Country to the OECD Council Decision establishing a Multilateral Consultation and Surveillance Mechanism for Sea Dumping of Radioactive Wastes [4]. Thus, the United States shares responsibility with other nations for the consequences of ocean dumping of radioactive wastes.

The Marine Protection, Research and Sanctuaries Act of 1972 (PL 92-532) gives EPA the regulatory responsibility for ocean dumping of all materials, including radioactive waste. This act prohibits the ocean dumping of high-level radioactive waste and requires EPA to control the ocean dumping of all other radioactive waste through the issuance of permits. In implementing its permit authorities, EPA issued an initial set of regulations and criteria, in 1973, to control the dumping of material into ocean waters [5]. It was in these regulations that EPA initially introduced the general requirement of isolation and containment of radioactive waste as the basic operating philosophy.

## 1.2 Present Ocean Disposal Requirements.

In 1977, EPA issued final regulations and criteria concerning the dumping of radioactive wastes in the ocean [6].

In supporting the containment philosophy, the regulations require that "the radioactive materials must be contained to prevent their direct dispersion or dilution in ocean waters" and that "the materials to be disposed of must decay, decompose or radiodecay to environmentally innocuous materials within the life expectancy of the containers and/or their inert matrix."

The IAEA recommendations to the London Dumping Convention emphasize that a general policy of isolation and containment of radioactive waste be pursued through the use of suitable packaging to keep radioactive releases "as low as reasonably achievable" [7].

Both EPA and IAEA have identified the waste package as a prime barrier for the containment of radioactive waste. However, there appears to be a significant difference in the acceptable package performance between the two organizations. The terms "innocuous" and "as low as reasonably achievable" are not defined as used in the domestic and international guidance, respectively. In the context of the EPA criteria, "innocuous" might be interpreted as concentrations of radionuclides which have radiodecayed to such low concentrations that they do not present a hazard to man. On the other hand, "as low as reasonably achievable" refers to limiting radioactive releases, taking into account the state of packaging technology, economics of package improvements, public health and safety, and other societal and socioeconomic considerations. In the absence of more definitive restrictions, package release rates could be designed or varied to reflect the social and economic pressures of an ocean dumping nation.

An alternative concept is proposed in this report, which envisions the waste disposal system as a series of barriers necessary to ensure that radionuclides are retained or their movement is retarded to prevent a hazard to man. A "mutibarrier" concept is presented which consists of a "containment system" and an "isolation system." The containment system includes barriers to radionuclide movement provided by packaging, including the waste form and the container. The isolation system consists of the containment system as well as the natural environmental barriers against radionuclide transport from dumpsite to man.

## 2. GLOSSARY

Some of the terminology used in this report is defined to provide the reader with a quick reference to uncommon terms, or terms having unique meanings in the field of radioactive waste management.

Although it is acknowledged that the definitions may not be universally accepted, it is anticipated that the inclusion of this glossary will result in a better understanding of the proposed criteria in Section 3.

Acceptable Limit:	Radioactivity or radiation limit acceptable to a regulatory body.
Activation Product:	A radioactive isotope produced from a stable isotope by absorption of a neutron.
Activity:	A measure of the rate of radioactive decay occurring in a given quantity of material, i.e., the number of nuclear disintegrations per unit of time. Activity is commonly expressed in curies (Ci).
As Low As Reasonably Achievable (ALARA):	ALARA refers to limiting release and exposure and is used by the Nuclear Regulatory Commission (NRC) (10 CFR 50.34) in the context of "...as low as reasonably achievable, taking into account the state of technology and the economics of improvements in relation to benefits to the public health and safety and other societal and socioeconomic considerations...".
Alpha Activity (decay):	The disintegration of radioactive nuclei by emission of alpha particles. This decay process is invariably accompanied by gamma ray emission and usually occurs for very heavy isotopes, i.e., those of $Z > 82$ .
Barrier:	Any medium, engineered or natural, which prevents or retards the movement of radioactive materials.
Beta Activity (decay):	The radioactive decay of a nucleus by emission of a beta particle. Beta decay is often accompanied by the simultaneous emission of one or more gamma rays.

Biosphere:	That portion of the earth's environment inhabited by living organisms. It comprises parts of the atmosphere, the hydrosphere (ocean, inland waters and subterranean waters) and the lithosphere.
Cladding Waste:	Nuclear waste composed of cladding hulls and assembly grid spacers for nuclear fuel elements. Generated during reprocessing when spent fuel assemblies are disassembled.
Compressive Strength:	The load per unit of cross-sectional area under which a solid block fails by shear or splitting.
Container:	The receptacle into which a waste form is placed for disposal.
Container Lifetime:	The time period during which the container effectively serves as a barrier to radionuclide movement.
Containment:	The retention of radioactive material by the use of suitable packaging in such a way that it is effectively prevented from being dispersed into the environment.
Criterion:	A standard on which a decision or judgment may be based. It may be qualitative or quantitative, e.g., the package shall have adequate density to ensure sinking to the sea bed.
Curie (Ci):	A unit of activity equal to $3.7 \times 10^{10}$ disintegrations per second.
Decommissioning:	The preparation required for the planned permanent retirement of a nuclear facility in accordance with requirements set by a regulatory body.
Deepsea:	In context of ocean dumping of radioactive wastes, it is that part of the ocean where water depth is in excess of 4,000 meters.
Dispersion:	The summed effect of those processes of transport, diffusion, and mixing which tend to distribute materials from wastes or effluents through an increasing volume of water. The ultimate effect appears as a dilution of the material.
Disposal:	The disposition of waste materials without the intention of routine retrieval.

Dumping: (also Ocean Dumping)	The deliberate disposal of wastes into the ocean from vessels, aircraft, platforms or other man-made structures.
Environment:	The sum of all the conditions and influences that affect the survival and development of an organism.
Fissile Materials:	Isotopes (principally U-235, Pu-239 and U-233) which undergo fission following absorption of a low energy (thermal) neutron.
Fission:	The process whereby a heavy nucleus splits into two or more fragments with a concomitant release of energy.
Fission Product:	A stable or radioactive isotope formed by the fission of a heavy nucleus.
Fuel Cycle:	All the steps involved in supplying and using fuel materials for nuclear reactors, including waste management operations.
Fuel Reprocessing:	The dissolving of spent fuel elements for the removal of waste materials and the recovery and segregation of reusable materials.
Gamma ray:	Electromagnetic radiation (photon) emitted from the nucleus of the atom. Gamma rays and X-rays are identical in nature, except X-rays emanate from the electrons which surround the nucleus. In general, gamma rays are more energetic than X-rays.
Half-life:	The characteristic time in which half the atoms of a particular radioactive substance disintegrate. Each radionuclide has a unique half-life.
Hazard:	A natural or manmade cause of a potential deleterious effect, as differentiated from an expected deleterious effect.
High-Level Waste (HLW):	Those aqueous wastes resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated wastes from subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuel.
Immobilization:	Conversion of a waste to a form that reduces the potential for migration or dispersion of radionuclides by natural processes during storage, transportation and disposal.

Isolation:	The segregation of radionuclides from the biosphere by containment and the restriction of their release into that environment in unacceptable quantities or concentrations through the action of natural barriers.
Isotope:	Atoms of the same atomic number but with different atomic masses. For a given element the chemical properties of its various isotopes are almost identical; however, the nuclear properties of each isotope are distinctly different.
Leachability:	The susceptibility of a solid material to the removal of its soluble constituents by the dissolving or erosion action of water or other fluids.
Low-Level Waste (LLW):	Radioactive waste not classified as either high-level radioactive waste, transuranic waste, spent nuclear fuel or uranium mill tailings, as defined in the Low-Level Radioactive Waste Policy Act (PL 96-573).
Matrix Material:	A material used to solidify or immobilize radioactive waste by forming a monolithic solid, e.g., cement, bitumen, or polymer.
Migration (radionuclide):	The movement of radionuclides through various media due to dissolution fluid flow and/or by diffusion.
Maximum Permissible Concentration (MPC):	Maximum levels of radioactivity in drinking water or in air for the occupational worker, as established by national authorities, based on former ICRP recommendations. [Levels an order-of-magnitude lower were generally set for the public. However, new ICRP recommendations for limiting the intakes of radionuclides by workers (ICRP No. 30) no longer include the MPC concept for drinking water; instead, annual limits on intake (ALI) are used. MPC is not defined for seawater. However, it is customary to use the same MPC as for drinking water.]
Multibarrier:	A system using two or more independent barriers to isolate the waste from the human environment. These can include the waste form, the container, other engineered barriers and the disposal medium and its environment.
Nuclide:	A species of the nucleus of an atom characterized by its mass number, atomic number, and nuclear energy state.

Package:	The waste form and any container(s) as it is prepared for handling, transport, storage and disposal.
Pyrophoric Material:	Any material (solid or liquid) that ignites spontaneously in dry or moist air at or below 130° F.
Quality Assurance:	Planned and systematic actions necessary to provide adequate confidence that an item, facility or person will perform satisfactorily in service.
Radioactive Decay:	A spontaneous nuclear transformation in which alpha/beta particles or gamma radiation are emitted, or X-ray radiation is emitted following orbital electron capture, or the nucleus undergoes spontaneous fission.
Radioactive Waste: (Radwaste)	Any material or equipment that contains or is contaminated with radionuclides at concentrations or radioactivity levels established by the regulatory authorities and for which there is no anticipated use.
Radiolysis:	Chemical decomposition of a waste or waste form by the action of ionizing radiation.
Risk:	A measure of the deleterious effects that may be expected as a result of a technology, traditionally quantified as the product of the probability and the consequence of the occurrence of an event or series of events.
Secular Equilibrium:	A limiting case of radioactive equilibrium applying to a chain of two or more successive radioactive decays in which the decay rate of the parent isotope is equal to the decay rate of each of the daughter isotopes. Secular equilibrium can occur only in situations where the half-life of the parent is very long in comparison to that of each of the daughters and the parent and daughters are both confined to the same system for a time that is long compared to the half-life of the parent.
Site (dump):	The area containing nuclear waste that is defined by a boundary and which is under effective control of the implementing organization.

Solidification:	Conversion of liquid radioactive waste to a dry, stable monolithic solid.
Solidification Agent:	See Matrix Material.
Specification:	A numerical value, or range of values, indicating quantitatively whether a criterion has been met.
Spent Fuel:	Nuclear fuel which has been discharged from a reactor after having been subjected to nuclear reactions. (Fuel is usually discharged because it has been consumed to the design limit or because of failure or for necessary reactor maintenance.)
Transuranic (TRU) Waste:	Wastes containing quantities of nuclides, having atomic numbers > 92, at concentration levels established by the regulatory authorities. Waste containing more than 10 nanocuries ( $10^{-8}$ Ci) of transuranic alpha activity per gram of waste has been defined as TRU waste by DOE (Public Law 96-573).
Waste Form:	A monolithic free standing solid resulting from the incorporation of waste into a matrix material (e.g., liquid in concrete, solids in bitumen).
Waste Management:	The planning and execution of essential functions relating to radioactive wastes, including treatment, packaging, interim storage, transportation, and disposal.
X-ray:	A penetrating form of electromagnetic radiation emitted either when the inner orbital electrons of an excited atom return to their normal state (characteristic X-rays), or when a metal target is bombarded with high-speed electrons. X-rays are always nonnuclear in origin (i.e., they originate external to the nucleus of the atom).

