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COMPATIBILITY OF TECHNOLOGIES WITH REGULATIONS
IN THE WASTE MANAGEMENT OF H-3, I-129, C-14, AND Kr-85.
PART I. INITIAL INFORMATION BASE

by

L. E. Trevorrow, G. F. Vandegrift, V. M. Kolba, and M. J. Steindler

Chemical Technology Division

August 1983

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NOMENCLATURE

NOMENCLATURE

AEC - Atomic Energy Commission
ALARA - As Low as Is Reasonably Achievable
ANL - Argonne National Laboratory
BNL - Brookhaven National Laboratory
BWR - Boiling Water Reactor
CFR - Code of Federal Regulations
CHW - Cladding Hulls Waste
DF - Decontamination Factor
DOE - Department of Energy
DOG - Dissolver Off-Gas
DOT - Department of Transportation
DSC - Differential Scanning Calorimetry
EIS - Environmental Impact Statement
EPA - Environmental Protection Agency
ERDA - Energy Research and Development Administration
FAA - Federal Aviation Administration
FRP - Fuel Reprocessing Plant
GCD - Greater Confinement Disposal
HIP - Hot Isostatic Pressing
HLW - High-Level Waste
HTGR - High Temperature Gas-Cooled Reactor
IAEA - International Atomic Energy Agency
ILW - Intermediate-Level Waste
IMCO - Intergovernmental Maritime Consultative Organization
INEL - Idaho National Engineering Laboratory

NOMENCLATURE (contd)

LANL - Los Alamos National Laboratory	SRL - Savannah River	SR
LIAW - Low-Level Aqueous Waste	SRP - Savannah River	SR
LLRWPA - Low-Level Radioactive Waste Policy Act	TGA - Thermogravimet	TG
LLW - Low-Level Waste	TRU - Transuranic	TR
LMFBR - Liquid Metal Fuel Breeder Reactor	TTC - Transportation	TT
LODC - London Ocean Dumping Convention	USCG - U.S. Coast Guard	USC
LSA - Low Specific Activity	USPO - U. S. Post Office	USP
LWR - Light Water Reactor	WIPP - Waste Isolation	WIP
MPC - Maximum Permissible Concentration		
MT - Metric Ton		
MTHM - Metric Tons of Heavy Metal		
NEA - Nuclear Energy Agency		
NRC - Nuclear Regulatory Commission		
NTS - Nevada Test Site		
NWTS - National Waste Terminal Storage		
OECD - Organization for Economic Cooperation and Development		
ONWI - Office of Nuclear Waste Isolation		
ORNL - Oak Ridge National Laboratory		
PITC - Polymer-Impregnated Tritiated Concrete		
PNL - Pacific Northwest Laboratories		
PWR - Pressurized Water Reactor		
RAMPAC - Radioactive Materials Packaging		
RTV - Room-Temperature Vulcanizing		
SLB - Shallow-Land Burial		

NOMENCLATURE (contd)

ver SRL - Savannah River Laboratory
ver SRP - Savannah River Plant
met TGA - Thermogravimetric Analysis
TRU - Transuranic
ion TTC - Transportation Technology Center
Gua USCG - U.S. Coast Guard
Off' USPO - U. S. Post Office
tion WIPP - Waste Isolation Pilot Plant

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ABSTRACT

This report summarizes the information base that was collected and reviewed in preparation for carrying out an analysis of the compatibility with regulations of waste management technologies for disposal of H-3, I-129, C-14, and Kr-85. Based on the review of this literature, summaries are presented here of waste-form characteristics, packaging, transportation, and disposal methods. Also discussed are regulations that might apply to all operations involved in disposal of the four nuclides, including the processing of irradiated fuel in a fuel reprocessing plant, packaging, storage, transport, and final disposal. The compliance assessment derived from this information is reported in a separate document.

I. INTRODUCTION

A. Function of this Report

Technologies for management of H-3, I-129, C-14, and Kr-85 need to be analyzed with respect to their potential for compliance with acceptance criteria and regulations. This report contains the information base that will be used in such an analysis.

Several topics are not included here that may conventionally fall into the general category of airborne waste management. (1) Several nuclides released to the atmosphere, especially at reactor sites, are considered to have such a small impact, mainly because of short half-life, that collection and packaging of the waste are not required. (2) Radon-222 is an airborne nuclide that arises in some parts of the nuclear fuel cycle, but it will not be managed by collection and packaging; therefore, this nuclide is not considered here. (3) Particulate solids bearing a number of radioactive nuclides, including actinide elements and semivolatiles, e.g., ruthenium, cesium, and strontium, would also be released into some gaseous streams of an FRP.* Management of particulate solids, however, is not considered here. These solids will be collected on High Efficiency Particulate (HEPA) filters whose development, efficiency, and use are considered elsewhere. The HEPA filters bearing the actinides and semivolatile fission products filtered from gaseous streams of the FRP will fall into the class of TRU wastes for which DOE supports a separate program and for which a separate disposal destination will be assigned.

* Acronyms used in this report are defined on pp. xi-xii.

Furthermore, although a number of airborne wastes result from operations at a variety of facilities of the uranium fuel cycle, e.g., mines, mills, UF_6 conversion plants, and fuel-fabrication plants, the four nuclides assigned in the charter of this work are released in significant amounts only at an FRP. For that reason, the operations of the FRP only are discussed here. Although the release of radioiodine at reactors is a matter of concern [MURPHY-1983], it is mainly short-lived I-131 that requires removal from the effluents. Because the half-life of I-131 is 8 d, long-term management and disposal do not enter into the plans for control of this isotope.

Thus, the nuclides addressed in this study are limited to those that will be carried in gaseous streams and that have sufficiently long half-lives to require storage and disposal. The work is not concerned with short-lived gaseous radionuclides that can be managed by delay beds; furthermore, it is not concerned with particulate solids carried by gaseous streams.

Development of disposal modes for airborne wastes (Kr-85, I-129, H-3, and C-14) and also development of regulations and acceptance criteria for such nuclear wastes have been carried out in several unrelated activities. Furthermore, workers engaged in development of disposal modes and regulations regarding them have pursued similar goals, but from widely different starting positions. On the one hand, the focus of immobilization and waste-form development has been physicochemical behavior of materials and processes, and the orientation of packaging and transportation development has been strength of materials and the design and mechanical performance of packages. Development of regulations, on the other hand, has centered on setting limits for radioactivity exposure of the public and workers in the nuclear industry. Because these efforts have been pursued with some degree of independence, it is not obvious whether the contemplated disposal modes can comply with regulations or what disposal sites can accept these wastes. The information summarized in this report forms the basis for a subsequent effort to characterize the proposed disposal modes, assess their capability for complying with applicable regulations, and recommend changes wherever necessary and possible.

B. Study Plan

The characteristics of airborne wastes will probably not place them in the categories of wastes that may be destined for deep geologic disposal, i.e., HLW and TRU; therefore, it would be convenient if such wastes were acceptable to some other defined mode of disposal, such as SLB. Much experience on handling LLW and on disposing of it by SLB has accumulated, and much recent attention has been devoted to improving this disposal mode. The plan of this study is to make a series of tests of the compatibility of conceptual disposal modes with regulations. The first test involves a determination of which airborne wastes would be acceptable for SLB disposal, based upon nine proposed airborne waste forms* and current packaging concepts. Those waste forms that do not

*Nine waste forms have been assigned as the current primary candidates: (1) krypton in cylinders, (2) krypton in zeolites, (3) krypton in sputtered metals, (4) iodine on silver sorbents, (5) iodine in barium iodate, (6) carbon in barium carbonate, (7) carbon in calcium carbonate, (8) tritium in concrete, and (9) tritium in molecular sieves.

seem acceptable for SLB are given a second test of compliance, in which some alteration in waste systems,* use of interim storage, or changes in waste forms and containers is assumed. For any waste forms that are not suitable for SLB, even with these changes, a third test will be made to determine whether current or alternative waste forms and containers would be acceptable in some other disposal mode, including seabed emplacement, deep geologic disposal, and greater confinement disposal. In this sequence of tests, for each airborne nuclide, at the first indication of compliance of a disposal mode with regulations, the testing would go no further, and the disposal mode for this nuclide would then be characterized.

C. Scope of the Study

The scope of this study will depend on how far conceptual waste forms are from complying with regulations. If the nine waste forms comply with regulations for SLB, then the scope will be simply limited to the given waste forms and the SLB disposal mode. At the extreme, the scope would have to be expanded to examine several other disposal modes and, ultimately, regulation changes would have to be recommended. Although the actual analysis (part II) may not need to proceed as far as this extreme, this information base describes all possible disposal modes for the different waste forms and lists the source of more detailed information on them in case they must be used in the analysis.

The information that is most relevant to the main purpose of this work concerns the physical and chemical properties of the waste forms intended for disposal. Nevertheless, some information on sources of these wastes, primary forms,[†] collection processes, and immobilization processes is also provided here to furnish a complete picture of the derivation of final forms from primary forms. Although much data are available on the properties of pure compounds (e.g., barium iodate), the final storage forms (e.g., barium iodate incorporated into concrete) have been less thoroughly characterized, and in several cases not quantitatively characterized at all.

Information that would best represent reference concepts (i.e., those most likely to be put into practice) have been collected from literature, expert advice, and experimenters. Information on regulations likely to affect airborne waste disposal, i.e., regulations for (1) LLW disposal modes, (2) other applicable disposal modes, and (3) transportation, have also been collected. Regulatory information has been analyzed to facilitate its use in evaluation of waste form performance.

From the many options for airborne waste management that have been explored, those few considered by current workers as most likely to be applied were selected for attention. These options include immobilization of Kr-85 by ion implantation, by zeolites, and by compression of gas into cylinders; immobilization of I-129 by silver sorbents and by barium iodate incorporated into concrete; immobilization of tritium by concrete and by molecular sieves;

* Inclusive of the entire set of steps necessary to accomplish disposal: collection, packaging, transportation, and final emplacement.

[†] Primary forms are those in which the nuclides appear in plant streams before collection.

and immobilization of carbon by barium or calcium carbonates incorporated into concrete. The few other options considered have been limited to concepts that seem most feasible [PROUT]. Furthermore, other elements of airborne waste disposal considered here include container technology and transportation modes, shallow-land burial, intermediate-depth burial, interim storage, deep-geologic disposal, and seabed emplacement. The regulations considered include those of DOE, EPA, NWTs, WIPP, DOT, and other appropriate sources addressing environmental limits.

The question that will be addressed eventually is whether the physical and chemical properties of the waste forms chosen by developers will be sufficient for the waste-form performance that is either specified or can be inferred from the regulations for handling, transportation, and disposal. Only the physical and chemical properties that are relevant to these questions are included here. Obviously, the properties that will be relevant are those generally related to mobility, e.g., volatility, solubility, and chemical and physical stability.

II. PRIMARY WASTE FORMS

A. Occurrence of Airborne Wastes

The four nuclides in irradiated nuclear fuels that are the focus of interest in this work, H-3, I-129, C-14, and Kr-85, have been referred to commonly as airborne wastes because they are likely to appear in the off-gas of an air sparge of the dissolver in a conventional Purex fuel-reprocessing flowsheet. In the U.S., the commercial fuel cycle (excepting experimental reactors), the weapons fuel cycle, and the naval propulsion fuel cycle involve irradiated fuels. Reprocessing has been carried out on a regular basis in the latter two fuel cycles; but, except for a few campaigns at the West Valley plant and scattered R&D activities at national laboratories, the reprocessing of commercial fuels on a routine basis has not been performed in the U.S.

The four airborne nuclides will be distributed throughout several streams of a nuclear fuel cycle, in a pattern that depends on the reactor type and on the reprocessing scheme. This study will focus on a reference system of zircaloy-clad uranium oxide fuels removed from an LWR for reprocessing by the Purex process. Some of the airborne nuclides are produced in cladding and coolant, resulting in releases of relatively small amounts at reactors [NUCLSAF]. Small amounts of the nuclides may also be released at storage basins, R&D facilities, and medical facilities, but these releases are not of concern here. The focus of this work is on the problem of managing the major fraction of the nuclides that are freed when irradiated fuels are disintegrated during reprocessing.

B. Composition of the Primary Waste Stream

Estimates of the composition of the off-gas stream carrying the airborne nuclides in a fuel reprocessing plant have been given in several works [ALTOMARE, DP-CFP-78-121, BLANCETT, BROWN-1978], but these estimates of composition disagree quantitatively. The general scheme for the reprocessing of spent fuel carrying airborne nuclides is shown in Fig. II-1. The major components of the gas stream are from the air that is used to sparge the dissolver. The composition of the gas stream will depend on (1) whether some other gas, such as argon, is used to purge the fuel shearing cell [EVANS] and (2) whether the "voloxidation"* operation is used to remove tritium from the fuel before dissolution.

* Defined as the oxidation of chopped fuel to convert elemental tritium to tritium oxide, which is then fixed on a desiccant. The objective of this operation is to prevent dispersion of the tritium in the relatively large volumes of dissolver solution.

Fig. II 1. Generic Flow Scheme for Airborne Nuclides in Fuel-Reprocessing Operations

* Purge gas: large fraction of noble gas inventory, small fraction of tritium inventory.

† Removal of tritium, noble gases, and, if thermal outgassing is used, a large fraction of iodine inventory.

III. TECHNOLOGIES OF AIRBORNE WASTE FORMS

In this section, the technologies of waste management for the four nuclides of interest are summarized. This summary includes information on the radioactivities of the four nuclides, methods for their collection from FRP streams and subsequent immobilization, and characteristics of the final waste form. Since many of the final waste forms involve a concrete matrix, a short review of pertinent properties of concrete is included.

A. Technology of Krypton Waste Forms

1. Decay Characteristics of Krypton-85

Krypton-85 has a half-life of radioactive decay of 10.72 y. It decays by β^- emission, with a 514-KeV γ ray associated with the less intense β^- particles (Table III-1); Kr-85 decay produces a heat output of 1.50×10^{-3} W/Ci = 5.88×10^{-1} W/g Kr-85. Because Kr-85 will be only about 6% of the Kr collected from a reprocessing plant's air streams, its specific activity of 393 Ci/g Kr-85 will be diluted to 24 Ci/g Kr.

Table III-1. Characteristics of the Radioactive Decay of Kr-85 [MARTIN]

Radiation Type	Energy, keV	Intensity	Δ (g-rad/ μ Ci-h) ^a
β_1^- max	173 $\pm$ 2		
avg	47.5 $\pm$ 0.6	0.43 $\pm$ 0.01	0.0004
β_2^- max	687 $\pm$ 2		
avg	251.4 $\pm$ 0.8	99.57 $\pm$ 0.01	0.533
Total β^- avg	250.5 $\pm$ 0.8	100.000 $\pm$ 0.015	0.534
γ_1	513.99 $\pm$ 0.01	0.43 $\pm$ 0.01	0.0047
Total $\Sigma(\Delta_i)$ i	---	---	0.538

^aOne g-rad/ μ Ci-h is equivalent to 2.78×10^{-3} W/Ci. For an infinite, homogeneous medium in which a source is uniformly dispersed with a concentration of 1 μ Ci-h/g, Δ gives the absorbed dose in rads. For each radiation type i,

$$\Delta_i = 0.00213 I_i \times E_i$$

where I_i = intensity of i and E_i = energy of i. The factor 0.00213 is the conversion factor for keV/disintegration to g-rad/ μ Ci-h. The equilibrium absorbed dose, Deq , in rads equals $C_{85Kr} \times \Sigma \Delta_i$, where C_{85Kr} is concentration in units of μ Ci-h/g.

2. Collection of Krypton

For collection of krypton-85, two processes are currently receiving most attention [PENCE-1981A]: fluorocarbon absorption and cryogenic distillation. Collection of noble gases by adsorption on inorganic solids has been investigated on a laboratory scale, but is not developed to the extent of the collection processes named above. It has received serious attention for application to cleanup of fuel-reprocessing off-gases only relatively recently [PENCE-1981B, MONSON, RUTHVEN]. Cryogenic distillation has been judged the most advanced collection technology [PENCE-1981A, MONSON, BENEDICT-1979]. Absorption by fluorocarbon has successfully passed pilot-scale tests in the U.S. for scrubbing krypton from a gas stream [MERRIMAN-1982]. Purification of the stripper product, however, has not been developed. Cryogenic distillation has been in use at the Idaho Chemical Processing Plant since the mid-1960s, although in a system that was not designed for quantitative removal of krypton. This process has been the subject of considerable development work in European laboratories [VON AMMON-1978, -1980, -1981, -1983], particularly in Germany and Belgium, and it will be demonstrated on a small plant for the first time at the Tokai Mura reprocessing plant in Japan [PENCE-1981A]. Although the Tokai Mura Plant and U.S. studies [BROWN-1978, DOE/EIS-0046D, MELLINGER] have selected cryogenic distillation as the reference process for noble gas collection, a substantial amount of large-scale development on noble gas collection in the U.S. has involved the process of absorption in liquid fluorocarbons. The cryogenic distillation technologies in the Federal Republic of Germany and Belgium can be obtained in the U.S. through international waste management exchange agreements [CHRISTIAN].

Noble gas collection technology is judged to be ready for testing in a large-scale plant in long-term, fully-simulated conditions.

a. Pretreatment of Primary Gas Stream

All three collection processes are said [PENCE-1981A] to require thorough pretreatment of the off-gas stream in preparation for application of krypton collection processes. The pretreatment is applied to remove particulate solids, a major part of the NO_x [VON AMMON-1983], semivolatile fission products (ruthenium), water, iodine, CO_2 , and methane. All the noble gas collection processes require reduction of water vapor in the gas stream to about 1 ppm [PENCE-1981A]. The major part of the water can be removed by a condenser and the residual can be cleaned up by solid inorganic desiccants, e.g., molecular sieves. The major portion of NO_x is expected [PENCE-1981A] to be removed by an aqueous absorption column and the remaining 10-20% in the DOG stream would be removed by (1) reaction with ammonia on a synthetic mordenite catalyst or (2) by reaction with hydrogen on a noble metal catalyst. The choice between these two pretreatment processes will depend on which collection process is to be used. For example, if cryogenic distillation is to be used, the destruction of NO_x by hydrogen is preferred because a slight excess of hydrogen removes oxygen from the system, thus avoiding the formation and collection of ozone and its accompanying explosion hazard. Studies have been aimed at determining the optimal hydrogen concentration and catalyst [VON AMMON-1980]. On the other hand, if krypton is to be collected by liquid absorption or by adsorption on inorganic solids, then there is no reason to

employ hydrogen for its secondary benefit of oxygen removal; instead, the reaction involving ammonia and mordenite can be used for NO_x destruction. Recent reports [BRUGGEMAN-1979] on the effectiveness of the ammonia reaction, however, seem to indicate that it is not completely without uncertainties. For example, some reduction in performance of the catalyst attributed to high catalyst bed temperatures was observed in pilot-scale studies. Furthermore, catalyst efficiency seems to be affected by excessive moisture concentration in the gas stream, making control of moisture content critical.

The order of pretreatment operations is important and depends on which collection process is to be used [PENCE-1981A] for the various gas streams. It is suggested [PENCE-1981A] that, if the platinum-catalyzed hydrogen reaction is used to remove residual NO_x , then iodine removal should precede NO_x removal to prevent iodine poisoning of the platinum catalyst. Furthermore, if alkaline-earth hydroxide hydrates are used to remove $^{14}\text{CO}_2$, then this operation should precede the water removal by adsorbents. If NO_x destruction is done by the mordenite-catalyzed reaction with ammonia, the iodine-removal reaction would be placed downstream of the NO_x destructor, which removes ruthenium before the iodine is collected and avoids interference of ruthenium with iodine removal.

b. Cryogenic Distillation

In a typical flow scheme (Fig. III-1) for the cryogenic distillation process, a three-component system of krypton, xenon, and nitrogen is compressed to 500-800 kPa and cooled to about -140 to -150°C before injection at the midpoint of a column packed with sieve plates or other appropriate surface [PENCE-1981A]. Some of the nitrogen is cooled to condensation at the top of the column by a liquid-nitrogen-cooled heat exchanger, causing a down-flow of liquid nitrogen inside the column. Krypton and xenon are absorbed in the condensed nitrogen and are collected in the bottom of the column. The condensed liquid at the bottom of the column is heated to create a reflux. The temperature profile in the column must be controlled to prevent freezing of xenon and resultant plugging. Krypton and xenon are separated in a subsequent rectification column, operated at a pressure of 300 kPa; separation would be carried out as a batch operation. It has been suggested [KLETT-1981] that, if krypton is to be stored as a compressed gas in cylinders, then separation of xenon may not be worthwhile. To avoid problems following from the freezing of xenon, one scheme [PENCE-1981A] removes the xenon by freezing it on a chilled surface (fp, -112°C), before it enters the primary column. In the Tokai Mura Plant, xenon will be removed with a silica gel bed at -168°C .

A cryogenic process using liquid argon as the solvent for krypton and xenon absorption is planned for study at Fontenay-aux-Roses. This process has the same pretreatment requirements as the nitrogen solvent process, but problems caused by freezing of xenon are eliminated. Ozone explosion limits in argon, however, may be too low for safety [PENZHORN-1981].

In pilot-plant studies, the DF for krypton removal from simulated off-gas streams was 10^2 - 10^3 , which is greater than the anticipated needs for regulatory compliance. The Karlsruhe facility has supported a major effort on development of cryogenic distillation for removal of krypton from

Fig. III-1. Flow Scheme for Collection of Kr by Cryogenic Distillation (derived from [PENCE-1981A])

fuel-reprocessing gases [VON AMMON-1978, -1981]. None of the development projects has been operated for long periods of time in a highly radioactive environment; thus, specifications for actual service have not been well defined [PENCE-1981A]. The Belgian pilot plant has been operated for 10,000 h [CHRISTIAN].

c. Liquid Absorption

Experimental investigation of liquid absorption for collection of fission-product noble gases began at the Oak Ridge Gaseous Diffusion Plant in 1967. Pilot-scale testing of the effects of various parameters, including tests with radioactive isotopes (Kr-85, Xe-133, and I-131), CO₂, NO₂, N₂O, NO, and H₂O, have been completed. The liquid absorption method is based on exploiting differences in solubility between the noble gases and other constituents of the off-gas streams in fluorocarbon solvents. The reference solvent is dichlorodifluoromethane, CCl₂F₂. Demonstrated efficiencies [MERRIMAN-1980] for removal of Kr and Xe from various carrier streams such as N₂, Ar, He, and H₂ are said to be 99.9%. It is also claimed that liquid absorption could not only remove ¹⁴CO₂ from off-gas streams, but also serve as a "backup" system for iodine collection [STEPHENSON-1976].

Initial pilot-scale work [STEPHENSON-1976] involved three columns: (1) an absorber column for the main separation of noble gases from bulk gas carrier, (2) a fractional stripper column for removal of the

lower-boiling carrier gas from the solvent, and (3) a final stripper for removal of all remaining gas from the solvent, which is to be recycled to the absorber column. Depending on the carrier gas composition, the recycled solvent may have to be purified in a plant by removing the less volatile constituents such as NO_x and I_2 ; solvent purification has not been developed [CHRISTIAN]. In later work, a single column was designed, built, and tested; this column is intended to carry out, through control of local temperatures and pressures, all three functions of the original three columns (Fig. III-2). The combination column, requiring less equipment and instrumentation than the single one, is expected to offer operational simplicity and economic advantages. The feasibility of the column is reported to have been established in testing [MERRIMAN-1980].

The purity of the krypton produced by the liquid absorption process can be adjusted to values $>90\%$, depending on the use of subsequent steps involving cryogenic trapping and solid sorbents to remove fluorocarbon, CO_2 , and xenon [LITTLE, MERRIMAN-1980].

d. Adsorption on Solids

The order of pretreatment operations for removal of NO_x , iodine, water, and CO_2 before krypton adsorption (Fig. III-3) is similar to that recommended for pretreatment preceding the liquid absorption process. After the pretreatment operations, xenon would be removed from the mixture of krypton, xenon, and air at ambient temperature and pressure by adsorption onto a bed of synthetic mordenite, probably silver mordenite [MONSON]. The gas stream that passes through this bed would be cooled to about -80°C and passed through a cooled bed of another type of zeolite, probably hydrogen mordenite [MONSON], which would adsorb most of the krypton. Other constituents of the

Fig. III-2. Flow Scheme for Collection of Kr by Liquid Absorption (derived from [PENCE-1981A, MERRIMAN-1980])

Fig. III-3. Flow Scheme for Collection of Kr by Adsorption on Solids (from description by [PENCE-1981A])

gas stream would pass through this second column to be vented as decontaminated effluent. The krypton is desorbed from this second column at 60°C and recharged to -80°C before it is passed into a third column for concentration (additional purification) by adsorption onto the same type of zeolite used in the second column. Finally, the krypton would be desorbed from the third column at 60°C, condensed into a cold trap, and then expanded into cylinders for interim or final storage. Laboratory-scale tests of the adsorption method have shown DF's of at least 4×10^2 for krypton and 4×10^3 for xenon [PENCE-1981A]. In a plant, beds of large diameter would be needed; the transfer of heat in large beds has not been addressed. Also, the treatment of krypton that is absorbed onto the xenon bed should be considered [CHRISTIAN].

3. Immobilization of Krypton

Although extensive R&D programs have investigated the production of zeolite and sputtered-metal waste forms, the compressed-gas form is considered by evaluators [MELLINGER] to be the only method that has been demonstrated and is available for immobilization of krypton. The solidification technologies, however, have been sufficiently developed to indicate success may be achieved after additional refinements [CHRISTIAN].

a. Preparation of Compressed-Gas Storage Form

The operation used to place krypton gas in cylinders for final storage will depend on the collection process employed. Since cryogenic equipment is involved in the collection operations of both cryogenic distillation and adsorption on solids, the krypton may be condensed by cryogenic methods

and allowed to expand into cylinders to the desired pressure. If, on the other hand, collection is done by liquid absorption, cryogenic equipment may not be a part of the facility--in which case purified, collected krypton gas will be placed into cylinders by a mechanical compressor [PENCE-1981A, BROWN-1978, MERRIMAN-1982]. The cylinders might also be filled without mechanical compression by condensing a measured amount of gas with liquid nitrogen coolant [YAMAMOTO].

b. Characteristics of Compressed Gas Storage Form

Although Kr-85 has been routinely collected, stored, and shipped in cylinders as a part of the operation of the Idaho Chemical Processing Plant [BENDIASLV-1974, -1971, -1973, -1975A, -1975B, -1976], the management of the Kr-85 from a full-scale fuel-reprocessing plant in cylinders is still at the conceptual stage. As yet, there is no generally accepted reference design. In the absence of a reference, a thorough description of this mode of krypton management is probably best given by listing the results of several studies, which is done in Table III-2. The question of whether the xenon concentration should be reduced substantially, to decrease the volume and, therefore, the cost of storage, has been explored in economic analyses. Klett [KLETT-1981] has concluded that for storage as compressed gas the cost of cryogenic distillation to remove the xenon would justify the economic benefit realized from storage of smaller volumes.

Only certain alloys, Type 316 stainless steel and Nitronic 50, have been found to be suitable for resistance to corrosion by the rubidium produced in decay of Kr-85 [PINCHBACK-1979B, NAGATA]. Experience at the Tokai Mura plant on cylinder storage of Kr-85 will be valuable. Valves have historically been sources of corrosion and leaking problems with storage of even small quantities of Kr-85 [McCLANAHAN].

c. Preparation of Zeolite Waste Form

In the zeolite waste form, krypton is sorbed into microcavities in the structure of zeolites (crystalline aluminosilicates that contain cavities joined by pores of a characteristic size). The gas is emplaced within cavities, and the zeolite structure is sintered to an amorphous material at high temperature and pressure to seal the krypton in the solid. The rate of leakage of gases from these solids is quite low at room temperature and at temperatures expected for terminal disposal of radioactive krypton (200-350°C). Early work in the U.S. involved sodalite, but Penzhorn [PENZHORN-1980A], in the Federal Republic of Germany, found that zeolite 5A offers the highest storage capacities and lowest leak rates. Subsequent development in the U.S. has involved zeolite 5A [CHRISTENSEN].

In the process developed in the U.S., dried zeolite 5A, in the form of small spheres and a powdered-glass binder, is loaded into a capsule, which is then evacuated and heated to 400-700°C. Control of the moisture content is important and is correlated with the temperature and pressure of encapsulation. Krypton is introduced into the capsule to pressures of up to 1000 atm, depending on the loading required. This pressure is balanced by a helium pressure on the outside surfaces of the capsule. After a soak time of 0.5-1.0 h, the krypton that has not been incorporated into the solid (about

Table III-2. Summary of Several Characterizations of Storage of Kr as Compressed Gas

	[KLETT-1981, -1982]							
					Central Repository		On-Site Repository	
	[FOSTER-1975A]		[WARNECKE]	[BROWN -1978]	Kr Only	1 Kr/11 Xe Mixture	Kr Only	1 Kr/11 Xe Mixture
Cylinder Vol., L	50	50	50	50	310	310	50	50
Peak Pressure, kPa ^a	3.45×10 ³	1.38×10 ⁴	2.0×10 ⁴	3.44×10 ³	5.2×10 ³	6.5×10 ³	1.2×10 ⁴	1.5×10 ⁴
kCi/cyl ^b	128	419	340	128	740	123	246	38.5
Peak Temp., °C	60	127	200	60	251	58.5	220	51.5
Heat generation/ cyl, W	186	611	534	186	1060	177	581	92.9
Cyl/GW(e)-y ^c	2.5	0.76	0.9		0.359	2.16	1.08	6.89
Dose Rate, rem/h			1.44×10 ⁴ (surface)	4.3×10 ² (at 3 ft)				
Leak Rate, %/y			3.2×10 ⁻⁶					
Kr, %	100		90		100	8.3	100	8.3
Kr-85, % of total Kr	6		6.8	6		6		6
Xe, %			10			91.7		91.7
N ₂ , %			0.4					
Ar, %			0.08					
O ₂ , ppm			1					
H ₂ O, ppm			1					
CO ₂ , ppm			1					
Cylinder ID, in.				9	11.25	11.25	9	
Cylinder length, in.				52	170.4	170.4	65.5	
Shielding thickness, in. Pb				3				
Cyl Material					316SS	316SS	316SS	

^a1 kPa = 0.00988 atm (approx. 0.01 atm).

^bCyl = cylinder.

^cGW(e)-y = Gigawatt (electric) - year.

65% of that introduced to the capsule) is recycled. During the process, the zeolite sinters to an amorphous, glassy state. Hot isostatic pressing (HIP) is employed to compact the zeolite and glass powder to a monolithic shape. The capsule containing the monolith is sealed into a canister for terminal storage [CHRISTENSEN]. At Exxon-Nuclear Idaho Co., the process has been demonstrated with nonradioactive krypton on a full scale. Also, several hundred Curies of Kr-85 have been encapsulated in a small-scale HIP unit. The samples will be observed for a decade to determine any long-term effects from rubidium formation on krypton leak rates and waste-form properties [CHRISTIAN].

d. Characteristics of Zeolite Waste Form

Characteristics of the zeolite 5A waste form for storage of krypton-85 are listed in Table III-3. Although earlier work was based on storage of krypton on sodalite and similar solids, it seems to be generally agreed, on the basis of the most recent work, that zeolite 5A is the best choice for storage of Kr-85 in this type of solid. It has been concluded [PLUNG] that, for storage in zeolite, prior removal of xenon from the gas seems to be justified by the increased krypton loading that is possible with high krypton concentrations.

A brief test on the effect of irradiation on krypton-loaded zeolites was made. A zeolite sample was irradiated with a 10^6 kJ/kg dose of gamma energy, simulating a 2.7-month storage of Kr-85. The sample contained 37 cm^3 (STP)/g of zeolite 5A. In this test, about 0.009% of the krypton was released [PENZHORN-1980B].

In a test of the effect of water on krypton-loaded zeolites, Penzhorn et al. [PENZHORN-1980B] found "no significant gas desorption" from a sample containing 37 cm^3 (STP) Kr/g zeolite 5A that was "stored under water for weeks." In a test of the effect of water on krypton-loaded zeolites by Knecht [KNECHT-1981], no release of krypton was observed from samples immersed in water.

e. Preparation of Sputtered-Metal Waste Form

In the preparation of sputtered-metal waste form, krypton is implanted into a metal surface by electrostatic acceleration of positively charged krypton ions. The surface is also coated with sputtered metal, covering the implanted krypton atoms. This ion implantation and film covering is rapidly alternated until a thick layer ($\sim 1 \text{ cm}$) of sputtered metal containing implanted krypton is produced.

This operation is carried out in a vacuum chamber. Krypton is admitted to a previously evacuated chamber and subjected to bombardment by electrons that are accelerated by a potential difference between a heated filament and an anode. This bombardment produces positively charged krypton ions, which are accelerated to a target, negatively charged with respect to the anode, of the metal to be sputtered. In striking the target, the Kr^+ ions eject atoms of the target metal, which travel to the substrate surface and deposit there. The substrate, like the target, is negatively charged with respect to the anode in order to attract the Kr^+ ions, and the ratio of substrate charge to anode charge is selected to maximize the krypton content and to produce a net deposit on the substrate.

Table III-3. Characteristics of Zeolite Waste Form for Storage of Krypton

Radiophase:	gas
Encapsulant Material:	Zeolite 5A
Estimated Leakage:	0.03% in 10 y @ 300°C ^a 0.3% in 10 y @ 400°C ^a
Density:	1 g/cm ^{3a}
Loading:	50 m ³ (STP) Kr/m ³ solid ^a
Effect of Irradiation:	γ energy, simulating 2.7-month storage, released 0.009% of Kr. ^b
Effect of H ₂ O:	No gas desorption from sample stored under water for weeks. ^b
	No release of Kr from samples immersed in water. ^c

^a[CHRISTENSEN].^b[PENZHORN-1980B].^c[KNECHT-1981].

Since a cylindrical chamber, target, and substrate are used in this operation, the waste form will also be cylindrical, *i.e.*, a layer of sputtered metal containing entrapped krypton on the inner surface of a metallic cylinder with a hemispherical, closed bottom. When the sputtered metal layer reaches the desired thickness, the vacuum chamber is detached from the vacuum system, sealed, and treated as the disposal canister.

In a plant-scale concept [PARSONS] of the sputtering method, the substrate is a standard 10-in. SS pipe and pipe cap that is seal-welded to provide encapsulation after deposition is complete. The target is a standard 6-in. steel pipe and pipe cap, with the outer surface initially coated with 1.3 cm of 79 wt % nickel and 21 wt % lanthanum, a sufficient quantity to result in a 1-cm thick deposit of Ni-La on the inner wall of the substrate pipe. The 6-in. target pipe is centered inside the substrate pipe. The annulus between the two pipes contains the appropriate gas phase and a tungsten filament. The substrate, target, and filament are all electrically insulated from one another. A disposal sputtering chamber, which serves as the primary disposal canister and is not opened after the sputtering process, has been designed and tested on a laboratory scale. It must be tested on the full scale. Remote operations must also be developed. Two 10-Ci samples of Kr-85 have been prepared and three 200-Ci samples will be prepared in FY 1983. These will be observed for a decade to determine long-term storage characteristics [CHRISTIAN].

f. Characteristics of Sputtered-Metal Waste Form

Some of the physical characteristics of the sputtered-metal waste form are listed in Table III-4.