### 3. PRELIMINARY WASTE PACKAGE PERFORMANCE CRITERIA AND SPECIFICATIONS FOR OCEAN DISPOSAL OF LOW-LEVEL WASTES

The basic criteria and specifications suggested in this section are directed specifically towards those conditions which impact the performance of the waste package during storage, transportation, handling and disposal. Therefore, it precludes criteria associated with other aspects of ocean disposal such as site selection, operations, and monitoring.

Specific performance criteria are suggested for the waste package and for the individual waste package components which include the waste container, the waste form and the waste type. Where possible, numerical specifications are listed to qualify criteria; however, many specifications cannot be determined fully at this time since the information needed to assume reasonable values is not readily available. It is expected that these values will be identified as more definitive criteria are developed. In the meanwhile, the phrase, "to be determined," is used to indicate these information gaps.

#### 3.1 Assumptions.

It was necessary to make a set of assumptions to enable the development of waste package performance criteria. These assumptions are:

- Existing Federal regulations govern the interim storage, transportation and disposal of radioactive wastes. Waste packages intended for ocean disposal should meet all minimum Federal requirements, including relevant United States international treaty commitments.

- Only low-level waste (LLW), as defined in the Low-Level Radioactive Waste Policy Act (PL 96-573), is considered for ocean disposal.

- The disposal site is located in waters at a depth in excess of 4,000 meters.

- The waste package is not intended to be routinely retrievable.

- Package performance specifications are based upon a multiple barrier concept considering the contributions of engineered barriers (waste form, container) and natural barriers (water column, sediment geochemistry, etc.).

### 3.2 Waste Package Performance Criteria.

- Criterion: The package shall have adequate density to ensure sinking to the sea bed.  
  
Specification: The specific gravity of the solidified waste form and container shall not be less than 1.2 to ensure sinking to the sea bed.  
  
Discussion: A waste package must be sufficiently dense to sink immediately. Although the specific gravity of surface sea water does not exceed 1.03, the package should be sufficiently dense to ensure that its movements during descent and on the sea bed are not readily influenced by currents.
- Criterion: The waste package shall remain intact upon impact on the ocean floor.  
  
Specification: To be determined. (The waste package shall be designed to meet or exceed National and IAEA Transport Regulations and to maintain its integrity upon impact on the ocean floor at a calculated and/or measured terminal velocity).  
  
Discussion: There should be no loss or dispersal of radionuclides upon impact of the waste package with the ocean surface during the ship dumping operations and upon impact of the package with the ocean floor. Tests conducted in the United Kingdom and in Japan for packages meeting IAEA Transport Regulations have shown that the impact of a package free-falling from a ship height of 30-50 feet (9-15 m) to the ocean surface has not resulted in package failure. The velocity of the package upon impact with the ocean floor has been calculated to be approximately 6-7 ft/sec (2.2-2.3 m/sec), depending on package density and configuration. [8,9].

- Criterion: The waste package shall be designed with adequate strength to maintain its integrity during the normal hazards of transportation, handling and disposal.
- Specification: To be determined. (All waste packages shall meet the minimum DOT Regulations 49 CFR 173).
- Discussion: Waste packages must be capable of withstanding the stresses encountered in handling, storage and transportation. Consideration should be given to the fact that the distance required to lift waste packages for emplacement aboard a ship may be significantly greater than that required in shallow-land disposal operations.
- Criterion: Waste packages shall be designed for safe handling and be compatible with handling equipment.
- Specification: To be determined.
- Discussion: Lifting rings and other auxiliary handling devices are required to facilitate package handling. Lifting devices should be offset or hinged in a manner which does not inhibit or compromise the integrity of the waste package.

### 3.3 Waste Container Performance Criteria.

- Criterion: The container shall be capable of maintaining its contents until the radionuclides have decayed to acceptable limits.
- Specifications: The waste container shall have an expected lifetime of 200 years in the deepsea environment.
- Discussion: The expected lifetime of the container is contingent on the types and amounts of radioactive materials in the waste form and the characteristics of the disposal site. In assuming isolation as the basic operating philosophy for the disposal of radioactive wastes in the ocean, both engineered and natural barriers contribute to controlling the release of radioactivity such that the amounts released would not constitute a significant hazard to man. This implies that the life expectancy of the container can be less than the time required for the radioactive materials to decay to environmentally acceptable

limits, where acceptable limits are those quantities of activity which, when the other barriers to migration are considered, will not pose a significant hazard to man. A life expectancy of 200 years is presumed adequate for the container, since the longest lived radionuclides of importance, Cs-137 and Sr-90, will have decayed to less than 1% of their initial activity in this time. (Depending upon the types of activity contained and their quantity, some containers may not require a lifetime as long as 200 years.)

- Criterion: Waste containers shall be of uniform size, configuration and construction.
- Specification: To be determined.
- Discussion: An important purpose for this criterion is to prevent exceeding the size limitations of handling equipment and facilities. However, several benefits also accrue from the use of standard containers. These benefits include: optimization of storage facilities, standardization of handling and transportation techniques, improved quality control and quality assurance, decreased packaging costs and reduction in personnel exposure. Specifications may include several container designs.

### 3.4 Waste Form Performance Criteria:

- Criterion: Liquid radioactive wastes shall be immobilized by suitable solidification agents.
- Specification: Aqueous wastes should be solidified to form a homogeneous, monolithic, free standing solid containing no more than 0.5 percent (by volume), or 1.0 gallon (3.8 liters) of free or unbound water per container, whichever is less.
- Discussion: Wet wastes such as ion exchange resins, sludges and evaporator concentrates may contain as much as 80% to 90% water by volume. This liquid must be immobilized by a solidification agent to form a monolithic matrix to prevent ready dispersion of activity and potential container corrosion from within.

- Criterion: Buoyant waste material shall be excluded or treated to preclude its movement or separation from the waste form during and after disposal.  
  
Specification: Buoyant material must be shredded, incinerated or processed prior to solidification to form a homogeneous free standing monolithic solid having a specific gravity not less than 1.2.  
  
Discussion: Buoyant material included in the waste must be treated to preclude its return to surface waters in the event that the waste package deteriorates or loses its structural integrity during descent to the sea bed and thereafter.
  
- Criterion: The waste form shall be able to withstand the hydrostatic pressure encountered during and after descent to the sea bed.  
  
Specification: The waste form shall have a uniaxial compressive strength not less than  $150 \text{ kg/cm}^2$ , provided that it does not contain large voids or compressible materials.  
  
Discussion: In the event that large voids cannot be eliminated, it will be necessary to incorporate approved pressure equalization devices to assure that the waste package will maintain its integrity during and after descent to the sea bed. Measures should be taken to ensure that any voids in the waste form and/or waste package are small and homogeneously distributed so as to resist the hydrostatic pressure encountered during descent. Work in Japan has shown that waste forms with uniaxial compressive strengths of  $150 \text{ kg/cm}^2$  will maintain their integrity under triaxial hydrostatic pressure of  $500 \text{ kg/cm}^2$  (corresponding to a depth of 5,000 meters) [10].
  
- Criterion: Particulate wastes such as ashes, powders and other similar materials shall be immobilized by a suitable solidification agent.  
  
Specification: Particulate wastes shall be immobilized to form a homogeneous, monolithic, free standing solid.

- Discussion: Particulate wastes are readily dispersible in air and water. Immobilization reduces the quantity of potentially respirable fines during handling and the dispersibility of radioactive material in the event of container failure in disposal.
- Criterion: The waste form shall be chemically compatible with the container material.
- Specification: To be determined.
- Discussion: The chemical composition of the wastes and/or solidification agent(s) should be such that it does not adversely affect the expected life-time of the container. The presence of corrosive liquids and/or the formation of gases and other materials through radiolysis or bacterial action can compromise the integrity of the waste form and the container.
- Criterion: The leach rate of the waste form shall be as low as reasonably achievable (ALARA).
- Specification: The fractional release rate for Cs-137<sub>3</sub> Sr-90 and Co-60 shall be no greater than  $10^{-3}$  Fraction Release  $\times (V/S)d^{-1}$  where V=volume and S=surface area of the waste form, in accordance with the proposed ANS 16.1 leach test for leaching in seawater [11,12].
- Discussion: Mitigation of hazard to man depends upon the performance of multiple barriers preventing radionuclide migration. One of these barriers is the waste form itself. Performance of the waste form as a barrier is often described in terms of a leach rate. A leach rate of  $10^{-3}$  Fraction Release  $\times (V/S)d^{-1}$ , or less for a 55-gallon drum size cement waste form is approximately equivalent to an activity release of 0.04% per year after container failure at 200 years (based on the initial activity content of a waste form containing only Cs-137 and Sr-90). Actual release rates by leaching will probably be lower in a disposal site since at container failure only a limited surface area will be subject to leaching and since the temperature is approximately 1-4°C. In addition, leach rates typically decrease as the valence of the radionuclide species increases (e.g., Cs > Sr > Co).

### 3.5 Waste Content Criteria

- Criterion: Compressed radioactive gaseous wastes shall be excluded from disposal at LLW ocean disposal sites.

Specification: No radioactive gaseous wastes shall be accepted for ocean disposal unless they have been immobilized into stable waste forms such that the over-burden pressure in the waste package does not exceed atmospheric pressure.

Discussion: Compressed gases, by their nature, present a hazard to operating personnel during shipboard handling and disposal operations. In addition, the possibility of failure resulting from hydrostatic pressures encountered during descent could result in an instantaneous release of radioactivity. The disposal of gaseous wastes such as Kr-85 and H-3, will require immobilization by methods such as ion implantation on metal surfaces, sorption on various substrates or reaction with transition metals to form metal hydrides.

- Criterion: Gas generation by all mechanisms shall not be sufficient to adversely affect the integrity of the waste package during storage, transportation and disposal.

Specification: To be determined.

Discussion: The rate at which gas is generated by all mechanisms during storage, transportation and disposal must not be sufficient to cause package failure as a result of pressurization. Such pressurization is of primary concern during storage and transportation but in ocean disposal at depths in excess of 4,000 meters, internal pressurization is compensated by high hydrostatic pressures.

- Criterion: Wastes in which the primary hazard is associated with chemical toxicity shall not be dumped in a LLW ocean disposal site.

Specification: To be determined.

- Discussion: Disposal sites will be selected and designed for the isolation of low-level radioactive wastes. Therefore, chemical toxic wastes may not be adequately isolated by such sites and may, in fact, compromise site performance for radionuclide retention.
- Criterion: Pyrophoric or other highly reactive materials shall be excluded from LLW ocean disposal sites.
- Specification: No pyrophoric materials shall be accepted for LLW ocean disposal unless they have been solidified with chemically stable materials such that the waste form is rendered non-pyrophoric.
- Discussion: Waste forms must be non-pyrophoric in order to minimize the possibility of shipboard fires during transportation and handling and the potential for chemical reactions which may compromise the integrity of the waste package during and after disposal.
- Criterion: Explosive materials shall be excluded from LLW ocean disposal.
- Specification: No explosive materials shall be accepted for LLW ocean disposal unless they have been solidified with chemically stable materials such that the waste form is rendered non-explosive.
- Discussion: Waste forms must be non-explosive in order to minimize the probability of package failure and radioactivity dispersal as the result of explosion during transportation and handling during and after disposal.
- Criterion: The waste shall be physically and chemically compatible with the solidification agent.
- Specification: To be determined.
- Discussion: Certain "problem" wastes, such as organic liquids, ion exchange resins and oils are difficult to solidify by one or more of the contemporary solidification processes as they are currently used. The properties of such waste forms may preclude them from ocean disposal.