Investigations of the behavior of krypton-loaded sputtered metal have shown that, for nickel-lanthanum alloys, a structure change from a glassy to crystalline state occurs at 350-400°C and that higher temperatures (550-800°C) produce a significant krypton release rate [TINGEY-1982A]. "No major increases in release rates were observed" on exposure of the sputtered material to liquid-phase water at 25 and 90°C; the results of long-term exposure are expected to become available in the future [LYTLE]. An alloy of nickel, lanthanum, and molybdenum was stable to a temperature of 740°C, but the molybdenum-containing alloy was considered less desirable because of its lower trapping rate for krypton. Exposure of the Ni-La-Kr material to oxygen at 320°C resulted in a temporary, small increase in krypton release rate; but, after the initial exposure or on removal of the oxygen, the krypton release rate was lower than normal. Evidently, a protective and diffusion-resistant oxide layer forms.

Table III-4. Characteristics of Sputtered Metal Waste Form for Storage of Krypton

Radiophase:	gas
Encapsulated Material:	$\text{Ni}_{0.81}\text{La}_{0.09}\text{Kr}_{0.10}^{\text{a}}$ (sputtered material)
Substrate Material:	SS p.p.e, 10 in. dia (serves as primary canister) ^b
Estimated Leakage:	0.5% in 10 y @ 350°C ^a
Loading:	34 cm ³ (STP) Kr/g deposit ^a
Effect of Water:	No release of Kr at 25 and 90°C. ^c

^a[TINGEY-1982B].

^b[PARSONS].

^c[LYTLE].

g. Alternative Krypton Storage Forms

In addition to the storage forms described above, a large number of others have been suggested and have been evaluated according to technical and economic feasibility to varying extents [THAMER-1978,-1979; FOSTER-1975B; KNECHT-1977]. These alternatives are not described in detail here because, at this time, they are considered much less likely to be applied than the three preferred storage forms. They would be seriously reconsidered only in the event that all three storage forms described above prove infeasible. These alternatives include (1) entrapment of krypton in clathrates [STAPLES-1974, CHLECK, MOCK]; (2) storage in porous, underground media as a gas [REIST, TADMOR, KING,

JACOBS, MUDRA, WINAR]; (3) incorporation of krypton into a grout that is forced underground into horizontal fractures (hydrofracture) created in shale beds [PERETZ]*; (4) storage at subatmospheric pressures in tanks [DUNSTER]; (5) storage of Kr-85 as a solution in organic liquids [WINAR] or in glass [GRAY-1978, -1979]. Several other alternatives have been suggested that are judged [THAMER-1978, -1979]--because of cost, lack of development, lack of data, high release rate, or other unfavorable properties of the storage form--to be less attractive than those listed above.

h. Accumulation of Rubidium in Stored Kr-85

The corrosive effect of the product of Kr-85 decay, rubidium, on the primary container has been a major concern in planning and development for Kr-85 storage. Because of this concern, the rubidium corrosion of alloys and possible coatings has been investigated [PINCHBACK-1979A, 1979B; NAGATA; KLETT-1981], which resulted in recommendations for the materials to be used in primary containers for storing Kr-85. The conclusions from corrosion and stress-corrosion tests on several alloys were that 316 SS and Nitronic 50 provide containment for 50-100 y [NAGATA]. The tests also showed that oxygen and water greatly increased the corrosion rates and should be held to concentrations of <100 ppm. Penzhorn [PENZHORN-1982] suggests that 316 SS will satisfactorily withstand corrosion by rubidium.

The possibility of using corrosion-resistant coatings on the interior surface of the primary container is rejected in one report [NAGATA]. Investigations of the possibility of using glass coatings were not carried to conclusion in other work [KLETT-1981]. Data available do not seem to permit definite conclusions on the containment life of packages of Kr-85 stored either as the zeolite or as the sputtered-metal waste form. If the critical factor is the corrosion of the container by rubidium, the corrosion data seem to promise that the integrity of the container is assured for a satisfactory time period. A container lifetime of 50 y (followed by slow release) might accomplish the same result as the proposals of interim storage for 50 y (followed by slow release). Perhaps valve integrity, more than container integrity, will be of greater concern for high-pressure cylinders.

B. Cements as Immobilization Media

A matrix often proposed for the immobilization of radioactive wastes is concrete prepared from portland cement [LOKKEN]. A brief review of the properties of concrete [HELMUTH] is, therefore, appropriate. As a matter of definition, concrete is a mixture of two components: hydraulic cement, which reacts with water to form a paste that eventually sets and hardens; and aggregate, which, when the concrete is used for construction, is a mixture of sand and gravel and, when used for the disposal of radioactive waste, is the solid containing the radionuclide [e.g., for encapsulation of I-129, the aggregate

*A suggested variation of this alternative involves mixing a zeolite powder containing the Kr-85 with the grout to provide additional entrapment [THAMER-1978].

would be $\text{Ba}(\text{IO}_3)_2$ or AgXI^*]. Dry cement mixes obtain their cementitious properties by hydration of the constituents. The reaction of water and the major constituents of portland cement are shown below:

Figure III-4 shows a simplified representation of the setting and hardening of portland cement.

Portland (i.e., hydraulic) cements have been specifically designed for construction of underground, marine, and hydraulic structures and consist mostly of mixtures of tri- and dicalcium silicates, with lesser amounts of tricalcium aluminate and ferrite. Calcium chloride is sometimes used to retard or accelerate setting times, depending on its concentration; calcium sulfate also controls setting times and enhances strength. Fly ash, when added to portland cement, enhances its cementitious properties by reacting with the calcium hydroxide formed during hydration. [The compound $\text{Ca}(\text{OH})_2$ is non-cementitious and can contribute to long-term instability of the concrete.] Standard portland pozzolanic cement, type I-P, is prepared from 80% type I portland cement and 20% fly ash. High-alumina cements, as the name implies, have higher than typical concentrations of Al_2O_3 . Although the hydration product of the high-alumina cements, aluminum hydroxide [like $\text{Ca}(\text{OH})_2$] is non-cementitious, its presence [unlike $\text{Ca}(\text{OH})_2$] appears to have no effect on the overall, long-term stability of concrete.

The choice of which portland cement to use is based on the special properties of each type of cement, which are as follows:

- Type I: when special properties are not required.
- Type II: for general use, especially when moderate sulfate resistance or moderate heat of hydration is required.
- Type III: for high early strength.
- Type IV: for low heat of hydration.
- Type V: for high sulfate resistance.

Table III-5 shows the chemical composition of some typical portland and high-alumina cements expressed as percentages of various metallic oxides. Table III-6 provides the same basic information in terms of potential compound composition of these cements.

Concretes exposed to seawater, sulfate-containing groundwater, or sewage require sulfate resistance; so, it would seem, would concretes designed for long-term storage of radioactive wastes, especially for ocean disposal.

*Where AgX is a notation for silver-exchanged faujasite.

Fig. III-4. Four Stages in the Setting and Hardening of Portland Cement (simplified representation of the sequence of changes): (a) Unreacted clinker grains disperse in water; (b) after a few minutes, hydration products eat into and grow out from the surface of each grain; (c) after a few hours, the coatings of different clinker grains have begun to join up, the gel thus becoming continuous (setting); (d) after a few days, further development of the gel has occurred (hardening). Courtesy of Academic Press Inc. (London) Ltd. [HELMUTH].

Table III-5. Chemical Composition of Some Typical Cements (expressed as percentages of metallic oxides) [HELMUTH]

	Composition, wt %						Loss, wt %	Insoluble residue, wt %
	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	SO ₃		
Type I	20.9	5.2	2.3	64.0	2.8	2.9	1.0	0.2
Type II	21.7	4.7	3.6	63.6	2.9	2.4	0.8	0.4
Type III	21.3	5.1	2.3	64.9	3.0	3.1	0.8	0.2
Type IV	24.3	4.3	4.1	62.3	1.8	1.9	0.9	0.2
Type V	25.0	3.4	2.8	64.4	1.9	1.6	0.9	0.2
High alumina	5.3	39.8	14.6	33.5	1.3	0.4	0	4.8

Table III-6. Composition of Some Typical Cements^a
in Terms of the Potential Compounds
Formed in the Mix [HELMUTH]

	Compound, ^b wt %			
	C ₃ S	C ₂ S	C ₃ A	C ₄ AF
Type I	55	19	10	7
Type II	51	24	6	11
Type III	56	19	10	7
Type IV	28	49	4	12
Type V	38	43	4	9

^a Calculated by the American Society for Testing and Materials.

^b C₃S = (CaO)₃(SiO₂);
 C₂S = (CaO)₂(SiO₂);
 C₃A = (CaO)₃(Al₂O₃);
 C₄AF = (CaO)₄(Al₂O₃)(Fe₂O₃).

The density of cured portland cements is 2 g/cm³ [BAUMEISTER]. Since there should be very little interaction between the cement and the aggregate of the concrete used for waste disposal, the density of the concrete can be arrived at by use of weight percent concentration of each constituent and its density. For example, for a concrete containing Ba(IO₃)₂·H₂O with 9.05 wt % I [17.7% Ba(IO₃)₂·H₂O, density = 4.66 g/cm³], the density of the concrete (weight/volume) would be:

$$\text{density concrete} = \frac{\text{lg concrete}}{\sum (\text{wt fraction constit.} / \text{density constit.})} \quad (\text{III-3})$$

$$= \frac{\text{lg concrete}}{(0.177 \text{ g Ba(IO}_3)_2 \cdot \text{H}_2\text{O}) \frac{1 \text{ cm}^3 \text{ Ba(IO}_3)_2 \cdot \text{H}_2\text{O}}{4.66 \text{ g Ba(IO}_3)_2 \cdot \text{H}_2\text{O}} + (0.823 \text{ g cement}) \frac{1 \text{ cm}^3 \text{ cement}}{2.0 \text{ g cement}}} \quad (\text{III-4})$$

$$= 2.2 \text{ g/cm}^3 \quad (\text{III-5})$$

The addition of an aggregate that has little physical integrity of its own tends to weaken a concrete. This effect is illustrated by the drop in compressive strength of concretes with the addition of increasing amounts of three sludges (10-40 wt %) that were made to simulate the high-level waste of SRP. These data are presented in Table III-7. It is important to note

Table III-7. Compressive Strength of Concrete Waste Forms [LOKKEN]

Sludge Type ^b	Wt %	Compressive Strength, psi ^a					
		I	II	III	V	I-P	HAC ^c
	0	10 824	11 284	13 478	11 898	11 916	9 311
A	10	8 402	8 243	8 694	8 829	8 296	9 574
	25	4 588	4 630	6 180	5 620	4 472	5 792
	40	464	1 259	3 054	3 054	2 380	4 363
B	10	8 973	9 045	9 321	11 159	7 692	9 624
	25	5 779	6 412	7 230	7 158	5 855	7 158
	40	3 932	3 352	4 736	4 234	3 311	5 884
C	10	9 313	7 557	7 603	8 490	7 761	8 465
	25	5 171	4 627	5 817	4 732	4 930	6 658
	40	2 388	2 884	3 317	2 700	3 088	3 371

^a1 psi = 6.9 kPa.

^bType A = 50 mol % Fe(OH)₃ and 50 mol % Al(OH)₃;
Type B = 40 mol % Fe(OH)₃, 40 mol % Al(OH)₃, and 20 mol % HgO;
Type C = 50 mol % Fe(OH)₃ and 50 mol % MnO.

^cHigh-alumina cement.

that the different sludge compositions affected each concrete differently; this finding is an indication that the sludge components reacted chemically with the cement components. Thus, the application of these data to a $\text{Ba}(\text{IO}_3)_2$ waste form is risky.

The mobility and permeability of water and ionic species are extremely low in concrete. The reasons for this are explainable by the hydration reaction that forms concrete. As concrete begins to gel, the originally continuous pore space becomes a system of discrete capillary cavities surrounded by a continuous gel. Inside the gel formation are extremely small pores (~ 3 nm) in which water is trapped. The water trapped inside these pores is greatly affected by the matrix and is, therefore, extremely viscous and tightly bound to the pore walls. The special state of this water is the cause of the extremely low permeability of concrete.

Cements have been studied by many sites as an immobilization medium for low-level wastes. In a survey report, Lokken [LOKKEN] reviews the effects of various parameters on physical properties related to the use of concrete for high-level wastes. The conclusions underline the attributes which make cement a candidate for low-level waste, *i.e.*, low cost, low processing temperatures, processing simplicity, and the requirement for low levels of decay heat and radioactivity to preserve the chemical properties and structural integrity of the monolith. A good general review for understanding the properties of portland cements is provided by Helmuth *et al.* [HELMUTH].

C. Technology of Tritium Waste Forms

Methods for the collection and immobilization of tritium are discussed below.

1. Decay Characteristics of Tritium

The radioactive half-life of H-3 is 12.35 y, and decay occurs strictly by β^- emission. These β^- particles have an average energy of 5.71 keV and maximum energy of 18.6 keV. This decay produces a thermal output of 3.39×10^{-5} W/Ci = 3.26×10^{-1} W/g H-3. Tritium's specific activity of 9.60×10^3 Ci/g will be substantially diluted (by a factor of 10^{-6} to 10^{-10}) in the water collected from a reprocessing plant's off-gas streams.

2. Collection of Tritium

Before discussion of the collection of tritium, it must be mentioned that tritium arising from activities related to fission may never be collected. Several assessments of the environmental significance of the tritium produced in fission-related activities have been made, some implying that collection may not be economically justified [BROWN-1976, COHEN]. This report does not attempt to resolve that question; it is concerned only with characterizing

two tritium waste forms and a few alternatives that would be used if future regulations make collection and disposal of fission-produced tritium necessary.

The EPA has proposed no ruling specifically addressed to tritium produced in the fission fuel cycle that would be analogous to the rulings on krypton and iodine [40 CFR 190].

If tritium is to be collected from fission fuels, the form in which it is available for collection will depend on the type of reactor. In an LMFBR fuel cycle, it will be available mainly as sodium tritide (NaT) in the cold traps used to continually purify the coolant [RENNER]. In an HTGR fuel cycle, a major fraction is expected to appear as water vapor in the off-gas, which is generated from burning of carbon that is part of the carbide fuels; a smaller fraction will appear in the LLAW from solvent extraction. In conventional Purex processing of LWR fuel, most of the tritium is expected to be available in two forms: tritiated water and discarded cladding hulls. Small fractions of the total amount may be available in elemental form in the off-gas from fuel shearing and dissolution. If recent observations are correct [HENRICH], the amounts of elemental tritium may be so small that installation of a process that would convert the element to water vapor by catalyzed oxidation and combine this vapor with the tritiated water from subsequent operations may not be justified.

Characterization of the distribution of forms and quantities of tritium in a plant that is reprocessing LWR fuel have varied widely [ALTOMARE, PHILLIPS]. Early observations suggested that most of the tritium in LWR fuel would appear in low-level aqueous waste of FRP [KULLEN]. A recent observation [HENRICH] of the distribution of tritium in irradiated PWR fuel, however, suggests that about 60% is associated with the zircaloy cladding and most of the remaining 40% is associated with the fuel matrix. Only the latter portion would appear in the LLAW.

Tritium bound in zircaloy cladding hulls is considered to be a form suitable for disposal (see Section III.C.3.c.1.). There are several options for collecting tritium from the fuel matrix: voloxidation or thermal outgassing as a part of the head-end of the processing flowsheet, water recycle to reduce the volume of tritiated aqueous waste, tritium scrub of solvent, or hydrogen isotope separation.

The tritium-removal head-end operations most seriously considered are voloxidation and thermal outgassing. The voloxidation operation, extensively investigated by ORNL, involves roasting chopped fuel in the presence of air or oxygen at 450°C to drive the airborne waste out of the fuel. If this operation is used, the tritium in the fuel matrix would be collected as tritiated water on a desiccant. Although voloxidation has been investigated extensively, it cannot be regarded as available technology; in fact, there are doubts that it will prove to be technologically feasible [MCKAY].

Thermal outgassing involves heating chopped fuel at 1100-1500°C in a vacuum or a helium stream to drive off the airborne wastes. The collection of tritium by this process has not been thoroughly characterized, but experimental demonstrations are now in progress [BURGER-1982A, BRAY]. Both processes endeavor to collect the tritium in a small volume so that dispersal is avoided in the large volume of aqueous solution used for fuel dissolution.

If tritium collection is to be practiced without a head-end tritium-removal operation, it is generally agreed [HALL, HENRICH, BURGER-1978, MCKAY] that the aqueous plant fluids must be recycled to limit the volume of water containing the tritium from the fuel matrix. According to [MCKAY] a ten-fold reduction in volume of tritiated aqueous FRP wastes could be obtained by recycle. It is asserted [MCKAY, BRUGGEMAN-1980], however, that even with recycle, the volume of tritiated LLAW would be too great to solidify directly and package for terminal storage. Accordingly, the volume of recycled aqueous waste would, in some schemes, be reduced by a factor of 10 through a tritium scrub of solvent [MCKAY, HENRICH] or by a factor of 10^4 or more through separation of hydrogen isotopes. Although several isotope-separation processes have been suggested for this purpose, the process that is currently favored by several organizations is the catalyzed exchange of hydrogen isotopes between hydrogen gas and liquid water. This process is being developed in laboratories in the U.S. [ELLIS], Canada [BUTLER], and Belgium [BRUGGEMAN-1980]. The same reaction, although involving hydrogen gas and steam to prepare water enriched in deuterium [BENEDICT-1955], was employed on a large scale at the Trail plant in British Columbia in the early 1940s. The technology of this early work has been greatly improved by recent introduction of a hydrophobic catalyst that permits direct contact of liquid water with the catalyst. Demonstrations have involved a stream of elemental, gaseous H-T mixtures flowing counter-current to a water stream running down through a packed bed of solids bearing the catalyst. An electrolytic cell at the bottom of the column generates the elemental form. Some concepts for large-scale application have proposed that a catalytic recombiner at the top of the column would regenerate the aqueous form [BUTLER, BRUGGEMAN-1980], but experimental setups have not included such a device [ELLIS]. In application to the tritiated liquid waste from FRP, water enriched in tritium could be bled off from the bottom of the electrolysis cell for disposal as tritiated concrete. One conceptual application to FRP [ELLIS] involved bleeding off tritium-enriched hydrogen from the electrolysis cell for disposal as a metallic hydride.

A diagram relating these process options is shown in Fig. III-5. Even though voloxidation and the isotope separation by hydrogen-water exchange over a hydrophobic catalyst cannot be considered proven and available technology, they are at an advanced stage of development compared to thermal outgassing. It seems likely, at any rate, that most of the tritium in irradiated fuels of the LWR type will be collected in cladding hulls and in tritiated water. Disposal of the tritium collected in water is the object of the following review of tritium-immobilization technology.

Fig. III-5. Proposed Tritium Collection Operations in Aqueous Fuel Reprocessing

3. Immobilization of Tritium

a. Immobilization of Tritium in Concrete

The incorporation of tritiated water into concrete has been investigated by several laboratories during the past decade. Observations of the characteristics of this waste form have been concerned mainly with release of tritium by vaporization and by leaching. Another property of tritium-containing concrete that has been studied is the rate of gas formation from radiolysis [WIENEKE-1979, BIBLER].

Leach tests have been made on samples in which variations were made in coating type, cement type, volume/surface ratio, packaging, cement additives, water/cement ratio,* and curing technique. Several additives to increase water loadings have been tried also, including silica gel, vermiculite, and plaster of paris. The comparison of results from different investigators is difficult because of differences in test conditions and expression of results. Nevertheless, the effect of some parameters is so great that general conclusions are obvious. It has been concluded [EMELITY, MCKAY, MATSUZURU] that cement alone is an unsatisfactory matrix for tritium retention. Satisfactory performance of cement forms in leach tests seems to have been obtained only by the aid of a combination of coating and packaging [MERSHAD-1977A, -1977B,].

The difficulty of retaining tritiated water in solid waste forms is indicated by some observations on water exchange rates. Water exchange rates for aqueous metal ions are extremely dependent on the metal ion, varying over 16 orders of magnitude at 25°C. The highest rate of water exchange has been measured for alkali metals ($\sim 10^9 \text{ s}^{-1}$) and the lowest rate measured for Rh^{3+} ($4 \times 10^{-8} \text{ s}^{-1}$). The rates for other metal ions, for example Cr^{3+} ($3 \times 10^{-6} \text{ s}^{-1}$), Co^{3+} ($\sim 10^{-1} \text{ s}^{-1}$), Al^{3+} ($\sim 10^1 \text{ s}^{-1}$), Fe^{3+} ($\sim 10^2 \text{ s}^{-1}$), V^{3+} and VO^{2+} ($\sim 10^3 \text{ s}^{-1}$), Ni^{2+} ($\sim 10^4 \text{ s}^{-1}$), Mg^{2+} ($\sim 10^5 \text{ s}^{-1}$), Mn^{3+} ($\sim 10^6 \text{ s}^{-1}$), and the lanthanides (10^7 - 10^8 s^{-1}), fall in between these extremes [BASOLO]. The exchange rate of tritiated water (HTO), however, is not affected by highly variable and relatively low substitution rates. Substitution of tritiated and natural waters occurs by a much faster path--hydrogen-ion-substitution reactions through the making and breaking of hydrogen-bonded water polymers.

The rate constants of the two reactions

have been estimated by nuclear magnetic resonance (NMR) techniques to be: $k_1 = 8.2 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$ and $k_2 = 4.6 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$. Averaging these two second-order rate constants and assuming unit activity for water gives an estimated first-order rate constant for the reaction

of $k_3 = 6 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$. This rate is higher than can be achieved by any water substitution reaction [GLASEL].

The point of the above discussion is that no chemical mechanism can successfully protect against natural-water/tritiated-water substitution. Tightly binding tritiated water to a chemical matrix will be completely ineffective because of the hydrogen ion transfer mechanism. If a ready diffusion path exists for the transfer of water in and out of the matrix, tritium ion transfer will occur.

*The water/cement ratio must be in the range 0.3-0.8 (weight). Below this range, the solid has insufficient strength, and above this ratio a layer of unincorporated water will exist on the solid.

Cement retains water by mechanisms that are complex and not well understood. Furthermore, the rates of exchange between water contained in cement and water in contact with the cement do not seem to be very well understood [MCKAY]. In portland cement, water seems to be held in several different physical and chemical forms that are not easily distinguished. Some of the hydration reactions seem to take place over a long period of time, perhaps years. For practical purposes, the water associated with portland cement can be roughly classified into evaporable and nonevaporable fractions. A block of cement has a characteristic water vapor pressure, which diminishes with curing. At relative humidities that provide a partial pressure higher than this characteristic pressure, the block will absorb water; at relative humidities that provide a lower partial pressure, it will give up water.

The development of a tritiated concrete waste form has included measurements of leach rates on (1) simple tritiated cement with and without several types of coatings [EMELITY, MATSUZURU], (2) polymer impregnated tritiated concrete (PITC) [COLUMBO-TSDPR], (3) PITC with and without a polyethylene container [WIENEKE-1979], and (4) a cement-plaster combination contained in a multi-walled package [MERSHAD-1977A, -1977B]. Only the last waste form, which will be referred to as the Mound Package, has been shown to have an acceptably low leach rate and has been accepted at a waste-disposal facility (the Nevada Test Site).

The coatings investigated for their ability to aid the retention of tritium in concrete [EMELITY] included asphalt, paints, greases, plastics and waxes. Such coatings reduced the amount of tritium leached in the first year from about 60% for uncoated concrete to at best about 5%; nevertheless, this simple waste form (blocks of coated concrete) was judged to have an unacceptably high leach rate. A suggested coating [MCKAY] is a layer of cement formed by pouring tritiated cement into a pre-formed cement box; it is asserted that the evaporable water could not be removed from such a package, but this concept has apparently never been tested.

The retention of tritium by PITC has been investigated rather extensively at BNL [COLUMBO-TSDPR], including collaborative observations with workers at Mound and Savannah River Laboratories. The polymer, usually polystyrene, fills the pores in cement, thereby reducing diffusion of water. The final report of the tests at Mound Laboratory [WIENEKE-1979] showed that the amount of tritium leached by water in the first year was about 1% for PITC samples contained in polyethylene jackets, but was much higher, 20-50%, for non-jacketed samples. All of the leach rates of polymer-impregnated cements, with and without polyethylene jackets, were considered to be unacceptably high.*

The waste form that is currently considered acceptable is the Mound Package--a combination of cement, nested containers, and sealants. The Mound Package was developed specifically to provide disposal for the tritiated wastes arising at the Mound Laboratory, some occurring as tritiated water. There seems to be no obvious reason why this package can not be applied to

*Details of the criteria for acceptability are not given; presumably they were determined by negotiations between Mound Laboratory and the disposal facility to which Mound proposed to ship the waste.

disposing of tritiated water from a fuel reprocessing plant, reduced in volume by one or more of the processes described in Section III.C.2.

The procedure for preparation of the Mound Package is as follows: the polyethylene primary container is loaded with 81 kg of a 1/3 (volume) mixture of cement/plaster. The polyethylene primary container has a capacity of 27 gal (100 L). About 27 L of the aqueous waste is then added to the cement-plaster mix. The lid of the polyethylene primary container is sealed with RTV sealant. The polyethylene container, after being placed in an asphalt-coated 30-gal steel drum, is completely covered with non-hardening asphalt. The lid of the 30-gal drum is then sealed with RTV sealant and a bolted ring. The sealed 30-gal drum is centered on a layer of about 4 gal of asphalt in a 55-gal steel drum. The annulus between the two drums is then filled with vermiculite to within a level of about 2 in. below the top of the 30-gal drum. The remaining void is then filled with asphalt, and the lid is placed on the 55-gal drum and sealed with RTV sealant and a bolted ring. Details related to this concept are summarized in Table III-8. A simple cross-sectional diagram of the Mound Package is shown in Fig. III-6.

Table III-8. Characterization of the Mound Package for Disposal of Tritium in Cement

Package^a	
Primary:	Polyethylene drum (27 gal)
Secondary:	Asphalt-coated 30-gal steel drum (DOT 17H)
Tertiary:	55-gal steel drum (DOT 17H)
Waste Form:^a Cement/plaster mix, 1:3 volume	
Weights^a	
Cement-plaster:	81 kg
Water:	27 kg
Vermiculite:	10 kg
Tritium Release Rate: 0.002% in 87 y (7 half-lives, extrapolated from experimental observations)^a; 10⁻³% after 400 weeks^b	

^a[MERSHAD-1977A, -1977B].

^b[LAMBERGER].

Since this Mound Package is the only currently practical technology for disposal of tritiated water in cement, it is considered here as the reference concept. To obtain an even lower release rate, the Mound Laboratory had proposed using PITC rather than the cement-plaster mixture in the Mound Package, but at present nothing other than the cement-plaster form has been used in the actual disposal of wastes [WIENEKE-1982]. Equipment has been assembled at the Mound Facility to immobilize their tritiated wastes [CHRISTIAN]. The drums containing solidified Mound wastes are overpacked by placing 13 or 14

Fig. III-6. Cross Section of the Mound Package for Disposal of Tritiated Waste in a Cement-Plaster Matrix [MERSHAD-1977A]

of them in a container that had been previously used to ship a jet engine in an inert atmosphere. The container is welded shut and sent for SLB [BLAUVELT].

b. Immobilization of Tritium on Molecular Sieves

Several proposals have been described for trapping tritium, either as the element [LAKNER, TURI] or as tritiated water [BROOThAERTS, HOLLAND, MCKAY], on molecular sieves. Molecular sieves are commercially available in the forms of large particles, cylindrical pellets, and spherical beads. It has been suggested that a desiccant for tritiated water storage should have the following properties [MCKAY]: (1) low water vapor pressure, (2) high water content, (3) low solubility, (4) high thermal and radiolytic stability, and (5) low cost. Much of the reported experimental work on reaction of tritium with molecular sieves is concerned not with retention, release, or leakage from the solid to the environment, but with demonstrations of cleanup of gas streams [DELDEBBIO] or with the capacity of the solid for incorporating tritium, hydrogen, or water. The capacity of Type A zeolite for water is 22 wt % when fully saturated as $\text{Na}_2\text{O} \cdot \text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2 \cdot 4.5\text{H}_2\text{O}$ [MCKAY].

There is much evidence that adsorption by molecular sieves offers an effective collection process for tritium, either as the element or as water. Unanswered, however, is the question of whether molecular sieves also offer a matrix that holds tritium without rapid leakage to the environment. The rate of exchange of hydrogen between water in the adsorbed state and water in the environment is thought to be the primary determinant of the technological feasibility of retention of tritium by a desiccant [MCKAY]. A low vapor pressure of water seems to be a necessary, but not sufficient, condition for tritium retention. It is suggested that for tritium retention the

vapor pressure of water held by the desiccant must be no greater than 10^{-4} bar (10 Pa) [MCKAY]. Exchange is thought to be only "loosely related to the vapor pressure," i.e., a low vapor pressure may not indicate a low exchange rate. Equilibrium constants for partitioning of tritium between water on molecular sieves and water in the environment have been measured [ONO]. The results showed that at equilibrium the thermodynamic (i.e., equilibrium) tendencies in this system favor the location of tritiated water on the molecular sieves rather than in the environment surrounding the sieves. The results of this study, however, do not indicate the rate of release of tritiated water adsorbed on molecular sieves. An observation of the rate of appearance of tritium in the environment of a sample of tritiated water held on molecular sieves is needed. Even if they do not provide retention, molecular sieves could be used as a medium for collection and localization of tritium, and some efficient means of packaging, perhaps similar to the Mound Package, would bear the burden of isolating the tritium from the environment.

In the preparation of a tritiated molecular-sieve waste form, McKay [MCKAY] has recommended that water vapor be contacted with the solid, rather than the solid being immersed in liquid water. One of the possible hazards of manipulation of molecular sieves loaded with tritium is the inhalation of tritium-bearing dust. For this reason, tritium-loaded molecular sieves might best be compressed into monolithic forms, by a process similar to the one proposed for dealing with krypton-loaded molecular sieves [BENEDICT-1979, CHRISTENSEN]. Several schemes involve conversion, by catalyzed oxidation, of elemental tritium to HTO for subsequent adsorption on molecular sieves [BROOThAERTS, BRUGGEMAN-1980, WITTENBERG, MULLER, BIXEL, SHERWOOD].

Elemental tritium has a toxicity of about 1/30,000 that of aqueous tritium. Once released to the environment, it converts to aqueous tritium with a conversion time of 6 to 10 y [MASON-1977, -1979].

Encapsulation of the elemental form of tritium by zeolites requires elevated temperatures (350-450°C) and rather high pressures (7-14 × 10^4 kPa) [LAKNER, FRAENKEL, TURI]. These requirements are disadvantages compared to adsorption of tritiated water vapor onto sieves at ambient conditions. Encapsulation of the elemental form at these high temperatures and pressures would be advantageous if it produces a tritium-encapsulated solid with a much lower leak or exchange rate than that for encapsulated tritiated water. It would have the additional advantage of containing tritium in a less hazardous form. Considering the relative toxicities of elemental and aqueous tritium (given above), a much higher leak rate of elemental tritium encapsulated by zeolites could be tolerated. Some hydrogen molecular-sieve systems may have acceptable thermal stability for applications such as short-term hydrogen fuel storage, but they lack promise for long-term retention of tritium without the aid of secondary containment. For example, at 25°C, potassium A-type zeolite lost 46% of its hydrogen load in 45 h, and cesium A-type zeolite lost 7% of its hydrogen load in 5 d [FRAENKEL]. In another test [TURI], about 3% of the hydrogen that had been loaded onto a cesium zeolite was released in 18 d at room temperature. As a test of the retention of hydrogen, this observation [TURI] was flawed because the gas released included

not only the hydrogen encapsulated in microcavities of the zeolite, but also the hydrogen adsorbed onto surface sites. A thorough assessment of the retention of elemental tritium of hydrogen by zeolites requires more detailed experimentation, but the general trend does not seem favorable for disposal application.

Many of the reported schemes involving molecular sieves do not include disposal of them. Rather, the sieves are used only for collection of tritium and are then regenerated for further tritium collection [MULLER, WITTENBERG]; the tritium is removed by heating the sieves and disposed of in some other, unspecified way. If, at saturation, the tritiated water held on molecular sieves is 20-22 wt %, storage of the entire dilute LLAW stream of a Purex reprocessing plant in this manner would involve unacceptably large volumes. Therefore, tritiated water stored on molecular sieves would have to be the product of one or more of the volume-reduction processes discussed in Section III. C.2, i.e., recycle, tritium scrubbing, voloxidation, or isotopic exchange.

Thus, a good deal of attention has been given to use of molecular sieves for their properties in cleanup of gas streams, i.e., their performance as getter for either hydrogen or water and also for their capacity, i.e., the quantity of H_2 or H_2O held by the sieves. Few observations, however, seem to indicate whether they would also perform well as a long-term retainer of tritium. Only experimental work on exchange of tritium between molecular sieves and environmental water will answer that question. A review [MCKAY] of the storage of tritium on solids suggests the following order of merit for the various candidates for matrix materials: (1) inorganic hydrates,* (2) metal hydrides, (3) organic polymers, and (4) cement.

c. Alternative Tritium Immobilization Technologies

(1) Zirconium Hydride

The capability of several transition metals for reacting readily with hydrogen to form stable hydride compounds [MUELLER] and the availability of waste zirconium metal in the LWR fuel cycle[†] have led to the suggestion that zirconium hydride be used as a waste form for tritium. Investigations of the feasibility of this concept have included experimental observations of suitable conditions for the reaction of zircaloy tubing with hydrogen [SCHEELE-1976] and also of the leaching behavior of zirconium hydride [COLOMBO-1977].

Reaction conditions consisting of a temperature of 630°C and 1 atm pressure were found capable of producing a high loading of hydrogen in solid zirconium (approaching $Zr/H = 2$) at a practicable rate [SCHEELE-1976].

*A category in which molecular sieves is included in this author's discussion.

[†]Zircaloy hulls contain ~98% zirconium.

The quantities of zirconium required to store the tritium from fuel reprocessing will depend on the amount of associated hydrogen and oxygen. As in the case of most other tritium-storage schemes, the zirconium hydride concept is feasible only if the volume of tritiated waste water is minimized. Thus, it would best be applied to the product of one of the volume reduction schemes described in Section III.C.2. For a product with H/Zr ratio of 2, the quantities of zirconium necessary to hold the hydrogen-tritium mixtures from fuel reprocessing have been estimated, as shown in Table III-9. Comparison of the required quantities of zirconium in this table with the estimated quantities of cladding hulls (3.7×10^5 kg/1500 MTHM, see Table III-9) suggests that, if cladding hulls are to be the sole source of zirconium, then the volume reductions obtainable through voloxidation or through recycle plus isotope separation are necessary.

The results of a limited series of leach tests on zirconium hydride samples are briefly summarized in Table III-10. These results indicate roughly that the leach rate of zirconium hydride is small; however, the reports upon which these results are based lack sufficient details concerning the effects of several parameters that generally affect leach rates.