## REFERENCES

1. Council on Environmental Quality, Ocean Dumping a National Policy, A Report to the President prepared by the Council on Environmental Quality, U.S. Government Printing Office, Washington, D.C., October 1970.
2. National Academy of Sciences, Disposal of Low-Level Radioactive Waste into Pacific Coastal Waters, National Academy of Sciences - National Research Council Publication No. 985, Washington, D.C., 1962.
3. "Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter," drawn up at the Intergovernmental Conference on the Dumping of Wastes at Sea, held in London, October 30-November 10, 1972.
4. Organization for Economic Cooperation and Development, Decision of the Council Establishing a Multilateral Consultation and Surveillance Mechanism for Sea Dumping of Radioactive Wastes, OECD, C(77)115 (Final), Paris (1977).
5. Environmental Protection Agency, "Transportation for Dumping of Material Into Ocean Waters," Chapter I - Environmental Protection Agency, Subchapter H-Ocean Dumping, Federal Register, 38 (198): pp. 28610-28621, Monday, October 15, 1973.
6. Environmental Protection Agency, "Final Revision of Regulations and Criteria," Chapter I - Environmental Protection Agency, Subchapter H-Ocean Dumping, Federal Register, 42(7): pp. 2462-2490, Tuesday, January 11, 1977.
7. International Atomic Energy Agency, "Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, - The Definition Required by Annex I, paragraph 6 to the Convention, and the Recommendations Required by Annex II, Section D," INFCIRC/205/Add.1/Rev.1, IAEA, Vienna, Austria, August 1978.
8. Mitchell, N.T., "The Principles and Practice of Design and Manufacture of Packages for Sea Disposal of Radioactive Waste from U.K." A paper distributed to the Committee Meeting, Ministry of Agriculture and Food, Directorate of Fisheries Research, Fisheries Radiobiological Laboratory, Lowestoft, Suffolk (1979).
9. Olivier, J.P., Sea Disposal Practices for Packaged Radioactive Wastes Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle, Denver, USA, 1976, Conf.-76-0701 (ABTS) U.S. Dept. of Commerce, Springfield, VA (1976) p. 667.
10. Seki, S. and H. Amano, "Integrity Test of Full Size Packages of Cement-Solidification Radioactive Wastes Under Deep-Sea Conditions," Nuclear And Chemical Waste Management, Vol. 1, pp. 129-138, Pergamon Press Ltd., 1980.

REFERENCES (CONT.)

11. American Nuclear Society, Measurement of the Leachability of Solidified Low-Level Radioactive Wastes, Second Draft of a Standard, American Nuclear Society Standards Committee Working Group ANS-16.1, April, 1981.
12. International Standards Organization, Long-Term Leach Testing of Radioactive Waste Solidification Products, Draft ISO Standard, ISO/TC 85/SC 5/WG 5 N 38., 1979.

## Appendix A. LOW-LEVEL RADIOACTIVE WASTE

In order to understand the concerns associated with the ocean disposal of low-level radioactive waste and to develop waste package performance criteria and specifications, it is useful to consider the kinds of wastes which comprise low-level radioactive wastes. This chapter will provide information concerning the sources and types of low-level wastes, including their physical form and chemical nature as well as current and projected generation rates.

### Definitions.

Low-Level radioactive waste (LLW) is defined by the Low-Level Radioactive Waste Policy Act (Public Law 96-573) as "radioactive waste not classified as high-level radioactive waste, transuranic waste, spent nuclear fuel or by-product material as defined in Section 11e.(2) of the Atomic Energy Act of 1954." The Act defines by-product material as "the tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore reprocessed primarily for its source material content."

High-level radioactive waste (HLW) is defined in the Marine Protection, Research and Sanctuaries Act of 1972 (Public Law 92-532) and EPA Ocean Dumping Regulations (40 CFR 227) as "the aqueous waste resulting from the operation of the first cycle solvent extraction system, or equivalent, and the concentrated waste from subsequent extraction cycles, or equivalent, in a facility for reprocessing irradiated reactor fuels or irradiated fuel from nuclear power reactors." This law and regulations also prohibits the ocean dumping of high-level wastes.

The Atomic Energy Commission (now the Department of Energy) defined transuranic waste as material excluding high-level waste which contains more than 10 nanocuries per gram of transuranic nuclides, with the exception of Pu-238 and Pu-241, but including U-233 and its daughter products (AEC Manual, Chapter 0511, 1973). The AEC Manual also mentions that the

value of 10 nCi/g is subject to modification based on long-term studies of nuclide migration in soil. On September 14, 1974, the AEC proposed the 10 nCi/g value in the Federal Register. This definition does not explicitly apply to commercially generated (NRC licensed) wastes. Recently, the NRC has proposed a 100 nCi/g limit (10 CFR 61) for land disposal of radioactive wastes.

#### Sources and Types of Low-Level Waste.

Low-level waste (LLW) is produced as a consequence of both federal government and commercial operations.

The majority of federal LLW is a consequence of defense related activities, including fuel fabrication, reactor operation, spent fuel storage, fuel reprocessing and associated chemical processing operations. (Although fuel reprocessing is primarily associated with the generation of high-level waste, significant quantities of low-level waste are also produced). In addition, the federal government also generates LLW during facility decontamination and decommission activities and from research and development activities.

Commercial generation of LLW results from both fuel cycle and non-fuel cycle operations. Commercial fuel cycle operations include uranium mining, uranium milling, uranium hexafluoride ( $UF_6$ ) production, uranium enrichment, fuel fabrication, reactor operations, spent fuel storage and facility decontamination and decommissioning. (There is currently no fuel reprocessing conducted by the commercial sector, although this may resume in the future.) These commercial fuel cycle activities are similar in nature to federal government activities and hence, most of the waste types produced are analogous. Non-fuel cycle operations, both institutional (including medical institutions and universities) and industrial (pharmaceutical and other industries) also produce LLW.

The types of LLW produced by the various sources are summarized in Table A.1. These wastes may be classified as either dry wastes or wet wastes.

TABLE A.1 Sources and Types of Low-Level Radioactive Wastes [1].

SOURCES	TYPES OF WASTE GENERATED												
	DRY WASTES				WET WASTES								
	COMBUSTIBLE		NONCOMBUSTIBLE		FILTER CAR- TRIDGES	SPENT RESINS	SLURRIES AND SLUDGES	AQUEOUS CONCEN- TRATES	SPECIAL AQUEOUS SOLUTIONS <sup>a</sup>	OILS	OTHER ORGANIC LIQUIDS	MEMBRANES <sup>b</sup>	BIOLOGICAL
	COMPAC- TIBLE	NONCOM- PACTIBLE	COMPAC- TIBLE	NONCOM- PACTIBLE									
GOVERNMENT DEFENSE	•	•	•	•	•	•	•	•	•	•	•	•	
D & D <sup>c</sup>	•	•	•	•		•			•				
RD & D <sup>d</sup>	•	•	•	•		•	•	•	•	•	•	•	•
COMMERCIAL FUEL CYCLE MINING	•	•	•	•			•	•					
MILLING	•	•	•	•			•	•					
UF <sub>6</sub> PRODUCTION	•	•	•	•			•						
ENRICHMENT	•	•	•	•			•			•	•		
FUEL FABRICATION	•	•	•	•			•			•			
POWER PLANTS	•	•	•	•	•	•	•	•	•	•		•	
SPENT FUEL STORAGE	•	•	•	•		•	•						
D & D	•	•	•	•		•			•				
NONFUEL CYCLE MEDICAL	•		•	•							•		•
PHARMACEUTICAL <sup>e</sup>	•			•							•		•
UNIVERSITIES	•		•	•		•					•		•
OTHER INDUSTRIES <sup>e</sup>	•			•		•							

a Decontamination, pickling, etching, electropolishing, etc. solutions.

b Membranes from processes such as ultrafiltration (UF) and reverse osmosis (RO).

c Decontamination and decommissioning (D &amp; D) operations.

d Research, development, and demonstration (RD &amp; D) programs.

e Data on these wastes are incomplete and difficult to obtain.

Dry wastes are solids and include items such as paper, glass, metal, wood, plastic, rubber and rags. Dry wastes may further be broken down into two classes: combustible or non-combustible and compactible or non-compactible. Combustible dry wastes may be incinerated to reduce volume. The resultant incinerator ash residue is highly dispersible and may require solidification prior to disposal.

The majority of wet wastes are produced from the cleanup of aqueous processes or waste streams prior to recycle or discharge. The type of waste resulting from these cleanup operations depends upon the process employed (filtration, ion exchange, evaporation, centrifugation, reverse osmosis, ultrafiltration, flocculation or sedimentation). Filtration produces filter cartridge and filter sludge wastes. Spent resin, powdered resin sludges, and regenerant solution wastes result from ion exchange operations. Evaporation, centrifugation, reverse osmosis, ultrafiltration, flocculation and sedimentation processes generate slurry, sludge and aqueous concentrate wastes. Reverse osmosis and ultrafiltration also produce membrane wastes. In addition, wastes resulting from these cleanup operations are often subjected to additional treatment to reduce their volume for disposal. For spent resins and sludges this may include a dewatering operation (settling, centrifugation, or filtration), while volume reduction of aqueous wastes is generally accomplished through some form of evaporation. Some wet wastes are combustible (resins, oils and organic liquids) and as such, an incineration process could be employed to reduce waste volume. More detailed descriptions of these wastes and their origins can be found in the literature [1-4].

Generally, wet wastes are solidified prior to disposal. Exceptions include filter cartridges, some spent ion exchange resins and filter sludges, and organic liquids. While past practice has been to simply dewater resins prior to disposal, governmental agencies and shallow land disposal site operators are requiring solidification of some spent resin wastes. Solidification agents commonly employed in the United States include portland cements and modified portland cements. Urea-formaldehyde is no longer used. Thermosetting resins and asphalt are beginning to be used for the solidification of LLW in this country. Sorbents, such

as vermiculite and synthetic calcium silicates have been used with wastes to immobilize liquids. While this practice has been greatly reduced at commercial fuel cycle facilities due to more restrictive attitudes of regulatory agencies, their use is still fairly widespread at some government installations and at commercial non-fuel cycle facilities.

Package performance specifications may require that low-level wastes to be disposed of by ocean dumping be monolithic free standing solids. As such, most wastes would require solidification; however, some solidified low-level waste forms may not meet performance specifications. This situation then requires either modification of current solidification formulations, selection of a more appropriate solidification agent (with use of a suitable formulation) or segregation of these wastes and subsequent disposal by another method. In addition, all other wastes must be solidified using proper formulations to assure that the resultant waste forms meet the designated performance specifications for ocean disposal.

#### Generation Rates of Low-Level Wastes.

The estimated annual volumetric generation rates of low-level wastes in the United States for 1980 are shown in Table A.2 [5]. The majority of this waste (52% by volume) originates from commercial sources. LLW produced by the federal government is not further broken down in Table A.2. This information is generally not available, since much of it relates to defense wastes. Some 58% of commercial LLW is generated by fuel cycle operations. Power reactor operations produce 87% of the volume of fuel cycle LLW. Non-fuel cycle wastes constitute 42% of commercial LLW volume. The quantity of LLW produced by institutional (hospitals, medical schools, colleges and universities) and industrial sources are estimated to be approximately equal [5].