Several considerations have to be taken into account in an evaluation of the feasibility of storage of tritium as the zirconium hydride. One advantage suggested for this mode of storage is the relatively small space required [SCHEELE-1976]. In addition, it is believed that this waste form would satisfy requirements of noncombustibility [ALTOMARE], although details of its behavior as a function of temperature and gaseous environment have yet to be investigated at realistic conditions. One of the disadvantages of metallic hydrides as a tritium waste form is the necessity for converting all tritium and associated hydrogen to the elemental form. To make this scheme practicable, the tritium would have to be isolated before dispersion in dissolver solution. Another disadvantage is the need to have a hydrogen-tritium mixture free of other contaminants [SCHEELE-1976], e.g., oxygen. If zircaloy hulls are used to fix the tritium, the radioactive properties of the neutron-activation products associated with the hulls are likely to determine the packaging necessary to meet transportation regulations and acceptance criteria at a disposal facility, e.g., SLB facility.

(2) Organic Compounds

The incorporation of tritium into organic compounds has been investigated experimentally [FRANZ-1975, -1976; COLOMBO-1974], and evaluated at some length [MCKAY]. Several candidate compounds have been suggested [MCKAY], and experimental work has been done on six. These compounds included polyacetylene [COLOMBO-1974], tritiated Bakelite, polyacrylonitrile, polystyrene, polyureylene-polymethane copolymer [FRANZ-1975, -1976], and polyvinyl chloride [DIAMOND]. The preparation of polystyrene would require the conversion of tritiated aqueous waste to hydrogen, which would be reacted with polystyrene in a solvent in the presence of a catalyst but in the absence of oxygen; the latter step is taken to discriminate against formation of T₂O. The preparation of the other compounds, however, could be carried out by direct reaction of tritiated water. Results of tests of the thermal stability of several of these polymers, summarized by [MCKAY] in Table III-11, showed that

Table III-9. Quantities of Zirconium Required to Fix Tritiated Aqueous Waste from Fuel Reprocessing as ZrH_2

Volume-Reduction Scheme	Volume of Tritiated Water, L/1500 MTHM	Quantity of Zirconium Required, ^a kg/1500 MTHM
High (~90%) Water Recycle ^b	3800	2.0×10^7
Isotope Separation with Water Recycle ^b	86.5	4.4×10^5
Voloxidation with Water Recycle ^b	14.4	7.3×10^4
Thermal Outgassing ^c	0.56	2.8×10^3

^aFor comparison, the quantity of zirconium expected from reprocessing LWR fuels, at a PWR/BWR ratio of 2/1, is about 3.7×10^5 kg/1500 MTHM. This estimate was derived from data in [DOE/EIS-0046F-4.11].

^bVolume reduction schemes and volumes as described by [GRIMES].

^cVolume from thermal outgassing as conceived by [BRAY].

Table III-10. Results of Leach Tests on Tritiated Zirconium Hydride

Leach Solution	Fraction of Tritium Released During Period ^a	
	6 months	12 months
Distil. H ₂ O	None detected	$(2.3-6) \times 10^{-5}$
HCl (pH=4)	None detected	1.6×10^{-4}
NaOH (pH=10)	Not measured	5.5×10^{-4}

^aSummarized by [MCKAY] from observations reported in [COLOMBO-TSDPR].

Table III-11. Thermal Stability of Tritiated Organic Compounds

Compound	Thermal Degradation Threshold, ^a °C
Bakelite-type	130
Polyacrylonitrile	200
Polyvinylacetate	150
Polyureylene/polyurethane	180
Hydrogenated polystyrene	280

^aFrom compilation in [MCKAY].

they are generally stable up to about 200°C, with varying stages of decomposition beginning at temperatures of about 130°-300°C. The thermal stability of these polymers is probably the main point to be considered in assessing their capability for complying with technical criteria in regulations for transport or for terminal storage. Simple leach tests have shown that the leachability of tritium from these solids is slight [MCKAY]. After an initial exposure to water in which 0 to 5% of the tritium was leached out, the quantity of tritium lost in additional exposure was undetectable in most cases.

Results of leach tests on tritiated polyvinyl chloride [DIAMOND] are not easily compared with the results on the other compounds reported [FRANZ-1975, -1976] and [COLOMBO-1974], but it is clear that leach rates for this compound were quite low; the surface penetration derived from the observations was 70 Å per day. From data in the description of this work [DIAMOND], it is possible to express the leach rate, for a surface/volume ratio of 2-3 cm⁻¹, as a fraction of about $2 \times 10^{-4}/d$.

Information on the radiation stability of several of these compounds were compiled by McKay [MCKAY] as shown in Table III-12. The G values listed in Table III-12 suggest that radiolytic gas formation rates in these compounds are not likely to be great enough to prohibit their use as storage media.

(3) Other Tritium-Management Options

Among the many other tritium-management options that have been suggested [BURGER-1974, -1976], two seem to be regarded as feasible. One is the tank storage of tritiated aqueous waste. This concept has not been developed much beyond enumeration of the probable parameters [BROWN-1980] and preliminary cost estimates [ARNOLD, NEA, GRIMES]. The other option, suggested both in U.S. studies [TREVORROW, PERKINS] and in German planning [HARTMANN], is deep-well injection of tritiated aqueous waste. Neither of these options afford the degree of immobilization obtainable by incorporation of tritium

Table III-12. Radiation Stability of Organic Compounds Proposed as Tritium-Storage Forms

Compound	G Value, ^a ion/100eV
Polyacetylene	0.15 (H ₂ only)
Polyethylene	2.1-3.9
Phenol-formaldehyde resins	0.1
Polyacrylonitrile	0.4
Polyvinylacetate	1.4
Polystyrene	0.08, 0.03

^aFrom compilation in [MCKAY].

into chemical compounds, but promise to be capable of delaying the dispersal of tritium to the environment until the radioactivity associated with the waste has been greatly reduced by decay.

D. Technologies of Iodine Waste Forms

1. Decay Characteristics of I-129-Containing Waste Forms

Iodine-129 has a half-life of radioactive decay of 1.57×10^7 y by β^- and γ decay of the energies shown in Table III-13. Data, also presented in this table, show that the thermal output of ^{129}I is almost equally divided between β^- and a series of low energy x-rays. Table III-14 gives the composition of iodine isotopes expected to be present in the blended fuel of an LMFBF from time of discharge to 10 y after discharge. One year after discharge, there are significant concentrations of only two iodine isotopes--stable I-127 (24.6 wt %) and I-129 (75.4 wt %). Table III-15 shows the differences in the amount of these two iodine isotopes produced in different reactor fuels.

Because of its long half-life of radioactive decay, I-129 has a low specific activity, 1.77×10^{-4} Ci/g I. Since the material for disposal is not 100% I-129, but rather ~75% (based on data presented in Table III-14), the specific activity of iodine as the waste form will be only 1.3×10^{-4} Ci/g I.

Based on the specific activity of I-129 and the energies and intensities of its decay modes (Table III-13), the thermal output of I-129 equals 4.62×10^{-4} W/Ci or 8.18×10^{-8} W/g. Based on the fact that reprocessing waste iodine is only 75% I-129, the heat release of the iodine would be 6.1×10^{-8} W/g I.

Table III-13. Radioactive Decay of I-129, Radiation Types with Their Corresponding Energy, Intensity, and Energy Emitted per Disintegration [MARTIN].

Radiation Type	Energy, keV	Intensity, %	$\Delta(\text{g-rad}/\mu\text{Ci-h})^a$
Auger-L	3.43	74 ± 4	0.0054
Conversion Electron-K-1	5.02 ± 0.03	79.10 ± 20	0.0085
Auger-K	24.6	8.8 ± 1.6	0.0046
Conversion Electron-L-1	34.13 ± 0.03	10.6 ± 0.3	0.0077
Conversion Electron-M-1	38.44 ± 0.03	2.10 ± 0.10	0.0017
Conversion Electron-NOP-1	39.37 ± 0.03	0.70 ± 0.10	0.0006
β^- max	150 ± 5		
avg	40 ± 5	100	0.0852
X-ray L	4.1	8.2 ± 2.5	0.0007
X-ray K α_2	29.4580 ± 0.0010	20.0 ± 0.6	0.0126
X-ray K α_1	29.7790 ± 0.0010	37.1 ± 0.9	0.0235
X-ray K β	33.6	13.2 ± 0.4	0.0094
	39.58 ± 0.03	7.50 ± 0.20	0.0063
Total ($\sum \Delta_i$) i	-----	-----	0.1662

^aEnergy emitted per disintegration in units of g-rad/ $\mu\text{Ci-h}$. For an infinite, homogeneous medium in which a source is uniformly dispersed with a concentration of 1 $\mu\text{Ci-h/g}$, Δ gives the absorbed dose in rads. For each radiation type i

$$\Delta_i = 0.00213 I_i \times E_i$$

where I_i = intensity of i and E_i = energy of i. The factor 0.00213 is the conversion factor for keV/disintegration to g-rad/ $\mu\text{Ci-h}$. The equilibrium absorbed dose, Deq , in rads equals $C_{129\text{I}} \times \sum \Delta_i$, where concentration is given in units of $\mu\text{Ci-h/g}$. One g-rad/ $\mu\text{Ci-h} = 2.78 \times 10^{-3}$ W/Ci.

Table III-14. Fission Product Iodine from Spent LMFBR Fuel and Its Decay. (Basis: 1 metric ton of fuel. Calculated by ORIGEN Code [CLARK-1975].)

Isotope	Time after Discharge from Reactor									
	0 d		90 d		150 d		365 d		10 y	
	Ci	g	Ci	g	Ci	g	Ci	g	Ci	g
I-127	0	74.6	0	76.8	0	77.5	0	78.6	0	78.9
I-129	3.90×10^{-2}	239	3.94×10^{-2}	241	3.94×10^{-2}	242	3.94×10^{-2}	242	3.94×10^{-2}	242
I-131	1.40×10^6	11.3	6.16×10^2	4.97×10^{-3}	3.51	2.84×10^{-5}	3.15×10^{-8}	2.54×10^{-15}	0	0
I-132	1.62×10^6	^a	7.40×10^{-3}	^a	2.14×10^{-8}	^a	2.50×10^{-28}	^a	0	0

^aNegligible.

Table III-15. Amounts of Iodine Isotopes Found in 1 MTHM of Different Reactor Fuels One Year After Discharge [CROFF-1980, CLARK-1975].

Reactor Type	Fuel Burnup, MWd/MT	Mass I-127, g	I-129		$\frac{\text{I-129}}{\text{I Total}} \times 100$
			g	Ci	
PWR	33,000	55	179	0.0316	76.5
BWR	27,500	45.6	149.7	0.02644	76.6
LMFBR Core + axial blanket	64,770	201.8	510.2	0.09011	71.7
LMFBR Blended fuel	---	78.6	242	0.0399	75.4

2. Collection of Iodine

Collection processes must be capable of removing several iodine species from process streams, including elemental iodine, inorganic iodine compounds, and organic iodine compounds. The latter are formed from interactions of iodine, nitric acid, solvent, and carbon contained in fuel materials [BURGER-1979A]. Methyl iodide, CH_3I , is the species usually mentioned in discussions of the problem, but the general term, organic iodides, is commonly employed, implying that more than one organic species, unidentified, may be present.

According to Burger et al. [BURGER-1981A], there are three currently favored techniques for removal of iodine from off-gas streams of fuel reprocessing plants: Mercurex, Iodox, and solid sorbents. A fourth process, caustic scrubbing, which has fallen into disfavor chiefly because of its inability to remove organic iodine compounds from the off-gas stream, is still considered by them to be a viable process. These methods are discussed in detail by several reviewers [BOND, BROWN-1980, BURGER-1979A, -1981A, HOLLADAY-1979, and MAILEN]. A description of each process based on these reviews follows.

a. Mercurex

The mercury nitrate-nitric acid scrubbing process has been demonstrated in both packed and bubble-cap columns and is capable of removing both inorganic and organic iodides from LWR-LMFBR fuel reprocessing off-gases. Adequate residence time must be provided to decompose organic iodide species; improved organic iodides removal is associated with higher nitric acid concentrations [BURGER-1979B]. Organic iodides react to form HgI_2 and organic nitrates or alcohols. The overall efficiency of the process is controlled by

the extent of bleed-off of the accumulated I^- and IO_3^- from the recycle solution.

The chemical reactions that are operative in this process are:

and

or

The sorption of NO_x from the off-gas stream, which forms nitrous acid, inhibits formation of $Hg(IO_3)_2$. Because of the low solubility of mercuric iodate in nitric acid solution, this sorption is advantageous to the process.

Some work has been performed to study the conversion of the aqueous HgI_2 product of the Mercurex process to $Ba(IO_3)_2$, a compound that is a currently acceptable chemical form for disposal in concrete [ROGERS]. This conversion also fulfills three other important requirements: (1) separating the iodine species from the scrubbing process, (2) reducing the mercury requirements of the process, and (3) eliminating the disposal of large quantities of mercury with the radioactive iodine. The conceptual plan, not yet fully developed, is (1) electrolytic [BURGER-1979B] or nitric acid [PARTRIDGE] oxidation of mercuric iodide to produce insoluble mercuric iodate, (2) contact of the mercuric iodate with sodium hydroxide to produce mercuric oxide and sodium iodate, and (3) precipitation of barium iodate from the reaction of aqueous sodium iodate with barium hydroxide. The nitric acid oxidation is effectual only at selected conditions [PARTRIDGE]. (The HgO product of the reaction of $Hg(IO_3)_2$ and $NaOH$ would be reacted with nitric acid to produce $Hg(NO_3)_2$ for reuse in the scrubbing process.) These steps, which most likely would be carried out in a batch mode, are slow, and the separation of the solid products would be difficult. Another disadvantage of this process is that aromatics in the feed gas greatly diminish the iodine DF [HOLLADAY-1979]. An alternative, in development at the Mol, Belgium plant, is electrolytic reduction to metallic mercury and precipitation of CuI , requiring a reductant for the I_2 that is formed [BURGER-1982B].

Advantages for this process are that I_2 and CH_3I can be removed simultaneously with appreciable DF's, and this removal is relatively unaffected by the presence of water vapor or trace amounts of dodecane or octane. Also, the presence of NO_x in the off-gas stream actually benefits the process by preventing the formation of the insoluble $Hg(IO_3)_2$. Because the concentration of nitric acid is subazeotropic (2-14 M), the scrubber equipment can be constructed of stainless steel, and no special in-plant equipment to produce hyperazeotropic HNO_3 is necessary. (Both of these points are advantages over the Iodex process, discussed next.) Retention factors of $>10^3$ have been

obtained in laboratory tests of the Mercurex process, but the full-scale process designed for the Allied-General Nuclear Services reprocessing plant claims retention factors of only 10-75; this process alone is therefore probably not adequate for process application, but may be sufficient if backed up by solid sorbents [BURGER-1979A].

b. Iodox

The Iodox process is a wet scrubbing system in which the off-gas stream of a fuel reprocessing plant is contacted by 20-22 M HNO₃ (hyperazeotropic nitric acid) in a bubble-cap scrubber. The reaction scheme for iodine removal is the sequence:

Reaction III-12 is fast and reaction III-13 is rate-determining. For aliphatic iodide compounds, there is an initial fast nitration reaction:

The oxidation of I₂ then follows reactions III-12 and -13. The acidic iodate can be concentrated to form HI₃O₈ or precipitated as the slightly soluble Ba(IO₃)₂, although this step is untested.

Iodox is capable of retention factors of >10⁴ and is probably adequate to meet the requirements of 40 CFR 190* [BURGER-1979A]. The major disadvantage to the Iodox process is the high capital cost associated with building its equipment from acid-resistant materials such as zirconium or titanium; the hyperazeotropic nitric acid would also need to be produced in-plant. If fluorides are introduced in the reprocessing flowsheet (e.g., in processing denatured thorium-uranium fuel in the Zirflex process), corrosion problems would be more severe. Another disadvantage is the potential for accumulating explosive organic nitrates in the Iodox scrubber.

There are two major advantages to the Iodox process: (1) no new chemicals are added to the reprocessing plant operation, and (2) the product of the process, HI₃O₈, is easily converted to Ba(IO₃)₂, a preferred disposal form.

c. Solid Sorbents

Silver-coated alumina and alumina-silica combinations yield iodine-retention factors of >10³, but there are occasional, unexplained reductions in iodine-retention factors in process conditions [BURGER-1979A].

* Pertinent sections of the Code of Federal Regulations are referenced at end of this report.

Silver-exchanged zeolites and silver-nitrate-impregnated amorphous materials, on the other hand, are becoming more highly regarded as means for collecting radioactive iodine from a fuel reprocessing plant's dissolver off-gas. Commercially available materials are a silver-exchanged zeolite, silver-exchanged faujasite (AgX), silver-exchanged mordenite (AgZ), and silver-nitrate-impregnated amorphous silicic acid (AC-6120). Silver-exchanged zeolites appear to trap iodine by at least two mechanisms: physical sorption and chemical reaction to form silver iodide. A simplified chemical equation for the reaction would be:

It is speculated that the reaction of aliphatic iodides is:

The reason for suspecting at least two mechanisms for the retention of iodine is that there appear to be two types of iodine species--one tightly bound and therefore difficult to remove, the other easily displaced. According to [JUBIN], a typical loading for AgX at 150°C would be 349 mg I₂/g AgX; 214 mg/g AgX is chemisorbed and 135 mg/g AgX is physisorbed. The large chemisorption of AgX compared to other metal zeolites is explained by the capacity of Ag⁺ to bond by the following mechanism:

Studies have shown that silver sorbents can be regenerated with hydrogen gas at 400-500°C with the liberation of hydrogen iodide gas:

Lead-exchanged zeolites (PbX) can be used to chemisorb the HI produced by this reaction. Because of the substantially lower cost of lead than silver, PbX is a potentially more favorable waste form. Early indications, however, are that PbXI is not a stable material and may not be a satisfactory disposal form imbedded in concrete.

When silver-nitrate-impregnated amorphous silica is the sorbent, the reaction sequence has been shown to be:

By analogy, the speculated mechanism for the reaction of methyl iodide would be:

Regeneration of the silver-nitrate-impregnated silicic acid, although untested, should also be possible.

The major disadvantage of silver sorbents is their high cost, which is very important if they are considered as a terminal waste form. It is, however, possible to convert the iodine to PbXI or even to $\text{Ba}(\text{IO}_3)_2$ in subsequent chemical steps. Silver sorbent collection has major advantages of being inherently less complicated than wet scrubbing methods and less expensive to operate.

d. Caustic Scrubbing

In theory, caustic scrubbers should be able to remove elemental I_2 from the off-gas stream by the well-known disproportionation reaction:

In practice, they have never been more than 90% efficient, partially because of their inability to remove organic iodides from the off-gas stream. They also trap CO_2 and NO_x ; the radioactive iodine and carbon could be separated from these mixtures as precipitated barium compounds, $\text{Ba}(\text{CO}_3)_2$ and $\text{Ba}(\text{IC}_3)_2$.

e. Conclusion

All four processes are capable of producing $\text{Ba}(\text{IO}_3)_2$ that can be incorporated in concrete for terminal disposal; the Iodex process does this most effectively. If sorption by silver sorbents is the preferred collection method, an alternative to disposal of iodine as $\text{Ba}(\text{IO}_3)_2$ in concrete would be disposal of AgZI , AgXI , and PbXI either as-received or encapsulated in concrete.

3. Immobilization of Iodine

Given below are the results of our investigation of the chemical and physical parameters important to methods for immobilizing iodine. The two primary methods are (1) incorporating $\text{Ba}(\text{IO}_3)_2$ into concrete and (2) collecting I-129 with silver sorbents and then disposing of them. Other iodine immobilization methods are also discussed.

a. $\text{Ba}(\text{IO}_3)_2$ in Concrete

(1) Solubility Data

Although barium iodate is spoken of as an insoluble compound, it does have a measurable solubility in water. The solubility data presented in Fig. III-7 are compiled from several sources and were measured with an ionic strength of 0 (no other anions or cations present). These

Fig. III-7. Solubility of $\text{Ba}(\text{IO}_3)_2$ vs. Temperature [BOUSQUET-1969A, GMELIN-1960 LINKE, FEDOROV, JONES-1973B, and STEPHEN]. (The data points below the line were ignored. They all came from the same experimenters, whose measurement at 25°C did not agree with the consensus of six independent measurements.)

data show a significant temperature dependence for the solubility of $\text{Ba}(\text{IO}_3)_2$ in water; there is a tenfold increase in the solubility over the temperature range 0-100°C.

Data on the solubility of barium iodate in pure water is not enough to describe the behavior of $\text{Ba}(\text{IO}_3)_2$ under practical conditions. Complexing of Ba^{2+} by other anions can increase the solubility of $\text{Ba}(\text{IO}_3)_2$ substantially, as can changes in the ionic strength of the aqueous environment. Fedorov *et al.* have shown this effect by varying the ionic strength with mixtures of $\text{LiClO}_4/\text{LiNO}_3$ between 0.5 and 4.0 M [FEDOROV]. The solubility of $\text{Ba}(\text{IO}_3)_2$ was increased by both increases in the ionic strength and the nitrate concentration. Formation constants for $\text{Ba}(\text{NO}_3)^+$ and $\text{Ba}(\text{NO}_3)_2$ were both measurable. Figure III-8 shows the effect of nitrate concentration on the solubility of $\text{Ba}(\text{IO}_3)_2$; ClO_4^- was used to maintain constant ionic strength and was assumed not to complex Ba^{2+} .

Fig. III-8. The Effect of NO_3^- and Ionic Strength (I) on the Solubility of $\text{Ba}(\text{IO}_3)_2$ at 25°C [FEDOROV].

The rate of dissolution of $\text{Ba}(\text{IO}_3)_2 \cdot \text{H}_2\text{O}$ was measured by Jones et al. [JONES-1973A]. In relating their data by a model that considered both slow chemical processes at the surface of the crystal and diffusion through the water layer near the surface, Jones et al. determined that the dissolution rate was basically controlled by diffusion; there was no great chemical resistance to dissolution.

(2) Thermal Stability

It is known that $\text{Ba}(\text{IO}_3)_2 \cdot \text{H}_2\text{O}$ is the stable form at ambient temperatures and humidities. According to [DUVAL], as $\text{Ba}(\text{IO}_3)_2 \cdot \text{H}_2\text{O}$ is heated, dehydration begins at 43°C , is barely discernible at 60°C , and accelerates near 79°C . A decrease in the rate of water weight loss with increasing temperature is observed when 75% of the water of hydration is removed and the loss of the last 25% proceeds more and more slowly. Complete loss is not obtained until 320°C . (On the other hand, [BISWAS] reports water loss of $\text{Ba}(\text{IO}_3)_2 \cdot \text{H}_2\text{O}$ is measurable in the range of 80 to 180°C . The water of hydration returns spontaneously as the $\text{Ba}(\text{IO}_3)_2$ is left to stand in the atmosphere.) There is no change in weight loss or in the differential thermal

analysis curve of pure $\text{Ba}(\text{IO}_3)_2$ between 320° and 476°C, until the Rammelsberg reaction begins:

Reaction III-24 becomes violent when the temperature 610°C is reached. This reaction was complete by 666°C, and no further reaction was seen before Duval et al. stopped their scan at 950°C.

The presence of a purely acidic substance, CO_2 , does not greatly change the decomposition temperature for $\text{Ba}(\text{IO}_3)_2$, but does change the decomposition product yields [GONTARZ-1974, -1980]. As $\text{Ba}(\text{IO}_3)_2$ decomposes in the presence of CO_2 , the reaction becomes:

These same authors studied the effect of BaO and I^- on the thermal decomposition of iodate. When the ratio of $\text{Ba}(\text{IO}_3)_2/\text{BaO}$ was 1/4, the onset of decomposition was measured at 330°C by differential thermal analysis; there was no weight loss during decomposition. The decomposition was determined to be one of disproportionation of iodate:

They also found that the addition of iodide to iodate salts substantially reduced the decomposition temperature. For example, a mixture of $\text{Ca}(\text{IO}_3)_2$ and 5CaI_2 began to decompose at 330°C; $\text{Ca}(\text{IO}_3)_2$ alone showed no signs of decomposition until 480°C. The decomposition reaction is believed to be:

In conclusion, the thermal decomposition of $\text{Ba}(\text{IO}_3)_2$ does not appear to be of great concern. The low amounts of heat generated by the radioactive decay of I-129 is not enough to significantly raise the waste form above its ambient temperature. It has been shown that the chemical environment affects both the minimum temperature of decomposition and the decomposition product. Unless there are extraordinary curing procedures for the concrete before storage, the stability of $\text{Ba}(\text{IO}_3)_2$ under thermal conditions likely to be encountered in a disposal site is assured. In a basic environment like concrete, reaction III-26 would likely occur on heating the waste form above 330°C. Since the iodine species produced in reaction III-26 are not volatile as Ba^{2+} or Ca^{2+} salts, there would be no immediate iodine release in a heat-producing transportation accident. The fraction of I^- , however, would decrease the leach resistance of the waste form because of the much greater water solubility of BaI_2 vs. that of $\text{Ba}(\text{IO}_3)_2$.

(3) Heat Capacity

The molar heat capacity of $\text{Ba}(\text{IO}_3)_2$ was measured by Bousquet et al. [BOUSQUET-1969B] between 10 and 650 K. In the range 300-650 K the molar heat capacity, C_p , in units of $\text{cal degree}^{-1} \text{mol}^{-1}$ can be calculated by the relationship

$$C_p = 59.64 - 1.48 \times 10^6 T^{-2} \quad (\text{III-28})$$

(4) Point of Zero Charge

The point of zero charge (PZC) is the concentration of the cation M^{z+} or anion A^{x-} at which point the net charge of the precipitate, M_xA_z , is neither positive nor negative. As pointed out by [HONIG], for $\text{Ba}(\text{IO}_3)_2$, if there are equal equivalents of Ba^{2+} and IO_3^- in solution ($[\text{IO}_3^-] = 2[\text{Ba}^{2+}]$), the precipitate has a positive charge. The surface charge of precipitated barium iodate does not become neutral until the concentration of iodate in the aqueous phase is equal to or larger than 0.11 M. (The concentration of Ba^{2+} is set by the solubility product.) Other anions in solution would of course also, to some extent, modify the positive charge of the precipitate.

The charge of a precipitate is very much a function of the cations and anions in the salt. Silver iodide, for example, has a negative charge at its equivalence point. A high surface charge of a precipitate is the major contribution to (1) the formation of the colloidal precipitate and (2) the stability of the colloidal suspension. The sorption of ions on a solid or one solid on another is a function of a materials surface charge.

(5) Redox Behavior of Iodine Species

Table III-16 is a compilation of relevant oxidation-reduction couples in acid solution. In a basic environment like portland cement, IO_3^- would appear to be stable to reduction to iodide by water or hydroxide. Once it is leached out of the concrete into an anoxic groundwater at pH 7-9, it would be reduced by Fe(II) to iodide [ALLARD-1980]. Allard et al. give the redox potential of deep anoxic groundwaters to be less than 0.12 V at pH = 8. The redox potentials for iodine species are also given by the following relationships:

$$\text{I}^-/\text{IO}_3^- \quad E = 1.09 - 0.0592 \text{ pH} \quad (\text{III-29})$$

$$\text{I}^-/\text{IO}^- \quad E = 1.32 - 0.0592 \text{ pH} \quad (\text{III-30})$$

$$\text{I}^-/\text{I}_{2(s)} \quad E = 0.536 + 0.0592 \text{ pI}^- \quad (\text{III-31})$$

According to Clark and Howerton [CLARK-1980], the presence of sulfite in portland cement reduces some of the $\text{Ba}(\text{IO}_3)_2$ encapsulated in the concrete to BaI_2 .

Table III-16. Redox Potentials Pertinent to $\text{Ba}(\text{IO}_3)_2$ in Concrete [DEAN, MCMANUS, PLATT-1978]

Half Reaction	Standard Potential at 25°C in Volts
$\text{Ba}^{2+} + 2\text{e}^- \rightleftharpoons \text{Ba}$	-2.91
$\text{H}_5\text{IO}_6 + \text{H}^+ + 2\text{e}^- \rightleftharpoons \text{IO}_3^- + 3\text{H}_2\text{O}$	+1.6
$\text{IO}_3^- + 5\text{H}^+ + 4\text{e}^- \rightleftharpoons \text{HIO} + 2\text{H}_2\text{O}$	+1.14
$\text{IO}_3^- + 6\text{H}^+ + 5\text{e}^- \rightleftharpoons 1/2 \text{I}_2 + 3\text{H}_2\text{O}$	+1.19
$\text{I}_2(\text{aq}) + 2\text{e}^- \rightleftharpoons 2\text{I}^-$	+0.621
$\text{IO}_3^- + 3\text{H}_2\text{O} + 6\text{e}^- \rightleftharpoons \text{I}^- + 6\text{OH}^-$	+0.26
$\text{SO}_4^{2-} + \text{H}_2\text{O} + 2\text{e}^- \rightleftharpoons \text{SO}_3^{2-} + 2\text{OH}^-$	-0.93
$\text{O}_2 + 2\text{H}_2\text{O} + 4\text{e}^- \rightleftharpoons 4\text{OH}^-$	+0.41 (1 M NaOH)
$\text{O}_2 + 4\text{H}^+ + 4\text{e}^- \rightleftharpoons 2\text{H}_2\text{O}$	+1.229
$\text{Fe}(\text{III}) + \text{e}^- \rightleftharpoons \text{Fe}(\text{II})$	+0.771
$\text{Fe}(\text{OH})_3 + 3\text{H}^+ + \text{e}^- \rightleftharpoons \text{Fe}^{2+} + 3\text{H}_2\text{O}$	+0.93

(6) Density of $\text{Ba}(\text{IO}_3)_2$ and $\text{Ba}(\text{IO}_3)_2 \cdot \text{H}_2\text{O}$

According to [DEAN], the density of $\text{Ba}(\text{IO}_3)_2$ (molecular weight = 487.15 g/mol) is 5.23 g/cm³ at 20°C. The density of its monohydrate, $\text{Ba}(\text{IO}_3)_2 \cdot \text{H}_2\text{O}$ (molecular weight = 505.16 g/mol), is 4.657 g/cm³ at 18°C.

(7) Leaching Behavior of $\text{Ba}(\text{IO}_3)_2$ in Concrete

According to [ALTOMARE], the incorporation of $\text{Ba}(\text{IO}_3)_2$ in concrete is a prime candidate for disposal of ¹²⁹I because:

- The encapsulation is a low temperature process. Iodine compounds are not stable at the high temperatures of glass formation; encapsulation of ¹²⁹I in borosilicate glass is therefore not practical.
- Barium iodate has low aqueous solubility.
- Barium iodate can be prepared directly from partially soluble barium hydroxide and the product of the Iodox process without the addition of superfluous ionic species. All the resulting slurry of $\text{Ba}(\text{IO}_3)_2$ can be incorporated in the concrete so there is no secondary liquid waste.

The incorporation of $\text{Ba}(\text{IO}_3)_2$ in concrete has been tested with a very strong emphasis on leaching characteristics by researchers at ORNL [CLARK-1976, -1977A, -1977B, -1978, -1980; MOORE; MORGAN-1979] and at PNL [BURGER-1981A, -1981B; CHIKALLA-1981A, -1981B, -1981C].

Researchers at ORNL have reported that concrete can be prepared in the laboratory, incorporating as much as 21 wt % iodine in the form of $\text{Ba}(\text{IO}_3)_2$. Because of mixing difficulties with high $\text{Ba}(\text{IO}_3)_2$ compositions, however, they recommend a practical maximum of 15 wt %. The standard technique for preparation of leach samples was to mix HIO_3 with a slight excess of barium hydroxide, with enough water in the resulting barium iodate slurry to make up the concrete aggregate. The type I portland cement and the $\text{Ba}(\text{IO}_3)_2$ slurry were mixed in a Waring-type blender, and the concrete mix poured into (5-cm diameter by 5-cm high) cylindrical polyethylene containers. After casting, the samples were allowed to stand for 28 d or longer in a water-saturated atmosphere at room temperature. Typical conditions were 5 wt % excess barium hydroxide and a 0.72 weight ratio of water/cement.

The leach tests at ORNL were done statically, but the leach solution was changed to sample periodically (once a day, once a week, or, sometimes, once a month). The data collected from these tests tended to fit the pattern of a conventional leach experiment whose mechanism was controlled by diffusion. The theoretical equation for diffusion-controlled leaching is:

$$\left(\frac{\sum a_n}{A_0} \right) \left(\frac{V}{S} \right) = 2 \left(\frac{D_e}{\pi} \right)^{1/2} (t)^{1/2} \quad (\text{III-32})$$

where a_n is the weight of iodine per leaching period;

A_0 is the initial iodine content of the specimen
in the same units as a_n ;

V is the leachate volume in cm^3 ;

S is the apparent specimen surface area in cm^2 ;

D_e is the modified effective diffusivity in cm^2/s ;
and

t is the leaching time in s.

Figure III-9 shows some of the leaching data, plotted as the normalized fractional loss $(\sum a_n/A_0) (V/S)$ vs. the square root of time. There appears to be a slowdown of leaching at long leach times; this slowdown, the experimenters feel, is a consequence of their changing the sampling period from daily, to weekly, and to monthly as the experiments progressed. The inflection points seen on the curves correspond to these changes in sampling procedure. The data suggest that, at least between 5 and 12 wt % iodine, there is little deterioration of concrete's ability to resist leaching by the addition of $\text{Ba}(\text{IO}_3)_2$. As shown in Fig. III-9, the leach resistance of concrete containing 2.9 wt % iodine is slightly higher than the other concretes.

Fig. III-9. The Effect of Iodine Content on the Leach Rate of Barium Iodate Concrete, No Additives [CLARK-1977B].

Table III-17 is a summary of the results of these studies in distilled water. The influence of iodine concentration in the concrete, curing time of the concrete, excess barium hydroxide, and radiation was nil to slight. The maximum radiation dose produced from a Co-60 source, 2×10^7 rad, was equivalent to 660 y of self-irradiation but at a rate one million times higher. The water/cement ratio and cement type had a slight effect on the leach rate. Leach rates determined for iodine in type I cement concrete, used for the majority of these leach tests, were qualitatively determined to be as low or lower than those measured for iodine in the type II, III, IP, IS, and high-alumina cement concretes. The addition of fly ash to the concrete and use of low temperatures were two effective means of lowering the leach rate.