Considering the mix of power reactor types in 1980 and their respective liquid processing systems (deep bed resins or precoat filters for boiling water reactors (BWRs) and condensate polishing systems (CPS) or no CPS for pressurized water reactors (PWRs) [2,6], about 48% of power reactor wastes are dry solids. The remaining waste fraction consists

TABLE A.2

Amounts of LLW Added to Burial Grounds in the United States in 1980 [5]

Source	Generation Rate $\text{m}^3/\text{yr}$	Percentage Total
GOVERNMENT	$7.4 \times 10^4$	44.4
OTHER GOVERNMENT OR UNCLASSIFIED <sup>(a)</sup>	$6.5 \times 10^3$	3.9
COMMERCIAL (Fuel Cycle + Non-Fuel Cycle)	$8.6 \times 10^4$	<u>51.7</u>
		100.0
<u>Fuel Cycle</u> (58% Commercial LLW)		
UF <sub>6</sub> Production	$1.6 \times 10^3$	3.2
Enrichment <sup>(b)</sup>	$2.0 \times 10^2$	0.4
Fuel Fabrication	$4.7 \times 10^3$	9.5
Reactor Operations	<u><math>4.3 \times 10^4</math></u>	<u>86.9</u>
Total Fuel Cycle	$5.0 \times 10^4$	100.0
<u>Non-Fuel Cycle</u> (42% Commercial LLW)		
Institutional <sup>(c)</sup>	$1.8 \times 10^4$	50.0
Industrial	<u><math>1.8 \times 10^4</math></u>	<u>50.0</u>
Total Non-Fuel Cycle	$3.6 \times 10^4$	100.0

(a) For example, waste from fuel fabrication for foreign reactors, or waste generated by government agencies, but shipped for commercial burial.

(b) Included in government waste total given above.

(c) Institutional sources include hospitals, medical schools, colleges and universities.

primarily of wet wastes which have been solidified prior to disposal. Dry solids constitute approximately 42% of institutional wastes [3] and as much as 90% of the volume of the LLW generated by some industrial producers [4]. Most of these dry wastes contain relatively small quantities of radionuclides. A significant fraction of this waste is "suspect" and may in fact not be contaminated.

Estimates of LLW volumetric generation rates through the year 2000 are listed by source in Table A.3 [5]. Projections of the volume of fuel cycle LLW are based upon a proposed reference growth scenario projecting 180 GW(e) of installed nuclear capacity by the year 2000. While the generation rate of governmental LLW is expected to remain approximately constant over the period from 1980 to 2000, the volume of commercial LLW produced annually is projected to increase more than 150% (These projections do not consider governmental LLW resulting from the decontamination and decommissioning of formerly utilized or surplused sites under the Formerly Utilized Sites Remedial Action Program (FUSRAP) or the Surplus Facilities Management Program (SFMP). (They also do not consider LLW resulting from a possible resumption of commercial spent fuel reprocessing.) Most of this increase is due to increased quantities of fuel cycle waste resulting from the expansion of installed power reactor capacity. The volume of fuel cycle LLW is anticipated to increase by 200% between 1980 and 2000, while non-fuel cycle wastes over this period may remain similar to that shown in Table A.2 for 1980; however, reactor operations in particular have significant incentive to reduce waste volume.

While volumetric LLW generation rates as presented in this section give a perspective of the magnitude of the problem, it is difficult to assess the hazards and risks implicit in ocean disposal without information concerning the radionuclide inventory. This information is presented in Appendix B for individual radionuclides in low-level wastes.

TABLE A.3

Projected Low-Level Waste Generation Rates, 1980-2000 [5]

<u>Year</u>	<u>Government, m<sup>3</sup>/yr</u>	<u>Commercial, m<sup>3</sup>/yr</u>		
	<u>Total</u>	<u>Fuel Cycle</u>	<u>Non-Fuel Cycle</u>	<u>Total</u>
1980	$5.7 \times 10^4$	$5.0 \times 10^4$	$3.6 \times 10^4$	$8.6 \times 10^4$
1985	$6.1 \times 10^4$	$8.5 \times 10^4$	$4.9 \times 10^4$	$1.3 \times 10^5$
1990	$6.1 \times 10^4$	$1.2 \times 10^5$	$5.9 \times 10^4$	$1.8 \times 10^5$
1995	$6.1 \times 10^4$	$1.3 \times 10^5$	$6.8 \times 10^4$	$2.0 \times 10^5$
2000	$6.1 \times 10^4$	$1.5 \times 10^5$	$7.9 \times 10^4$	$2.3 \times 10^5$

## APPENDIX A REFERENCES

1. Kibbey, A.H. and H.W. Godbee, A State-of-the-Art-Report on Low-Level Radioactive Waste Treatment, ORNL/TM-7427, Oak Ridge National Laboratory, Oak Ridge, Tennessee, 1980,.
2. Phillips, J. , F. Feizollahi, R. Martineit, W. Bell and R.A. Stouky, Waste Inventory Report for Reactor and Fuel Fabrication Facility Wastes, ONWI-20/NUS-3314, NUS Corporation, Rockville, Maryland, 1979.
3. Beck, T.J., L.R. Cooley and M.R. McCampbell, Institutional Radioactive Wastes-1977, NUREG/CR-1137, University of Maryland, Baltimore, Maryland, 1979.
4. General Research Corporation, Study of Chemical Toxicity of Low-Level Wastes, NUREG/CR-1973, General Research Corporation, Santa Barbara, California, 1980.
5. Oak Ridge National Laboratory, Spent Fuel and Radioactive Waste Inventories and Projections as of December 31, 1980, DOE/NE-0017, Oak Ridge National Laboratory, Oak Ridge, Tennessee, 1980.
6. Mullarkey, T.B., T.L. Jentz, J.M. Connelly and J.P. Kane, A Survey and Evaluation of Handling and Disposing of Solid Low-Level Nuclear Fuel Cycle Wastes, AIF/NESP-008, NUS Corporation, Rockville, Maryland, 1976.

The purpose of this section is to identify the principal radioactive isotopes found in low-level radioactive waste which might be considered for deepsea disposal. The isotopes normally present in LLW are discussed in terms of their longevity, toxicity and relative abundance.

### Introduction.

The IAEA, acting in a technical advisory capacity to the London Dumping Convention has analyzed the radiological impacts of ocean disposal of radioactive materials [1-4]. The analyses take account of some 80 radionuclides [4]. For each isotope, conservative estimates of population dose commitments were made using 12 pathways from disposal site to man. For the "sake of administrative convenience and analytical simplicity," the results for the individual isotopes were subsequently combined into three broad categories (see Table B.1). Annual release rate limits (curies per year) were assigned to each category, which were based on ICRP dose limits for critical groups or individual members of the public. The annual limits refer to total disposal by all countries, which implies that individual countries will eventually be assigned a quota not to be exceeded [1].

The IAEA analyses did not take credit for the retention of radionuclides in the disposal packages. In essence it was assumed that as soon as the package reached the sea bed, all radioactive contents would be discharged into the water. This approach yields a conservative (pessimistic) radiation dose estimate, particularly for the dose contribution of isotopes having half-lives in the range of a few years or less.

In the following sections, the disposal question will be examined from a more detailed point of view which will allow an evaluation of the benefits that might accrue from waste packages ("engineered barriers")

which effectively immobilize the radioactive contents for varying periods of time. Such engineered barriers would add an additional margin of safety to the already conservative IAEA recommended limits.

TABLE B.1

Release Rate Limits Recommended by IAEA  
for Ocean Disposal of Radionuclides [1]

Group	Release Rate Limits (Ci/year)	
	<u>Single-site</u>	<u>Finite Ocean Volume</u> <sup>a)</sup> ( $10^{17} \text{ m}^3$ )
Alpha emitters, but limited to $10^4$ Ci/yr for Ra-226 and supported Po-210	$10^5$	$10^5$
Beta/gamma emitters with half-lives of at least 0.5 years (excluding tritium) and beta/alpha emitters of unknown half-lives	$10^7$	$10^8$
Tritium and beta/gamma emitters with half-lives less than 0.5 years	$10^{11}$	$10^{12}$

a) The "Finite Ocean Volume" takes into account uniform mixing in the total ocean basin of all isotopes of half-lives less than or equal to 40,000 years, from all disposal sites.

#### Radionuclides Commonly Present in LLW.

More than 2,000 radioactive isotopes exist in nature or have been produced by nuclear reactions. The vast majority of these are not of concern in waste disposal because their half-lives are much shorter than the time required to collect and process the waste. After a decay time equal to 6.645 half-lives, only 1% of the original activity of a given radioactive isotope remains; after 10 half-lives less than one part in a thousand ( $10^{-3}$ ) remains, and after 20 half-lives less than one part in a million ( $10^{-6}$ ) of the original activity remains. Thus, isotopes having half-lives  $< 0.1$  years will have decayed to  $< 0.1\%$  of their original activity after one year of aging and to  $< 0.0001\%$  after two years.

Most of the isotopes appearing in LLW are those which are products of nuclear fission or neutron activation of stable elements contained in reactor cores (coolant, fuel hardware and cladding, reactor core structures, control rods, etc.). A few additional isotopes manufactured by charged particle accelerators appear in industrial and institutional LLW, e.g., Na-22, Cl-36, Cd-109.

The distribution of radionuclides appearing in various low-level waste streams of reactors is quite diverse, depending on the reactor design, degree of fuel burnup and the details of the waste processing equipment. In general, the wastes have been poorly characterized. Reliable assays have been limited. Consequently, large inconsistencies appear in published data. Recently, R. E. Wild et al. [5] have completed a comprehensive analysis of radwaste data which appears to be particularly thorough. Their report has served as the primary resource for the numerical tabulations given in this chapter. Some additional data have been taken from other sources, as cited in the tables' references.

Tables B.2, B.3 and B.4 list the radionuclides commonly found in reactor waste streams and in industrial/institutional wastes. Isotopes with half-lives  $< 0.5$  years have been omitted from the tables, since they will have decayed to negligible concentrations within the first five years after packaging, and therefore do not contribute significantly to the long-term radioactive inventory in the disposal environment. Isotopes with half-lives in the range of 0.5 to 5 years have been included because they must be considered when handling the wastes during disposal operations.

Table B.5 compares the 1980 United States LLW inventory with the IAEA release rate limits listed in Table B.1. If the United States is to use ocean disposal for LLW at an annual rate equal to the 1980 inventory, only a small fraction of the IAEA release rate limits would be approached. Therefore, it can be concluded that the IAEA release rate limits will not be a major restrictive factor if the United States should decide to resume ocean disposal of LLW on a limited scale.

TABLE B.2

Radioactive Isotopes Commonly Found in Wastes from  
Commercial Nuclear Power Plants

Only those isotopes with half-lives greater than 0.5 years have been included. The annual quantities listed in column 3 are the estimated activities which will eventually be appearing in wastes as a consequence of operations in the year 1980. The "relative hazard index" (RHI) is defined as the quantity of water (in liters) required to dilute 1  $\mu\text{Ci}$  of the isotope to MPC listed in 10 CFR 20, Appendix B, Table II, Column 2.