The parameter that was determined to be the most important to the leach rate of iodine from barium iodate incorporated in concrete was the composition of the leachant. The diffusivity of iodine into various leachants is shown in Table III-18. The explanation for the much lower leach rates than in distilled water for the water that contains salts is the formation of an insoluble barrier on the face of the concrete. Scanning electron

Table III-17. Summary of Iodine Leach Study Results from Ba(IO₃) in Concrete [MOORE]

Parameter	Range	Effect on Leachability	Minimum Order of Magnitude of Diffusivity, cm ² /s
Iodine Concentration	9-21 wt %	none	10 ⁻¹⁰
Curing time	7-112 d	none	10 ⁻¹⁰
Excess Ba(OH) ₂	5-25 wt %	none	10 ⁻¹⁰
Radiation	0-2 × 10 ⁷ rads	none	10 ⁻¹⁰
Fly Ash	0-12%	inverse	10 ⁻¹¹
Water/Cement Weight Ratio	0.5-0.9	slight direct	10 ⁻¹⁰
Cement Type	type I, II, III, IS, IP, high-alumina	slight	10 ⁻¹⁰
Temperature	3-50°C	slight direct	10 ⁻¹¹

microscopy on a concrete sample leached in simulated sea water revealed a 0.5-1.0 mm surface film composed primarily of magnesium hydroxide. The surface of concretes leached by tap and groundwaters showed high concentrations of calcium and zinc, with low concentrations of strontium, iron, and titanium. (Only CaCO₃ was formed on the surface of concrete leached by distilled water.)

Although it seems clear that these films are responsible for retarding the leaching of iodine from concrete, the mechanism of retardation is not at all clear. Morgan et al. listed three possibilities [MORGAN]; the films

- (1) act to stop erosion of the concrete,
- (2) form an adsorption layer for the iodine, and
- (3) form a layer of very low diffusivity between the concrete and the leachant.

They then discussed reasons why none of these mechanisms was possible. They felt that mechanism (1) could not be important because the accumulated release of iodine was measured to be clearly proportional to the square root of time

Table III-18. Effect of Different Leachants on the Iodine Diffusivity (D_e) from $Ba(IO_3)_2$ in Concrete [MOORE].

Leachant	D_e , cm^2/s
Distilled H_2O	1.3×10^{-10}
Simulated Sandia Brine	2.4×10^{-11}
Simulated Sea Water	1.5×10^{-14}
0.03 M $MgCl_2$	1.7×10^{-14}
ORNL Tap Water	5.9×10^{-14}
Oak Ridge Groundwater	7.9×10^{-15}
0.01 M $ZnCl_2$	5.7×10^{-14}

for all the leachants studied. If destruction of the concrete were the mechanism of iodine release, the accumulated loss would be linearly proportional to time. Mechanisms (2) and (3) were negated by x-ray fluorescence analysis data. While iodine was clearly visible in the concrete matrix, none was visible in the films; if the films were acting as either an adsorption or diffusion barrier, iodine would have been found in higher concentrations than in the concrete.

Morgan *et al.* conclude that their measured iodine leach rates should be considered to be an upper limit to what could be expected for the release of radioiodine from barium iodate incorporated in concrete. Leaching experiments in which the leachant is changed daily, with a zero concentration of iodine at the surface of the specimen, is not a likely condition for waste buried in a deep geologic formation or beneath the ocean floor, where flow rates are measured in the order of meters per year. Diffusion and dissolution should be greatly attenuated under these conditions.

Altomare *et al.* [ALTOMARE] also addressed this issue and made some additional strong arguments against extrapolating these data. They argue that these data were collected by measuring the leaching of relatively small samples for a few hundred days and, therefore, that it is dangerous to apply them to full-size waste canisters over several hundred or thousand years. Because the penetration of water into concrete is extremely slow (Section III.B), once the iodine close to the surface is depleted, the leaching will slow down. It has been shown by [HATTA] that it takes sea water 1 y to penetrate 1 cm into a concrete block. As the readily accessible iodine near the surface is depleted, the inverse dependency of volume to surface area given in Eq. III-32 would be expected to break down due to the depth-of-penetration effects.

In spite of this criticism, Altomare et al. [ALTOMARE] used these ORNL data for distilled water:

$$D_e = 1.9 \times 10^{-4} e^{-4163/T} \text{ cm}^2/\text{s} \quad (\text{III-33})$$

(T is in K) to project to a full-scale waste-disposal situation. This diffusivity translates into a penetration rate of 1×10^{-2} cm/y based on the specimen size tested. The projected time of complete iodine leaching from a cubic waste container 2.3 m on a side filled with 6000 kg of concrete containing 10% iodine was calculated to be 1.7×10^4 y. Even with all these approximations, it is obvious that this time is insignificant compared to 1.7×10^8 y, the ten half-lives of decay necessary to reduce the amount of ^{129}I to less than 0.1% of its initial quantity.

Researchers at ORNL also studied, to a limited extent, the effect of concrete additives on the leaching of iodine $\text{Ba}(\text{IO}_3)_2$ in concrete (9.05 wt % iodine). The additives tested were:

- 0.5% Na_2SiO_3
- 0.5% H_2SiF_6
- 0.9% butyl stearate
- 1.2% polybutene 24 plus 0.5% aerosol OT
- polybutene 20 coating of specimen
- 1.4% PbO

All the additives tested but lead oxide caused some improvement in leach resistance for the concrete; the greatest improvement was found for butyl stearate, which lowered the leach rate by ~ 3 fold. Fluosilicic acid showed a great discrepancy for the leaching behavior of the two samples tested, which were prepared with an attempt to formulate them identically. The average leach rate retardation was close to that of butyl stearate and, therefore, Clark et al. considered fluosilicic acid worthy of further study.

In leach tests (both static and dynamic) performed at PNL on iodates and iodides in portland type III concrete, Burger et al. [BURGER-1981A] reached many of the same conclusions mentioned above. Their results can be summarized as follows:

- (1) Approach to dynamic leach conditions increased the measured leach rates.
- (2) High temperatures increased leach rates.
- (3) High carbonate leachate concentrations decreased leach rates.
- (4) Iodine loading in cement of more than 5 wt % increased leach rates.

- (5) Low iodine compound solubility decreased leach rates (AgI , $K_{sp} = 8.3 \times 10^{-17}$ vs. $\text{Ba}(\text{IO}_3)_2$, $K_{sp} = 1.5 \times 10^{-9}$).
- (6) No additives or coatings had a significant effect on leach rates.

It is significant that the leach rate for iodine was measured to be within a factor of four for $\text{Ba}(\text{IO}_3)_2$, $\text{Sr}(\text{IO}_3)_2$, and $\text{Ca}(\text{IO}_3)_2$, while their solubility products increase in the order given above as 1.5×10^{-9} , 3.3×10^{-7} , and 7.1×10^{-7} . Based on its lower solubility, $\text{Ba}(\text{IO}_3)_2$ would be predicted to leach at a much lower rate than $\text{Ca}(\text{IO}_3)_2$. A probable reason for the essentially equivalent leach rates of barium and calcium iodates is the sizable amount of sulfate ion in portland III cement (3.1 wt % -- see Section III. B for details). Because of the lower solubility of BaSO_4 ($\sim 2 \times 10^{-5}$ g/L) than $\text{Ba}(\text{IO}_3)_2$ ($\sim 3 \times 10^{-3}$ g/L), at least some of the barium should be converted to BaSO_4 during the preparation of the cement; the iodate becomes incorporated in the cement as the more soluble $\text{Ca}(\text{IO}_3)_2$.

In conclusion, studies of the leaching behavior of $\text{Ba}(\text{IO}_3)_2$ in concrete indicate that the rates of iodine leaching are much too high to contain ^{129}I for the times necessary for its decay. No additives studied thus far have effectively altered this conclusion. The only significant parameter for reducing the leaching of iodine from barium iodate incorporated in concrete was the leachant. The use of waters containing various salts and minerals was determined to lower the diffusivity constant of iodine by 4-5 orders of magnitude from that measured in distilled water.

(8) Radiation Studies of Barium Iodate in Concrete

Based on the data presented in Section III.D.1, a concrete waste form containing 9.05 wt % iodine (75% I-129) would receive a dose rate of 2.00 rad/h. This dose rate corresponds to 5.6×10^{-9} J/s-g concrete or 3.5×10^{10} eV/s-g concrete.

Clark and Howerton studied how this self-irradiation would influence the radiolytic reduction of $\text{Ba}(\text{IO}_3)_2$ in concrete to the more soluble BaI_2 [CLARK-1978, -1980]. To do this, they subjected concrete cylinders (2.4-cm diameter by 2.5-cm high) in a ^{60}Co γ flux of $4.4\text{--}5.3 \times 10^{15}$ eV/s-g for a period of up to one week. The concrete specimens contained 9.05 wt % iodine as $\text{Ba}(\text{IO}_3)_2$ and were cured for 28 d before use. The amount of I^- present in the concrete specimens after various irradiation times was detected by conventional analytical techniques. No elemental I_2 was ever detected in any specimen.

The data obtained from analyses of irradiated and un-irradiated concrete specimens are shown in Fig. III-10 as a plot of mg I^- /g concrete vs. irradiation time in days. The slope through the data corresponds to an iodide production of 7.22×10^{-10} g I^- produced/s-g concrete. The non-zero intercept of the iodide content of the barium-iodate-incorporated

concrete can be explained by the presence of reductants, primarily sulfite, in the portland cement. During the concrete-mixing process, the reaction

was taking place. Analysis of the cement confirmed the existence of enough SO_3^{2-} to produce the iodide found in the unirradiated concrete specimen.

The linearity of the data of Fig. III-10 implies that any back reaction is negligible. The slope of this line predicts a G value for the radiolytic reduction of iodate to iodide of 0.083 I^- ions/100 eV. Based on the self-irradiation of I-129 as $\text{Ba}(\text{IO}_3)_2$ in concrete of 3.47×10^{10} eV/s-g concrete, $2.1 \times 10^{-4}\%$ of the IO_3^- would be converted to I^- per year. In 1000 y, only 0.21% of the iodate would be converted to iodide—a trivial amount compared to the IO_3^- that would be leached from the concrete.

Fig. III-10. Radiolytic Reduction of Iodate to Iodide in a Barium Iodate Concrete Containing 9.05 wt % Iodine [CLARK-1980].

Fig. III-11. Effect of the Addition of 1.8 wt % of Ag_2O , HgO , or PbO on the Radiolytic Reduction of Iodate to Iodide in Barium-Iodate Concrete Containing 9.05 wt % Iodine [CLARK-1980].

These same researchers also measured the effect of adding three oxides of metals that form low-solubility iodide salts (Ag_2O , HgO , and PbO). The data for those studies are presented in Fig. III-11. The G values calculated from the data of Fig. III-11 are presented in Table III-19. Both Ag_2O and HgO appear to increase the G value for the formation of I^- from IO_3^- , while PbO decreases it slightly. The acceleration of iodide formation due to the presence of Ag_2O seems certain, but the modest decrease and increase in iodine formation shown for PbO and HgO , respectively, are uncertain within the analytical limits of the iodide measurement.

In conclusion, the effects of self-irradiation on the reduction of $^{129}\text{IO}_3^-$ ion to $^{129}\text{I}^-$ is negligible when compared to predicted leaching of iodine from $\text{Ba}(\text{IO}_3)_2$ incorporated in concrete. Moreover, the addition of Ag_2O , HgO , and PbO to the concrete will have a negligible effect on the already negligible radiolysis reaction.

Table III-19. Effect of Additives on the G Values for the Production of I^- Ion from $Ba(IO_3)_2$ in Concrete (additive concentration in concrete = 1.4 wt %)

Additive	G Value, I^- ion/100 eV
none	0.083
Ag_2O	0.151
HgO	0.105
PbO	0.060

b. Silver Sorbents

(1) General Considerations

(a) Properties of Silver-Exchanged or Silver-Nitrate-Impregnated Sorbents

The use of silver sorbents to collect I-129 has been given much study over the last ten years and has been reviewed by [JUBIN, HOLLADAY-1979, and BURGER-1979A]. Once these sorbents are loaded with ^{129}I , there are three options for disposal of the collected iodine:

Option (1). Regeneration of the silver sorbents for reuse by removal of the radioiodine. The radioactive iodine can be either loaded on lead-exchanged zeolite for disposal by options (2) or (3) below, or can be chemically converted to AgI or $Ba(IO_3)_2$ for disposal in concrete [CHIKALLA-1982, ALTOMARE, THOMAS, JUBIN, HOLLADAY-1979, MURPHY-1977].

Option (2). Direct disposal of the iodine-loaded solid sorbent in metal canisters for long term disposal [BURGER-1979A, -1981A, -1981B; PENCE-1974; CHIKALLA-1980; and WOOD].

Option (3). Encapsulation of the solid sorbent into concrete for long-term disposal [BURGER-1979A, -1981A, -1981B; CHIKALLA-1980, -1981A, -1981B, -1981C; PLATT-1980; BEONE].

Each of these options will be discussed in detail in the following sections. Before these disposal options are discussed, it seems appropriate to discuss pertinent chemical and physical characteristics of the silver sorbents being considered for the collection and disposal of iodine-129. The three most studied types are two silver-exchanged zeolites, silver-exchanged faujasite (AgX) and silver-exchanged mordenite (AgZ), and a silver nitrate-impregnated amorphous silicic acid (AC-6120, commercially available, is the most researched of this type).

The silver-exchanged faujasite most studied is one prepared from Linde-type 13X molecular sieve. This material is commercially available from Linde as the sodium-exchanged form and can be easily converted to the silver form. This zeolite is a high-porosity material with three types of channels running through it. The faujasite unit cell is described by [BRECK] as $\{(Na_2, K_2, Ca, Mg)_{29.5} [(AlO_2)_{59}(SiO_2)_{133}] \cdot 235H_2O\}$. The unit cell of the zeolite X is designated as $\{Na_{86}[(AlO_2)_{86}(SiO_2)_{106}] \cdot 264H_2O\}$. Linde's synthetic zeolite-X has the formulation $\{(0.83 \pm 0.05)Na_2O \cdot Al_2O_3 \cdot (2.48 \pm 0.03)SiO_2 \cdot nH_2O\}$ [JUBIN].

Mordenite has only about one third of the ion-exchange capacity of faujasite because of the much higher silica-to-alumina ratio of its structure. This higher SiO_2/Al_2O_3 ratio has the beneficial effect, however, of increasing its hardness and resistance to acid decomposition [JUBIN]. These attributes appear to be important factors in the much greater capacity for regeneration of iodine fixation of AgZ vs. that of AgX. The unit cell of natural mordenite has the chemical composition $Na_{48}[(AlO_2)_8(SiO_2)_{40}] \cdot 24H_2O$ [BRECK]. Zeolon-900, sold by the Norton Company, is a self-bonded synthetic mordenite with the chemical formulation $(Na_2O)_4 \cdot (Al_2O_3)_4 \cdot (SiO_2)_{40} \cdot 24H_2O$. The silver ions that are exchanged into these mordenites can be converted to a metallic form by pretreating the AgZ with H_2 gas at $500^\circ C$ for ~ 16 h [BURGER-1981B]. Experiments have shown that the metallic silver mordenite Ag^0Z has a higher capacity for iodine chemisorption than AgZ [JUBIN].

Table III-20 lists several physical parameters of Linde 13X and Zeolon 900 molecular sieves important to their use in the fixation of iodine-129. Table III-21 shows the volume distribution (in units of volume percent) in terms of a single zeolite particle and in terms of a bed (or canister) of zeolite particles. Breck [BRECK] makes the important point that the interpellet void in Table III-21 is an equilibrium value. At initial packing the particles tend not to be as densely packed, and the void volume is closer to 38 vol %.

For long-term storage of ^{129}I , silver-nitrate-impregnated materials offer the advantage that silver iodide is clearly the product of their fixation reaction; the iodine is, therefore, held very tightly to the waste form as insoluble silver iodide (the solubility of AgI vs. temperature is shown in Fig. III-12). Another important factor is that these materials cost only $\sim 1/10$ as much as the molecular sieve materials [HOLLADAY-1979]. According to [JUBIN], there have been three commercial types of this material tested:

GX-135 (North American Carbon Company). This silver nitrate containing aluminum silicate was highly effective at fixing iodine from both I_2 and CH_3I in tests at ORNL, but was discontinued because of the handling problems caused by its fine mesh size (8×16 mesh) [JUBIN].

H-151 (Aluminum Company of America). There is a limited amount of work on the use of this high-alumina-content (90%) material impregnated by $AgNO_3$ and other nitrate salts: only silver nitrate was found to bind iodine irreversibly. Table III-22 gives the physical specification of this material [JUBIN].

Table III-20. Properties of Commercially Available Molecular Sieves,
Linde 13X and Zeolon 900 [BRECK]

Property	Linde 13X		Zeolon 900	
	1/16" (Pellet)	1/8" (Pellet)	1/16" (Extruded)	1/8" (Extruded)
Form	Na ⁺		Na ⁺	
Structure Type	Zeolite X		Mordenite	
Pore Size, Å	10		7	
Bulk Density, lb/ft ³ (g/cm ³)	38 (0.61)		40 (0.64)	
Crush Strength, lb	12	25	--	
Water Content, wt %	4.5		--	
Water Capacity per 100 lb of Activated Adsorbent, lb	28.5 (25°C, 17.5 torr)		10	

Table III-21. Volume Distribution in a Single Molecular
Sieve Pellet and in a Bed of Molecular
Sieve Pellets [BRECK]

Constituent	Pellet, vol %		Bed, vol %	
	1/16" size	1/8" size	1/16" pellets	1/8" pellets
Intracrystal Void	28.1	28.8	19.1	19.6
Intercrystal Void	33.4	31.5	22.7	21.4
Solid Portion of Crystal	29.2	30.1	19.9	20.5
Solid Portion of Binder	9.3	9.6	6.3	6.5
Interpellet Void	--	--	32.0	32.0

Fig. III-12. Solubility of AgI vs. Temperature [MEYER].