ISOTOPE <sup>a)</sup>	HALF-LIFE <sup>b)</sup> (years)	ANNUAL <sup>a)</sup> QUANTITIES (1980) (Ci/year)	RELATIVE HAZARD INDEX (RHI) AFTER VARIOUS PERIODS OF AGING <sup>c)</sup> (liters/initial $\mu\text{Ci}$ )			
			0 yrs	10 yrs	150 yrs	600 yrs
H-3	12.33	90	0.33	0.19	neg. <sup>d)</sup>	neg.
C-14	5730	12	1.3	1.3	1.28	1.21
Fe-55	2.7	$1.3 \times 10^5$	1.3	0.1	neg.	neg.
Ni-59	$7.5 \times 10^4$	120	5	5	5	5
Co-60	5.27	$1.7 \times 10^5$	20	5.4	neg.	neg.
Ni-63	100.1	$1.2 \times 10^4$	33	30.8	11.7	0.5
Nb-94	$2.03 \times 10^4$	2.7	(2.5) <sup>e)</sup>	(2.5)	(2.5)	(2.4)
Sr-90	28.8	80	3333	2620	90	$1.8 \times 10^{-3}$
Tc-99	$2.14 \times 10^5$	0.21	3.3	3.3	3.3	3.3
I-129	$1.56 \times 10^7$	0.58	17000	17000	17000	17000
Cs-135	$2.95 \times 10^6$	0.21	10	10	10	10
Cs-137	30.17	$5.6 \times 10^3$	50	39.7	1.6	neg.
U-235	$7.04 \times 10^8$	0.14	33	33	33	33
U-238	$4.47 \times 10^9$	1.1	25	25	25	25
Np-237	$2.14 \times 10^6$	$2.2 \times 10^{-5}$	333	333	333	333
Pu-238	87.71	$2.2 \times 10^3$	200	185	61	1.7
Pu-239	$2.41 \times 10^4$	$3.0 \times 10^3$	200	200	200	197
Pu-240	$6.57 \times 10^3$		200	200	197	188
Pu-241	14.38	$5.9 \times 10^4$	5	3.1	$4 \times 10^{-3}$	neg.
Pu-242	$3.76 \times 10^5$	6.5	33	33	33	33
Am-241	432	9.8	250	246	197	95
Am-243	$7.37 \times 10^3$	0.67	250	250	246	236
Cm-243	28.5	0.58	200	157	5.2	neg.
Cm-244	18.1	6.7	143	98	0.5	neg.

a) Significant isotopes were identified and quantities estimated from data given Ref. 5 and based on 28.34 GWe-yr for 1980.

b) Ref. 6.

c) The RHI entries are for 1  $\mu\text{Ci}$  at 0 years. Values shown for subsequent years take the radioactive decay into account.

d) neg. = negligible, less than  $10^{-3}$ .

e) Estimated value. Nb-94 is not listed in 10 CFR 20, Appendix B.

Table B.3

## Radioactive Isotopes Commonly Found in Institutional and Industrial Wastes

Only those isotopes with half-lives greater than 0.5 years have been included. The "relative Hazard index" (RHI) is defined as the quantity of water (in liters) required to dilute 1  $\mu$ Ci of the isotope to MPC listed in 10 CFR 20, Appendix B, Table II, Column 2.

<u>ISOTOPE</u> <sup>a)</sup>	<u>HALF-LIFE</u> <sup>b)</sup> (years)	ANNUAL <sup>a)</sup> QUANTITIES (1980) (Ci/year)	RELATIVE HAZARD INDEX (RHI) AFTER VARIOUS PERIODS OF DECAY <sup>c)</sup> (liters/initial $\mu$ Ci)			
			<u>0 yrs</u>	<u>10 yrs</u>	<u>150 yrs</u>	<u>600 yrs</u>
H-3	12.33	544	0.33	0.19	neg. <sup>d)</sup>	neg.
C-14	5730	31	1.3	1.3	1.28	1.21
Na-22	2.6	3	25	1.7	neg.	neg.
Cl-36	$3 \times 10^5$	$7 \times 10^{-2}$	12.5	12.5	12.5	12.5
Co-57	0.74	7	2	neg.	neg.	neg.
Co-60	5.27	55	20	5.4	neg.	neg.
Ni-63	100.1	1.1	33	30.8	11.7	0.5
Zn-65	0.67	0.3	10	neg.	neg.	neg.
Sr-90	28.8	19	3333	2620	90	$1.8 \times 10^{-3}$
Tc-99	$2.14 \times 10^5$	$2.1 \times 10^{-5}$	3.3	3.3	3.3	3.3
Cd-109	1.24	0.3	5	neg.	neg.	neg.
Cs-137	30.17	27.8	50	39.7	1.6	neg.
Gd-153	0.66	4.3	5	neg.	neg.	neg.

a) Significant isotopes were identified and quantities estimated from data given in Ref. 5 and 7.

b) Ref. 6.

c) The RHI entries are for 1  $\mu$ Ci at 0 years. Values shown for subsequent years take the radioactive decay into account.

d) Neg. = negligible, less than  $10^{-3}$ .

Table B.4

## Radioactive Isotopes Commonly Found in Special Industrial Wastes

Only those isotopes with half-lives greater than 0.5 years have been included. These wastes arise from commercial isotope production (radiopharmaceuticals) tritium production, tritium accelerator targets, sealed radiography sources, and miscellaneous high activity wastes from neutron activation. The RHI's for these isotopes are listed in Table B.2

<u>ISOTOPE</u> <sup>a)</sup>	<u>HALF-LIFE</u> <sup>b)</sup> (years)	ANNUAL <sup>a)</sup> QUANTITIES (1980) (Ci/year)	<u>ISOTOPE</u> <sup>a)</sup>	<u>HALF-LIFE</u> <sup>b)</sup> (years)	ANNUAL <sup>a)</sup> QUANTITY (1980) (Ci/year)
H-3	12.33	$2.39 \times 10^5$	U-235	$7.04 \times 10^8$	$1.51 \times 10^{-3}$
C-14	5730	3.06	U-238	$4.47 \times 10^9$	$5.64 \times 10^{-3}$
Fe-55	2.7	$8.56 \times 10^3$	Np-237	$2.14 \times 10^6$	$7.89 \times 10^{-11}$
Ni-59	$7.5 \times 10^4$	4.88	Pu-238	87.71	$2.92 \times 10^{-2}$
Co-60	5.27	$1.55 \times 10^4$	Pu-239	$2.41 \times 10^4$	$8.21 \times 10^{-3}$
Ni-63	100.1	$2.01 \times 10$	Pu-240	$6.57 \times 10^3$	
Nb-94	$2.03 \times 10^4$	$3.33 \times 10^{-2}$	Pu-241	14.38	1.05
Sr-90	28.8	$7.02 \times 10^3$	Pu-242	$3.76 \times 10^5$	$1.42 \times 10^{-5}$
Tc-99	$2.14 \times 10^5$	$4.84 \times 10^{-2}$	Am-241	432	$3.05 \times 10^3$
I-129	$1.56 \times 10^7$	$4.03 \times 10^{-4}$	Am-243	$7.37 \times 10^3$	$1.85 \times 10^{-4}$
Cs-135	$2.95 \times 10^6$	$4.84 \times 10^{-2}$	Cm-243	28.5	$2.44 \times 10^{-2}$
Cs-137	30.17	$7.39 \times 10^3$	Cm-244	18.1	$4.26 \times 10^{-5}$

a) Significant isotopes were identified and quantities estimated from data given in Ref. 5.

b) Ref. 6.

TABLE B.5

Comparison of the 1980 United States LLW Inventory  
with the IAEA Criteria Shown in Table B.1

If the U.S. used ocean disposal at the rate of 1980 generation of wastes, the IAEA guidelines would not be exceeded. The U.S. 1980 data are the summation of the Column 3 entries in Tables B.2, B.3 and B.4.

<u>Group</u>	<u>Release Rate Limit (Ci/yr) IAEA Single-Site</u>	<u>1980 U.S. LLW Inventory<sup>a)</sup> (Ci/yr)</u>	<u>Fraction US/IAEA</u>
Alpha <sub>4</sub> emitters, but limited to $10^4$ Ci/yr for Ra-226 and supported Po-210	$10^5$	$5.2 \times 10^3$	0.052
Beta/gamma emitters with half-lives of at least 0.5 years (excluding tritium)	$10^7$	$3.6 \times 10^5$	0.036
Tritium and beta/gamma emitters with half-lives less than 0.5 years	$10^{11}$	$3.1 \times 10^{5b)}$	$3.1 \times 10^{-6}$

a) Allowance for defense LLW would double the numbers in this column.

b) The entry includes  $2.4 \times 10^5$  Ci/yr of H-3 listed in Tables B.2, B.3 and B.4, plus an additional  $7 \times 10^4$  Ci/yr of beta/gamma emitters with half-lives less than 0.5 years which have been omitted from the tables.

Toxicity of Radionuclides.

The radioactivity of a sample, measured in units of curies, is not an accurate reflection of its toxicity. (Of course, for a given isotope, 2 curies is twice as toxic as 1 curie.) The toxicity of a radionuclide depends also on the type and energy of the radiation it emits (alpha particles, beta particles, gamma rays, x-rays) and on the biochemical behavior of the chemical compounds of the isotope. The latter determines the routes by which an isotope can enter a living organism, which organs will be affected, and the average residence time in each of the organs.

The toxicity of individual isotopes has been the continuing subject of investigation over the past 54 years by the International Commission on Radiological Protection (ICRP), the National Committee on Radiation Protection (NCRP), and various other national and international bodies. The recommendations of these organizations have been adopted with a slight modification by the U.S. Nuclear Regulatory Commission (NRC). The maximum permissible concentration (MPC) for each isotope in air and water is listed in 10 CFR 20, Appendix B [8]. The inverse of the MPC values can be regarded as a measure of the toxicity of each isotope.

#### The Relative Hazard Index.

In order to express the toxicity in a form which can be more easily visualized, the data in Appendix B of 10 CFR 20 can be inverted to give ml/ $\mu$  Ci and then converted to liters/ $\mu$  Ci. This gives the quantity of water (in liters) required to dilute 1  $\mu$  Ci of a given isotope to acceptable levels for human consumption.\* This quantity will be referred to as the "relative hazard index", where  $RHI = 1/MPC \times 10^{-3}$ . The values, listed in Column 4 of Tables B.2 and B.3, were calculated from the data [8] using the entries for the soluble form of the isotopes for unrestricted areas, since these entries reflect the most conservative scenario for ocean disposal.

In a waste package, each original curie (Ci) of a given isotope will decay according to the characteristic half-life of the isotope. Thus, the RHI of an initial 1  $\mu$  Ci will decrease with time. Columns 5-7 of Tables B.2 and B.3 list the decreasing values of RHI arbitrarily selected for 10, 150 and 600 years of decay.

The relative toxicity,  $T_i$ , of isotope  $i$  in the LLW inventory at any future time can be obtained from the equation:

---

\*It is recognized that seawater is not actually an ingestion pathway, but the MPC is used only to provide an approximation of the relative hazard of various radionuclides.

$$T_i \text{ (future year)} = \text{RHI (future year)} \times (\mu\text{Ci})_i,$$

where  $(\mu\text{Ci})_i$  is the original inventory of isotope  $i$ .

#### Quantities of Radionuclides in LLW.

The relative abundance of radioactive isotopes appearing in particular LLW waste streams depends on the source of the waste and its age. Most waste consists of the fission products of U-236 (U-235 + neutron) and Pu-240 (Pu-239 + neutron). In the fission event, the parent isotope can split in many ways, yielding a very large combination of stable and/or radioactive products. The most probable events yield two fission products, usually one light and one heavy isotope.

The absolute fission yields as a function of mass number are shown in Figure B.1 for U-235 plus a neutron. The two prominent peaks correspond to the light and heavy products. The yields of the isotopes between and on each side of the peaks are extremely small (note that the vertical axis of the figure is a logarithmic scale). The corresponding curve for Pu-239 plus a neutron is similar, except the low peak is shifted to the right by about 4 mass numbers.

From the data in Figure B.1 it can be seen that LLW which has a fission product origin will be composed mostly of isotopes in the mass range from 85 to 110 and from 125 to 155. Column 3 of Table B.2 lists the quantities of each isotope which are estimated to appear in the LLW at commercial nuclear power plants in the year 1980. The relatively large quantities of Sr-90 and Cs-137 are accounted for by the fission yield curve. Most of the inventory of fission products remains trapped in the spent fuel and does not enter the LLW streams. (If recycling of spent fuel is resumed, the LLW streams from reprocessing plants will substantially increase the quantities of fission products for LLW disposal, even though most of the inventory will go into high-level waste streams.)

The yield of radioactive isotopes produced by neutron activation depends on the materials present in the reactor core and its immediate surroundings. Such materials include the coolant, fuel assembly hardware, control materials and the reactor structural materials. Column 3 of Table B.2 includes the estimated inventory of activation products (H-3 through Co-60) produced from commercial power plants in the year 1980.

Industrial and institutional LLW originates from many sources, including reactors and charged particle accelerators, and thus does not show typical fission product distribution. Quantities of industrial and institutional LLW have been estimated for the year 1980 and are listed in Column 3 of Table B.3.

Table B.4 lists estimated quantities of special industrial wastes resulting from isotope production, accelerator targets, and sources for radiography.

The inventories shown in Tables B.2-B.4 omit several important sources of waste, e.g., DOE production and research reactors and the nuclear navy, since such information is not in the public domain. However, as a rough estimate it can be assumed that the inventories will have a distribution similar to that shown in Table B.2, and that the listed inventories would about double.

#### Identification of Most Important Isotopes.

The data in Tables B.2, B.3 and B.4 make it possible to identify the most important isotopes from the viewpoint of disposal. These are the isotopes which have long half-lives, large RHI, and which are produced in large quantities.

The isotopes fall into two major groups: 1) those with half-lives of 30 years or less, and 2) those with half-lives greater than 30 years. The rationale for this division is that practical engineered barriers can be designed to immobilize the isotopes in Group 1 for the duration of

their significant toxicity. For the isotopes in Group 2, factors other than engineered barriers, i.e., natural barriers, must be relied upon for long-term radiation protection.

Group 1: Isotopes with Half-Lives of 30 Years or Less.

Except in unusual circumstances, e.g., cases in which chemical separations have been performed on wastes prior to disposal, a relatively few isotopes will dominate the waste packages and will determine the performance specifications envelope. Packages which can immobilize these problem isotopes can adequately retain the less important isotopes.

Tables B.2, B.3 and B.4 show quite clearly that three isotopes are of dominating importance to disposal considerations for typical waste streams. These are: Sr-90, Cs-137 and Co-60. A disposal package which will adequately contain these three isotopes would also contain the other short-lived (< 30 years) isotopes listed in Table B.2.

The design of the waste containers and/or the choice of solidification agents must adequately immobilize those elements which, because of their chemical properties, normally display high mobility, e.g., cesium.

A reasonable design objective for the average life expectancy of an engineered waste container might be in the range of 200 years, at which time approximately 1% of the initial Cs-137 inventory (half-life = 30 years) remains. After 200 years, the package would slowly deteriorate, releasing the remaining (declining) Cs-137 inventory.

Past experience indicates that a 200-year design objective is feasible at reasonable costs. Included in this experience are: 1) the condition of archaeological materials salvaged from the oceans after prolonged periods of immersion; 2) the condition of concrete pilings and piers, and 3) the condition of waste drums recovered from the ocean [9].

## Group 2: Isotopes with Half-Lives Longer than 30 Years.

It is prudent to assume that after several hundred years economically practical waste packages on the ocean floor will gradually deteriorate and the residual radioactivity will be discharged into the surrounding environment. The consequences of the ultimate releases must be examined in geochemical terms. These include:

- concentrations of identical or similar radionuclides already present in ocean waters from natural sources;
- abundance of stable isotopes of the element relative to the inventory of radioisotope released;
- volume of water required to dilute inventory to MPC\*, and
- chemical and geochemical behavior of the radionuclides in the disposal environment.

In the following sections, groups of the long-lived isotopes are discussed in the geochemical context.

### Long-Lived Isotopes Which Decay by Alpha Emission.

A. Natural Decay Chains: Three natural radioactive decay chains occur in nature. These originate, respectively, from Th-232 (designated the  $[4n]$  decay series), U-238 ( $[4n + 2]$  decay series), and U-235 ( $[4n + 3]$  decay series)\*\*. Each of the radioactive decay chains undergoes a series of alpha and beta decays, finally terminating in a stable isotope of lead or bismuth.

---

\*As noted earlier, the maximum permissible concentration (MPC) for drinking water does not apply to seawater, which is not directly ingested by humans; however, the MPC provides a highly conservative basis for comparing the relative toxicity of the various radionuclides.

\*\*A fourth natural decay series  $[4n + 1]$ , originating with Np-237, existed on earth for the first 50 million years of its history, but like the dinosaur is long since extinct. However, unlike the dinosaur it has been revived by man, originating from Pu-241.

If the parent isotope and all its daughters are isolated and retained together for periods of time which are long compared with the longest half-life in the chain, the parents and daughters approach the condition of secular equilibrium. In this situation, the decay rate of each daughter product is equal to the decay rate of the parent. For example, 1 Ci of U-238 in secular equilibrium has an additional 17 Ci of associated daughter products, of which a total of 8 Ci are alpha emitters.

B. Natural Alpha Decay in Ocean Waters: The global oceans contain approximately  $5 \times 10^9$  tonnes (metric) of U-238,  $3.6 \times 10^7$  tonnes of U-235, and  $7.8 \times 10^7$  tonnes of Th-232 [10]. Much larger quantities of each are present in ocean sediments. The daughter products of each of these isotopes are also present, both in the water and in the sediments.

In the water, secular equilibrium does not exist. There are several reasons for the departure from secular equilibrium:

- Daughter products formed in terrestrial rocks are in a more mobile chemical state than the parent, and hence leach from the rocks at accelerated rates. For example, the successive decays,  $\text{U-238} \rightarrow \text{Th-234} \rightarrow \text{Pa-234} \rightarrow \text{U-234}$  result in a higher leach rate for U-234 than for U-238. Hence, ocean waters contain about 15% more U-234 than would be the case for secular equilibrium.
- The isotopes of thorium are subject to rapid sedimentation. The residence time of thorium in seawater is about 200 years. Thus, many of the daughter products are formed in the sediments rather than in the water.
- Ra-226, a daughter of U-238 (via Th-230), formed in the sediments, has a long enough half-life and is sufficiently mobile to reemerge from the sediments into the water.

- o Concentrations of Ra-226 are highest in the bottom water. The average concentration is about 10% of that expected from secular equilibrium.

The net result is that the concentration of alpha activity in ocean water is somewhat less than would be the case if secular equilibrium existed. For example, for U-238 and its daughter products the total alpha activity is approximately 2.6 Ci per Ci of U-238, rather than 8.0 Ci for the case of secular equilibrium. The total alpha activity accompanying the  $5 \times 10^9$  tonnes of uranium in ocean water is  $4.4 \times 10^9$  Ci. The total natural alpha activity inventory including all daughter products of the three natural decay chains is about  $5.0 \times 10^9$  Ci.\*

As discussed earlier, the toxicity of the radionuclides should not be evaluated in terms of curies, but rather in terms of RHI x Ci. It is possible to normalize the toxicity of each decay chain to the toxicity of the parent by calculating a weighted average:

$$T_{\text{norm}} = \frac{(RHI \times Ci)_i}{(Ci)_i} \bigg/ \frac{(RHI \times Ci)_{\text{parent}}}{(Ci)_{\text{parent}}}$$

where  $T_{\text{norm}}$  is the toxicity of the decay chain normalized to the toxicity of the parent.

For U-238 and its daughters in ocean water,

$$T_{\text{norm}}(\text{U-238}) \approx 350 .$$

Thus, taking into account all of the alpha emitting daughters, the chain is 350 times more toxic than U-238 alone. The large value of  $T_{\text{norm}}$  is the consequence of the very large RHI's of the daughter products,

---

\*In addition to the natural isotopes, fallout from weapons testing, re-entry of space vehicles carrying "systems for nuclear auxiliary power" (SNAP), outfall from nuclear fuel reprocessing plants, and releases from nuclear powered vessels, have contributed to the alpha decay inventory of the oceans. These man-made sources of radioactivity in the oceans probably amount to an aggregate alpha activity in the range of  $1$  to  $20 \times 10^6$  Ci.

e.g., Ra-226, RHI = 33,333; Rn-222, RHI = 100,000; Po-210, RHI = 100,000. The  $T_{\text{norm}}$  for the other two decay chains are similar in magnitude.

The total alpha decay toxicity in ocean water can be expressed in terms of U-238 curies equivalent, where

$$\begin{aligned} Q &= [T_{\text{norm}} \times \text{Ci}(\text{U-238})] \\ &= [350 \times 5.0 \times 10^9] \\ &= 1.75 \times 10^{12} \text{ U-238 curies-equivalent} \end{aligned}$$

C. Comparison of Alpha-Decay Isotopes in Radwaste with Natural Alpha-Decay Isotopes in Ocean Water: The isotopes appearing in the lower half of Table B.2 (from U-235 to Am-243) decay by alpha emission, with the exception of Pu-241 which decays 99.99% by beta emission. The Cm-243, 244 isotopes are omitted from this consideration because of their short half-lives which could be contained in an assumed 200 year life expectancy of the integrity of the waste container.

The toxicity of the mixture of alpha emitting isotopes listed in Table B.2 can be normalized to U-238 in the same way that was done in the previous section. The result is:

$$T'_{\text{norm}}(\text{U-238}) = 5.64,$$

where  $T'_{\text{norm}}$  is the toxicity of the mixture of alpha isotopes relative to U-238. The calculation was made for waste that has aged in the dump site for 200 years at which time the integrity of the engineered barriers was assumed to have failed.

To place this in the context of ocean disposal, let us consider a hypothetical case:

### Assumptions:

1. Low level waste from commercial nuclear power plants generated at the annual 1980 rate (Table B.2) is dumped in an ocean site for a period of 100 years (in this example the alpha emitters of importance are: Pu-238, Pu-239 and 240).
2. The engineered barriers for each annual batch fail after 200 years. Residual contents are released over an unspecified time.
3. The isotopes released all remain in solution and are uniformly mixed in the ocean.

The U-238 equivalent placed in the ocean would be:

$$\begin{aligned} Q' &= \text{years} \times \text{Ci/year (1980)} \times T'_{\text{norm}} \text{ (U-238)} \\ &= 10^0 \times 5.2 \times 10^3 \times 5.64 \\ &= 2.9 \times 10^6 \text{ U-238 curies-equivalent} \end{aligned}$$

This value can be compared to Q, the U-238 curies equivalent of natural alpha decay toxicity calculated previously:

$$\begin{aligned} Q'/Q &= 2.9 \times 10^6 / 1.75 \times 10^{12} \\ &= 1.7 \times 10^{-6}. \end{aligned}$$

This represents the increase in the alpha toxicity of ocean water, for a well-mixed ocean. Studies on diffusion and mixing have led to a principle which, in general terms, states that except very close to the source, concentrations in any given volume of water are seldom much greater than the long-term concentrations for a well-mixed ocean [3]. Thus, except in the immediate vicinity of the disposal site, the waste referred to in the example above would lead to a fractional increase in alpha decay toxicity of about 1.7 parts per million.

### Long-Lived Isotopes Which Decay by Beta Emission.

The LLW inventory tables (Tables B.2, B.3 and B.4) list 8 long-lived isotopes which decay by beta emission. The total 1980 annual LLW inventory of these beta emitters is summarized in Table B.6. The quantities for each of these isotopes are a small fraction of the IAEA "Release Rate Limits for a Single Site" [11]. The release rate limits were derived by critical pathway considerations. These isotopes are discussed in a different context, taking into account their geochemical properties.

The volume of water, V, required to dilute these isotopes to MPC can be calculated as follows:

$$V = (\text{inventory in Ci}) \times \text{RHI (150 years)} \times 10^3 \text{ m}^3.$$

The values are summarized in Column 5 of Table B.6. As can be seen, the dilution volume for several of the isotopes is not unreasonable, being in the range of  $10^3$  to  $10^6$  cubic meters. However, there are two radionuclides, Ni-63 and I-129, which would require 0.16 and 0.01 cubic kilometers, respectively, for dilution to MPC.

The case of each of the isotopes in Table B.6 is discussed below, assuming a hypothetical case in which LLW waste is disposed of for 100 years at the 1980 rate of generation.

C-14: The annual 1980 inventory of C-14 would require  $5.9 \times 10^6 \text{ m}^3$  to dilute to MPC. This corresponds to a cube 180 meters on a side. The oceans contain approximately  $1.5 \times 10^7$  Ci of C-14 from natural sources. The annual production of C-14 in the atmosphere by cosmic radiation is  $4.2 \times 10^4$  Ci/yr. Weapons testing has added an additional  $6.7 \times 10^6$  Ci to the atmosphere. Most of the natural and weapons testing C-14 will eventually be deposited in the ocean. The 1980 annual LLW inventory of 46 curies is negligible in comparison with the natural inventory already in the oceans and the annual additions from natural sources and weapons testing fallout.

TABLE B.6

Total 1980 LLW Inventory of Long-Lived Isotopes  
Which Decay by Beta-Emission

The dilution volume is the volume of water (cubic meters) required to dilute the annual (1980) inventory of each isotope to MPC after 150 years of decay.

Isotope	Half-Life (Years)	1980 Inventory <sup>a)</sup> (Ci/year)	IAEA Release Rate Limits <sup>b)</sup> (Ci/year)	Dilution Volume <sup>c)</sup> (m <sup>3</sup> )
C-14	5730	46	$6.1 \times 10^6$	$5.9 \times 10^6$
Cl-36	$3.0 \times 10^5$	$7 \times 10^{-2}$	$3.7 \times 10^9$	$8.8 \times 10^2$
Ni-59	$7.5 \times 10^4$	125	$3.7 \times 10^6$	$1.3 \times 10^5$
Ni-63	100.1	$1.4 \times 10^4$	$6.2 \times 10^6$	$1.6 \times 10^8$
Nb-94	$2.03 \times 10^4$	2.73	N.L. <sup>d)</sup>	$6.8 \times 10^4$
Tc-99	$2.14 \times 10^5$	0.26	$5.9 \times 10^5$	$8.6 \times 10^3$
I-129	$1.56 \times 10^7$	0.58	$6.8 \times 10^5$	$9.9 \times 10^6$
Cs-135	$2.95 \times 10^6$	0.26	$2.0 \times 10^7$	$2.6 \times 10^3$

a) Summation of Tables B.2, B.3 and B.4.

b) Ref. [11].

c) "Dilution Volume" is the volume of water in cubic meters required to dilute the 1980 LLW isotope inventory to MPC after 150 years of decay.

d) N.L. = Not listed in IAEA Release Rate Limits

Cl-36: The annual inventory of Cl-36 in LLW would constitute a negligible addition to the ocean inventory of beta activity, particularly in view of the high concentration of stable chlorine in the form of sodium chloride salt.

Ni-59, Ni-63 and Nb-94: Seawater contains approximately  $2 \times 10^{-3}$  gm/m<sup>3</sup> of stable nickel isotopes and  $1 \times 10^{-5}$  gm/m<sup>3</sup> of stable niobium isotopes [10]. The 1980 inventory of Ni-59 (125 Ci) and Ni-63 ( $1.4 \times 10^4$  Ci) correspond to 1548 and 2471 grams of Ni-59 and Ni-63 respectively. After 150 years of decay these quantities become respectively 1545 and

874 grams. To obtain a 50% dilution (one to one ratio of radioactive to stable nickel isotopes) requires a volume of seawater of  $1.2 \times 10^6 \text{ m}^3$  ( $10^{-3} \text{ km}^3$ , or a cube 100 meters on a side). Thus, a modest amount of seawater would dilute the radioactive annual inventories of Ni-59 and Ni-63 to levels comparable to the stable nickel already present.

Calculations similar to the above for the 1980 annual inventory of Nb-94 (2.73 Ci or 14.6 gms) give a dilution volume of  $1.46 \times 10^6 \text{ m}^3$  to reach 50% dilution (one to one ratio of Nb-94 to stable niobium).

These 50% dilution volumes indicate the assimilative capacity of the well-mixed ocean for quantities of Ni-59, Ni-63 and Nb-94 commonly found in LLW streams.

The primary sources of these isotopes in LLW is from decontamination of primary coolant piping in light water reactors and from scrap reactor internals hardware. It would be practical to segregate these waste streams for land disposal. As more reactors are decommissioned, the quantities of scrap metals containing these activation products will increase. Ocean disposal of very large quantities of such scrap materials will require special studies to assess the long-term implications.

Tc-99: The annual inventory of Tc-99 is small. This isotope appears to present no significant problem in the quantities involved.

I-129: Because of its long half-life, the specific activity of I-129 is low,  $1.78 \times 10^{-4} \text{ Ci/gram}$ . It decays by means of a low energy beta particle (0.150 MeV) accompanied by a soft gamma ray (0.040 MeV). The low specific activity and the low energy radiations make detection and quantitative assay difficult. The oceans contain about 15 to 20 curies of I-129 from natural sources, mostly from spontaneous fission of U-238. It is also produced from spontaneous fission of Th-232 and U-234, reactions induced in Te-128 and Te-130 by free neutrons, and cosmic ray interactions with atmospheric xenon.

Iodine is a biologically active element which concentrates in the human thyroid. This behavior has caused the regulatory authorities to place very restrictive limits on the release of radioisotopes of iodine

to the environment. For example, the EPA Radiation Protection Standards for Nuclear Power Operations places a limit on I-129 entering the general environment from all uranium cycle activities of 5 millicuries per gigawatt-year of electric energy production [12]. The 1980 U.S. commercial nuclear power plant production was 28.34 gigawatt-years (net) of electric energy. Thus, the EPA standards would limit I-129 releases to 0.142 Ci for all operations associated with the 1980 fuel cycle. The 1980 LLW inventory of I-129 was 0.58 Ci, about 4 times the EPA limit.

The EPA limit was designed to restrict airborne releases where the pathways to man are more direct, particularly via the route from forage crop to milk. It can be argued that a less restrictive standard is appropriate for ocean releases. Critical pathway considerations of the IAEA yield a release rate limit of  $6.8 \times 10^5$  Ci/yr [11]. The reason for the large divergence between the EPA "airborne" standard and the IAEA limit is that the ocean water contains a substantial amount of iodine,  $60 \text{ mg/m}^3$ , in the form of the stable isotope I-127.

In the uptake of iodine in the marine food chains, there is on the average no discrimination between the stable isotope I-127 and the radioactive isotope I-129. For public health purposes, the critical factor is the ratio I-129/I-127 which must be less than 1/50 to avoid exceeding maximum dose limits to the thyroid [13]. Of course, regulatory authorities may be expected to set standards which are much more restrictive.

The waters of the North Atlantic contain approximately  $6 \times 10^{15}$  grams of stable I-127. A ratio of I-129/I-127 = 1/50 would be equivalent to  $1.2 \times 10^{14}$  grams of I-129, which is equal to  $2.1 \times 10^{10}$  Ci. This quantity corresponds to all of the I-129 fission product inventory from the fissioning of  $5.27 \times 10^5$  tonnes of U-235, or about  $4.2 \times 10^5$  GWyr of electric energy. In 1980, the total U.S. electric energy production was only 28.34 GWyr from nuclear power plants. If we assume that world production of nuclear energy eventually reaches a constant level of 500 GWyr/year, 840 years would be required to produce  $1.2 \times 10^{14}$  grams of

I-129. Most of this would become a part of the waste inventory for high-level waste disposal and would not be considered for disposal in the ocean.

From these considerations, it can be concluded that I-129 releases in ocean water are not a serious threat to man under any plausible scenario.

Cs-135: The 1980 annual inventory of Cs-135 is extremely small (0.26 Ci) and is negligible in comparison to the IAEA release rate limits ( $2.0 \times 10^7$  Ci/year). The dilution volume for 0.26 Ci required to reach MPC is only  $2.6 \times 10^3 \text{ m}^3$  (see Table B.6). Thus, the quantities of Cs-135 in 1980 LLW inventory appears to be of no significance.

#### Conclusions.

The isotopic content and quantities of radioactivity in low-level waste at current annual generation rates in the United States could be accommodated by ocean disposal without exceeding the limits recommended by the IAEA. Engineered barriers combined with natural barriers would enhance the margin of safety by retaining most of the shorter lived isotopes for the duration of their toxicity. The desirable time span of 200 years for the integrity of the outer container barriers is dictated primarily by the critical isotopes, Sr-90 and Cs-137, because of their high fission yields and their half-lives of approximately 30 years. Based on plausible disposal scenarios, the longer lived isotopes that remain after the engineered barriers have failed would not constitute a significant increase in the radiological toxicity of the ocean waters relative to the natural radioactivity already present in the ocean waters.

## APPENDIX B. REFERENCES

1. International Atomic Energy Agency, "Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, - The Definition Required by Annex I, paragraph 6 to the Convention, and the Recommendations Required by Annex II, Section D," INF/CIRC/205/Add.1/Rev.1, IAEA, Vienna, Austria, August 1978.
2. International Atomic Energy Agency, Radioactive Waste Disposal into the Sea, IAEA Safety Series No. 5, Vienna 1961.
3. International Atomic Energy Agency, The Oceanographic Basis of the IAEA Revised Definition and Recommendations Concerning High-Level Radioactive Waste Unsuitable for Dumping at Sea, Technical document, IAEA-210, Vienna, 1978.
4. International Atomic Energy Agency, The Radiological Basis of the IAEA Revised Definition and Recommendations Concerning High-Level Radioactive Waste Unsuitable for Dumping at Sea, Technical document, IAEA-211, Vienna, 1978.
5. Wild, R.E., O.I. Oztunali, J.J. Clancy, C.J. Pitt and E.D. Picazo, Data Base for Radioactive Waste Management, Waste Source Options Report, Dames and Moore, Inc., NUREG/CR-1759, Volume 2, November, 1981.
6. Lederer, C.M. and V.S. Shirley, editors, Table of Isotopes, 7th Edition, John Wiley and Sons, Inc., New York, 1978.
7. Beck, T.J., L.R. Cooley and M.R. McCampbell, Institutional Radioactive Wastes-1977, NUREG/CR-1137, University of Maryland, Baltimore, Maryland, 1979.
8. Title 10, Code Federal Regulation, Part 20 (10 CFR 20), Appendix B, Table II, Col. 2.
9. Colombo, P., R.M. Neilson, Jr., and M.W. Kendig, Analysis and Evaluation of a Radioactive Waste Package Retrieved from the Atlantic 2800 Meter Disposal site, BNL-51102, U.S. EPA Report No. 520/1-82-009, Washington, D.C., May 1982.
10. Fairbridge, R.W. (editor), The Encyclopedia of Oceanography, Reinhold Publishing Corporation, New York, 1966, p. 518.
11. Nuclear Energy Agency, Review of the Continued Suitability of the Dumping Site for Radioactive Waste in the North-East Atlantic, NEA/OECD, Paris, France, April 1980.
12. Environmental Protection Agency Radiation Protection Standards for Nuclear Power Operations, 40CFR190, 42FR2858, January 13, 1977.
13. Soldat, J.K., Environmental Behavior and Radiation Doses from Iodine-129, Battelle Pacific Northwest Laboratory, Richland, WA, BNWL-SA-4879, June 1974.

## Appendix C. DOMESTIC AND INTERNATIONAL REGULATIONS WHICH POTENTIALLY IMPACT THE OCEAN DISPOSAL OF LOW-LEVEL WASTES

A review of domestic and international regulations concerning the ocean disposal of low-level radioactive wastes is presented. These regulations potentially impact the development of waste package performance criteria, and therefore, must be considered.

### United States.

#### Environmental Protection Agency (EPA).

The Environmental Protection Agency was created under Reorganization Plan Number 3 of 1970 to consolidate in one agency various Federal pollution abatement activities which had been performed under separate organizations. EPA regulations are published as Title 40 of the Code of Federal Regulations. The EPA has the authority to control the ocean disposal of all wastes, including radioactive wastes as specified in the Marine Protection, Research and Sanctuaries Act of 1972.

The Marine Protection, Research and Sanctuaries Act of 1972 (Public Law 92-532) promotes a national policy to regulate the dumping into ocean waters of all materials which would adversely affect human health and welfare or the marine environment, ecological systems or economic potential. The Act prohibits the dumping or transportation for the purpose of dumping any radiological, chemical or biological warfare agent or any high-level radioactive waste into the territorial ocean waters of the United States (defined as 12 nautical miles from U.S. territory). It also describes conditions whereby permits may be issued by EPA for ocean disposal of materials not otherwise prohibited. This Act was amended in 1974 (Public Law 93-254) to implement the provisions of the Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter (London Dumping Convention), and for other purposes.

In January 1977, EPA published regulations concerning the transportation and dumping of wastes in the ocean (40 CFR Parts 220-229). These regulations contain general requirements for all wastes. They also establish a policy of isolation and containment for radioactive waste through two specific criteria:

- (1) Radioactive materials must be contained to prevent their direct dispersion or dilution in ocean waters.
- (2) The materials to be disposed of must decay, decompose or radiodecay to environmentally innocuous materials within the life expectancy of the containers and/or their inert matrix.

In December, 1982, the Ocean Dumping Act was further amended by PL 97-424 to provide for a Radioactive Material Impact Assessment which includes a specific consideration of structural aspects of each radioactive waste container when evaluating any permit for disposal.

#### Nuclear Regulatory Commission (NRC).

The development of commercial nuclear activities has been subject to regulation since 1954. At that time, the Atomic Energy Act of 1954 created the authority to regulate the development of a civilian nuclear power program. The regulatory functions were performed by the Atomic Energy Commission and are currently the responsibility of the Nuclear Regulatory Commission, which was created by the Energy Reorganization Act of 1974. The basic authorities derived from the Atomic Energy Act include the licensing and regulation of production, use, ownership and distribution of special nuclear materials, source material and by-product materials, as well as licensing and control over the manufacture, production, possession, use, importation or exportation of production and utilization facilities.

Nuclear Regulatory Commission regulations are issued under Title 10 of the Code of Federal Regulations. Radioactive waste disposal regulations are primarily found in 10 CFR Part 20 - Standards for Protection

Against Radiation. In particular, it requires NRC authorization for ocean disposal of wastes from NRC licensed facilities. Ocean disposal also requires a permit from the Environmental Protection Agency under the Marine Protection, Research and Sanctuaries Act of 1972 for all non-prohibited radioactive wastes, and for all potential disposers.

#### Department of Transportation (DOT).

The Department of Transportation was established by Congress (PL 89-670) in 1967 to administer and coordinate Federal government transportation programs. The Department of Transportation is authorized to regulate the transportation of explosives and other dangerous articles, including radioactive materials, in interstate and foreign commerce. Under the terms of a memorandum of understanding between the DOT and the NRC, the Department of Transportation has primary responsibility for regulations concerning the transportation of NRC-licensed materials, except for shipments of intermediate and large quantities of radioactive materials and shipments of fissile materials, which are primarily under NRC's jurisdiction. The DOT's primary responsibility is to develop safety standards for the classification and labeling of all packages of radioactive material and regulation of carriers and freight forwarding operations. The Department of Transportation regulations governing transportation of radioactive materials are largely found in Title 49, Parts 170-179 of the Code of Federal Regulations.

#### International.

##### The Nuclear Energy Agency of the Organization for Economic Cooperation and Development (NEA/OECD).

The OECD Nuclear Energy Agency (NEA) was established in 1972, replacing OECD's European Nuclear Energy Agency (ENEA). NEA now includes all the European Member Countries of OECD as well as Australia, Canada, Japan and the United States (23 countries as of April 1980).

NEA's responsibilities lie with the Member Countries of OECD and, recently, fall under a Decision of the OECD Council establishing a Multilateral Consultation and Surveillance Mechanism for Sea Dumping of Radioactive Waste (the OECD Decision) [1]. This mechanism is designed to further the objectives of the London Dumping Convention and it provides for the establishment and review of standards, guidelines and procedures for the safe disposal of radioactive wastes, taking into account the provisions of the London Dumping Convention of 1972 and the IAEA Definition and Recommendations of 1978.

In October 1978, the NEA guidelines were revised to conform to the requirements of the London Dumping Convention and the IAEA Recommendations. This revision was published in April 1979 [2].

#### The London Dumping Convention.

The Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, commonly referred to as the London Dumping Convention, was adopted by an intergovernmental conference in London in November 1972. The London Dumping Convention is an international convention to control marine pollution from the dumping of waste, including the dumping of radioactive material, in international waters. It applies only to those countries, including the United States, who have ratified or acceded to the Convention (47 countries as of October 1980). The International Maritime Consultative Organization (IMCO) was designated as the formal secretariat for the Convention during a meeting of the parties in December 1975.

#### International Atomic Energy Agency (IAEA).

The London Dumping Convention provides for the IAEA (as the competent international body) to define "high-level radioactive wastes or other high-level radioactive matter as unsuitable for dumping at sea" [3]. The IAEA also is entrusted with the responsibility of establishing recommendations that the Contracting Parties to the Convention should

consider in issuing permits for the dumping at sea of radioactive wastes or other radioactive material not otherwise prohibited by the IAEA Definition. Consequently, IAEA responsibilities for recommendations and guidance on the dumping of radioactive waste at sea comes from the London Dumping Convention with the recommendations and guidance being taken into account by the Contracting Parties to the Convention. Such recommendations and guidance were issued by IAEA in 1978 [4].

## APPENDIX C. REFERENCES

1. "Decision of the OECD Council of 22 July 1977 Establishing a Multilateral Consultation and Surveillance Mechanism for Sea Dumping of Radioactive Waste," NEA Sixth Activity Report 1977, OECD Nuclear Energy Agency, Paris, France (1978).
2. Guidelines for Sea Dumping of Radioactive Waste, Revised Version, Nuclear Energy Organization for Economic Cooperation and Development, Paris, France (April 1979).
3. "Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter," drawn up at the Intergovernmental Conference on the Dumping of Wastes at Sea, held in London, October 30-November 10, 1972.
4. International Atomic Energy Agency, "Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, - The Definition Required by Annex I, paragraph 6 to the Convention, and the Recommendations Required by Annex II, Section D," INFCIRC/205/Add.1/Rev. 1, IAEA, Vienna, Austria, August 1978.

**TECHNICAL REPORT DATA**  
(Please read Instructions on the reverse before completing)

1. REPORT NO. EPA 520/1-82-007		2.		3. RECIPIENT'S ACCESSION NO.	
4. TITLE AND SUBTITLE Development of a Working Set of Waste Package Performance Criteria for Deepsea Disposal of Low-level Radioactive Waste				5. REPORT DATE November, 1982	
				6. PERFORMING ORGANIZATION CODE	
7. AUTHOR(S)  P. Colombo, M. Fuhrmann, R.M.Neilson, Jr., and V.L. Sailor				8. PERFORMING ORGANIZATION REPORT NO.  BNL 51525	
9. PERFORMING ORGANIZATION NAME AND ADDRESS Nuclear Waste Research Group Department of Nuclear Energy Brookhaven National Laboratory Upton, New York 11973				10. PROGRAM ELEMENT NO.	
				11. CONTRACT/GRANT NO. Interagency Agreement No. EPA-IAG-AD-89-F-1-558-0	
12. SPONSORING AGENCY NAME AND ADDRESS Office of Radiation Programs U.S. Environmental Protection Agency 401 M Street, S.W. Washington, D.C. 20460				13. TYPE OF REPORT AND PERIOD COVERED Final	
				14. SPONSORING AGENCY CODE  ANR-461	
15. SUPPLEMENTARY NOTES					
16. ABSTRACT  The United States ocean dumping regulations developed pursuant to PL92-532, the Marine Protection, Research, and Sanctuaries Act of 1972, as amended, provide for a general policy of isolation and containment of low-level radioactive waste after disposal into the ocean.  In order to determine whether any particular waste packaging system is adequate to meet this general requirement, it is necessary to establish a set of performance criteria against which to evaluate a particular packaging system. These performance criteria must present requirements for the behavior of the waste in combination with its immobilization agent and outer container in a deepsea environment.  This report presents a <b>working set</b> of waste package performance criteria, and includes a glossary of terms, characteristics of low-level radioactive waste, radioisotopes of importance in low-level radioactive waste, and a summary of domestic and international regulations which control the ocean disposal of these wastes.					
17. KEY WORDS AND DOCUMENT ANALYSIS					
a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS		c. COSATI Field/Group	
Radioactive Waste Packaging Low-Level Radioactive Waste Ocean Dumping/Sea Disposal Radioactive Waste Disposal/Nuclear Waste Disposal					
18. DISTRIBUTION STATEMENT  Unlimited Release		19. SECURITY CLASS (This Report) Unclassified		21. NO. OF PAGES 64	
		20. SECURITY CLASS (This page) Unclassified		22. PRICE	