Table III-22. Specification of Alumina-Base Adsorbent: Alcoa Alumina H-151 [JUBIN]

Composition, %	
Al ₂ O ₃	90.0
Na ₂ O	1.4
Fe ₂ O ₃	0.1
SiO ₂	1.7
Loss on ignition (1100°C), %	6.0
Surface area, m ² /g	390
Bulk density (packed), lb/ft ³	53
Specific gravity	3.2
Particle sizes, cm	0.34 and 0.12

AC-6120 (United Catalysts, Inc.). Silver-nitrate-impregnated silicic acid has been developed by the Federal Republic of Germany as a non-regenerable solid sorbent for iodine-129 collection and disposal. This material, which has a silver content of 7-12 wt %, has been used successfully to remove iodine from DOG at the Reprocessing Plants in Karlsruhe, Germany, and Centre de Marcoule, France [BURGER-1979A].

(b) The Stability of Silver Sorbents to Loss of Iodine

Maximum and practical loading limits of iodine per gram of sorbent are important for the collection process. Another equally important parameter for disposal of iodine is the amount of iodine that is chemically and physically bound to the sorbent. Chemically bound iodine will have an expected chemical stability and insolubility as good as or better than those of silver iodide. Physically bound iodine is expected (1) to have much inferior properties to those of AgI and (2) to be much more susceptible to leaching from the waste form. Table III-23 tabulates the iodine loading capacities of several sorbents in terms of their theoretical capacities and in terms of the practical concern of keeping collection efficiency of iodine from the DOG above 99.9%.

The higher loading capacity of AgX makes it appear much more attractive than AgZ or AC-6120. Unfortunately, collection of iodine is only the short-term problem. When one considers the long-term problem of protecting the biosphere from high concentrations of I-129, the virtues of AgZ and AC-6120 become apparent. Workers [BURGER-1981A, -1981B; SCHEELE-1981] have determined that much of the iodine (>50%) sorbed on AgX was removable by Soxhlet extraction using carbon tetrachloride, tetrachloroethylene, toluene, or hexane; in contrast, no iodine could be stripped from AgZI by this same technique. This finding suggests a more stable fixation of iodine by AgZ than AgX. However, both AgXI and AgZI release iodine when heated [BURGER-1982B]. Because the chemical form of iodine in the structure of AC-6120 is certainly AgI, it would also be expected to not release iodine by Soxhlet extraction.

Thermal measurements (TGA and DSC) on AgX, AgZ, Ag^oZ, and AC-6120 loaded with iodine show that the sorption of iodine by those sorbents is very complex, and there are many forms of iodine in their structures that cannot be identified as simple silver compounds. Even AC-6120 does not correspond exactly in its thermal behavior to AgNO₃ and AgI. Figure III-13 shows the results of the thermogravimetric analyses up to 1000°C of AgZ, Ag^oZ, AgX, and AC-6120 loaded with iodine to 50% of their maximum capacities. All the weight loss is not I₂; much of the low-temperature weight loss is water, and some volatilization of AgI along with iodine may have been seen at higher temperatures. It appears that no iodine is lost from AC-6120; rather, the measured weight loss is due to the decomposition of nitrate ion [BURGER-1981A, -1981B; CHIKALLA-1981A; SCHEELE-1981].

Thermogravimetric analyses of several samples of AgZ with varying loadings of iodine have shown that the final iodine content for all loadings tends toward 45 mg I/g AgZ at 1000°C. When the equilibration of the sorbent was reached slowly, this stable fraction appeared to be constant whether the iodine was loaded from a continuous stream of I₂ or CH₃I. Figure III-14 shows the comparable thermal behavior of two samples of AgZ loaded to different degrees by a continuous feed over three weeks of either I₂ or CH₃I.

Table III-23. Maximum and Practical Iodine Loading Capacities of Silver and Lead Sorbents [JUBIN]

Sorbent	Maximum (Theoretical) Sorption, mg I/g	Practical ^a Sorption for Collection, mg I/g
AgX	429	220
PbX	437	390
AgZ	237	190
PbZ	247	398 ^b
AC-6120 (7 wt % Ag)	73	10
AC-6120/H ₁ (12 wt % Ag)	143	10
H-151	88.2	5
GX-135	---	2

^aTo keep collection efficiencies above 99.9%.

^bThis higher-than-theoretical amount of iodine collected may be due to formation of H₂PbI₄ [MURPHY-1977]; it may also just be wrong (discussed later in this section).

In that same figure is the thermal analysis curve of a sample of AgZ loaded in a batch mode over 24 h. The sample for this curve begins its loss of iodine at the same temperature, but has less iodine at 1000°C than the other two samples shown and the many others tested [SCHEELE-1981].

The relative stability to iodine loss of lead-exchanged zeolites was reported by [MURPHY-1977], who measured the vapor pressure of iodine over lead-exchanged zeolites loaded with iodine to 38 wt % for Pb[°]XI, to 40 wt % for PbXI, and to 38 wt % for PbZI vs. temperature. These iodine-loaded lead zeolites were prepared from the reaction of the lead-exchanged zeolites with HI. The effects of hydrogen, nitrogen, and air atmospheres on the I₂ vapor pressures were also studied. The results of this study, presented in Table I-II-24, are somewhat in doubt because of conflicting reporting of these data in this report. It is also not clear how they achieved 38 wt % loading of iodine on Pb[°]X when other data in this same report show a maximum loading of only 192 mg HI/g zeolite (19 wt %) of iodine on Pb[°]X. Their reporting of over 100% (161%) of the theoretical loading of iodine onto PbZ is disturbing.

It has been mentioned by several sources that the solubility of iodide in silver-exchanged and silver-nitrate-impregnated sorbents should be even lower than that of silver iodide [JUBIN, ALTOMARE], but no data have been found to verify this. From the Soxhlet extraction and

Fig. III-13. Thermogravimetric Analyses for (a) AgZ, (b) Ag^oZ, (c) AgX, and (d) AC-6120 (all with 50% capacity loadings of iodine; TG = weight loss; DTG = differential weight loss $\Delta W/\Delta T$) [BURGER-1981B]

Fig. III-14. Effect of Iodine Loading Rate and Species on the Thermal Release of Iodine from AgZI [SCHEELE-1981].

Table III-24. Extrapolated Vapor Pressure of I_2 above $PbXI$, $PbZI$, and Pb^*XI in H_2 , N_2 , and Air Atmosphere at $20^\circ C$ [MURPHY-1977]

Sorbent	Vapor Pressure, atm		
	in air	in H_2	in N_2
$PbZI$	4.7×10^{-6}	4.5×10^{-6}	1.9×10^{-6}
Pb^*XI	9×10^{-19}	8.7×10^{-19}	2.5×10^{-12}
$PbXI$	4.0×10^{-8}	4.0×10^{-11}	4×10^{-13}

thermal studies discussed previously and from the leach data yet to be discussed, it seems this observation is certainly not true, at least for the majority of iodine fixed in these sorbents. A measurement of the solubility of PbI_2 from $PbXI$, however, does show that the solubility of iodide is less than expected from the solubility of PbI_2 in water [THOMAS]. These researchers measured a solubility of 26 mg I/100 mL water at room temperature. The solubility of iodide in water as PbI_2 at $20^\circ C$ is 35 mg I/100 g H_2O [DEAN]. This finding is an indication that the solubility of PbI_2 is a factor in the case of lead-exchanged zeolites.

(2) Disposal Options

(a) Regeneration of the Silver Sorbent

Of the silver sorbents mentioned in this text, only silver-exchanged mordenite appears to be regenerable without great loss of capacity [JUBIN, BURGER-1979A, and THOMAS]. According to [THOMAS], after five regeneration cycles at 400 to $500^\circ C$, the iodine loading capacity of AgX dropped by two fold. At $600^\circ C$, its capacity dropped three fold after 3 cycles. No decline was seen at all for AgZ after the same tests. After 13 recycles the capacity of AgZ was down by 20%. The apparent reason for the loss of capacity for iodine by these zeolites is a collapse in pore structure caused by the recycle. The higher silica content of mordenite leads to a harder structure, more resistant to pore collapse.

If the sorbent is regenerated with the silver-exchanged mordenite, there is still a choice to be made: should the radioactive hydrogen iodide gas be trapped on a lead-exchanged zeolite or should it be scrubbed into an aqueous solution to be converted to an insoluble form, presumably $Ba(IO_3)_2$? Some experimental work has been performed on the scrubbing of HI by various aqueous phases and such a procedure is possible [PLATT-1979A, -1979B, -1979C]. The other option, collection by lead-exchanged zeolites has had a great deal more study and also seems quite possible. Figure III-15 is a block diagram for this process. If this option is selected, the disposal form for long-term storage is (1) $PbXI$ or $PbZI$ with no matrix or (2) encapsulation in a concrete matrix. These alternatives will be discussed in the following sections.

Load Mode Reaction: $2\text{Ag}^{\circ} + \text{I}_2 \rightleftharpoons 2\text{AgI}$
 Regeneration Mode Reaction: $2\text{AgI} + \text{H}_2 \rightleftharpoons 2\text{Ag}^{\circ} + 2\text{HI}$
 Storage Mode Reaction
 Approximated by: $\text{PbO} + 2\text{HI} \rightleftharpoons \text{PbI}_2 + \text{H}_2\text{O}$

Fig. III-15. Block Diagram of Iodine Recovery Process (derived from [THOMAS])

(b) Ag-Exchanged, AgNO₃-Impregnated, or Pb-Exchanged Sorbents as a Disposal Form without Matrix

According to Pence and Staples [PENCE-1974], "Processing of the iodine-laden adsorbent from the primary system to obtain a more concentrated form would not appear to be necessary or desirable." They go on to say: "The product should be ideally suited for long-term storage, provided excess NO₂ and moisture are purged from the adsorbent prior to packaging. Because the decay energy of I-129 is so low, a negligible amount of decay heat will be generated." Wood and Richardson [WOOD] also consider disposal of iodine-loaded sorbents in a container to be an option.

To calculate the specific activity of I-129 in this waste form, one must assume that the exchange of silver ion for sodium ion and the subsequent sorption of iodine does not increase the bulk volume of the material, only its density; this assumption is clearly compatible with what is known about these materials. The maximum and practical loadings of each sorbent are listed in Table II-23. The bulk density of zeolytic sorbents was calculated by correcting for (1) the higher mass of silver and lead ions vs. that of sodium ion and (2) the additional mass increase caused by iodine sorption. The silver-nitrate-impregnated sorbents were corrected for the greater mass of the iodide ion vs. that of the nitrate ion. Tabulated for each potential waste form in Table III-25 are the calculated bulk densities, amount of sorbent necessary to store the iodine produced in a model reprocessing plant of 1500 MTHM/y. and the specific activity of the waste form.

Table III-25. A Comparison of the Waste Disposal Forms for Various Solid Sorbents Alone or Encapsulated in Concrete

Sorbent	Bulk Density of Iodine-Loaded Sorbent, MT/m ³ (lb/ft ³)		Mass to Contain 625 kg of Iodine, ^a MT		Volume to Contain 625 kg of Iodine, ^a m ³		Specific Activity, ^b Ci/MT		Specific Activity, ^b Ci/m ³	
	Maximum ^c	Practical ^d	Maximum ^c	Practical ^d	Maximum ^c	Practical ^d	Maximum ^c	Practical ^d	Maximum ^c	Practical ^d
AgX	1.12(70)	0.99(62)	1.54	3.00	1.38	3.03	52.8	27.1	58.9	26.8
PbX	1.12(70)	1.02(64)	1.51	1.69	1.35	1.66	53.8	48.1	60.2	48.9
AgZ	1.02(64)	0.99(62)	2.78	3.47	2.73	3.50	29.2	23.4	29.8	24.4
PbZ	1.02(64)	0.99(62) ^e	2.67	3.30 ^e	2.62	3.33 ^e	30.4	24.6 ^e	31.0	49.0 ^e
AC-6120 ^f	0.88(55)	0.85(53)	9.04	66.0	10.3	77.6	9.0	1.2	7.9	1.0
AC-6120/H ^g	0.98(61)	0.91(57)	4.62	66.0	4.71	72.5	17.6	1.2	17.2	1.1
H-151	0.90(56)	0.85(53)	7.48	132.0	8.31	155.0	10.9	0.6	9.8	0.5
GX-135 ^f	--	0.85(53)	--	330.0	--	388.0	--	0.02	--	0.02
34 wt % AgZI in concrete ^h	--	2.1(131)	--	11.5	--	5.48	--	7.09	--	14.8
20 wt % PbXI in concrete ⁱ	--	2.1(131)	--	8.45	--	4.02	--	9.62	--	20.2

^aThe amount of iodine release projected from a 1500 MT/yr reprocessing plant in one year [ALTOMARE].

^bBased on 625 kg radioactive iodine (75% ¹²⁹I) or 81.2 Ci.

^cAssuming the maximum theoretical loading of iodine was loaded on the sorbent as 75% I-129 and 25% I-127.

^dAssuming the practical loading limit of iodine (Table III-22) was loaded on the sorbent as 75% I-129 and 25% I-127.

^eBecause there is uncertainty for the value of the practical loading of HI on PbZ, 200 mg iodine/g was used based on the behavior of AgZ or PbX.

^fThe bulk density of H-151 (53 lb/ft³) was used for this calculation.

^gThe greater concentration of the high-density silver nitrate was used to calculate a bulk density (based on that of H-151) of 57 lb/ft³.

^hThe AgZI is loaded with 190 mg I/g AgZ, which is comparable to 160 mg I/g AgZI.

ⁱThe PbXI is loaded with 370 mg I/g PbX.

Another criterion that has been used in picking a disposal form for I-129 is its resistance to leaching of I-129 into the biosphere. There has been very little information discovered in the literature as a basis for comparing the leach resistance of the solid sorbents listed in Table III-23, these same waste forms in concrete, and $\text{Ba}(\text{IO}_3)_2$ encapsulated in concrete. Preliminary static leach studies by Scheele and Wiemers [CHIKALLA-1980] determined that after 28 d the silver sorbents studied (70% loaded AgZI , AgXI , and AC-6120 and 95% loaded Ag^*ZI) had leached two to four orders of magnitude less of their iodine than comparable materials encapsulated in concrete. The fraction of iodine leached from these materials after 28 d varied from $<2.0 \times 10^{-7}$ to 2.0×10^{-5} . In later reports of this work [BURGER-1981A, -1981B, SCHEELE-1981], no additional data were presented; it was merely stated that there was "no improvement when placed in cement." However, [BURGER-1981A] states: "The solid sorbents considered are potential disposal forms as used. However, granular or powder forms present handling problems in addition to much lower resistance to leaching or volatilization. We believe immobilization in a monolithic form is desirable."

The statement on leach resistance does not seem to follow directly from the data presented in [CHIKALLA-1980], but the statement on handling problems of granular or powder forms is an important consideration.

No reports of the leach resistance of the lead zeolites were discovered in reviewing the literature, but, based on the orders-of-magnitude higher solubility of PbI_2 compared to AgI , the lead zeolites would be expected to have orders-of-magnitude higher leach rates than the silver zeolites.

(c) Disposal of Ag-Exchanged, AgNO_3 -Impregnated or Pb-Exchanged Sorbents in Concrete Matrix

The loading levels of AgZI and PbXI in concrete reported by Burger *et al.* [BURGER-1981A] are 0.8 mmol I/g dry concrete and ~ 1 mmol I/g dry concrete, respectively. These levels correspond to ~ 34 wt % AgZI and ~ 20 wt % PbXI in cured concrete. The estimated density of the concrete is 2.1 MT/m^3 . The calculated waste volumes and specific activities of these potential waste forms found in Table III-25 are based on these values. Because of the 32% void volume of packed zeolites, the specific activity of zeolites in terms of Ci/m^3 in concrete is not much lower than that of zeolites without the presence of concrete. The higher density of cement *vs.* that of air makes for large differences in specific activities between zeolite alone and zeolite in concrete when expressed in terms of Ci/MT .

The leach resistance of AgZI in concrete was measured to be about an order of magnitude greater than that for a comparable amount of $\text{Ba}(\text{IO}_3)_2$ in concrete in a dynamic leach test; in static leach tests, the leach rates of these two potential disposal forms were experimentally equivalent [BURGER-1981A]. The dynamic leach rate of pure AgI in concrete is a factor of 10 lower than that for AgZI . The higher leach rate for AgZI *vs.* that of AgI can be explained on the basis of the thermal analysis of AgZI , which has shown that only 45 mg I/g AgZ is held as AgI ; the rest of the 150 mg I/g AgZ is held in more labile forms. At the measured leach rate of $4 \times 10^{-6} \text{ cm/d}$, it would take 100 y to leach 1% of the iodine from a typical 208-L monolith of AgZI encapsulated in concrete.

When PbXI was added to cement, the mixture turned purple, indicating the hydrolysis of the lead-zeolite-iodide material with release of elemental iodine [BURGER-1981A]. Static leach tests for two samples showed how poor the leach resistance for this form is; after 5 d the normalized fraction of iodine released was 0.07-0.2 cm (a rate of $1-4 \times 10^{-2}$ cm/d).

Beone and Di Stefano [BEONE] studied the physical characteristics of concrete containing several concentrations of AgX sorbents. The cement they studied was pozzolanic (approximate composition would be 80% portland I cement/20% fly ash), which is more expensive than regular portland cements but has the advantages of high chemical resistance and a very high encapsulation capacity. The silver-exchanged zeolite they tested was LMX-13X (Union Carbide Europe). The concrete samples were cured in a 100% humidity environment and then allowed to air dry to a constant weight. Figure III-16 shows the rate of water loss during this drying period for several samples of differing composition; these various compositions are listed in Table III-26. The amount of water loss as well as the rate of water loss increase as the zeolite loadings are increased.

Figure III-17 shows the density of concretes at various loading levels of silver-exchanged zeolites. These densities are lower than the ones used for previous assumptions in this section because pozzolanic is a lower-than-average-density cement and the silver-exchanged zeolite was not loaded with iodine.

Beone and Di Stefano [BEONE] also reported results of crush-strength studies of concrete-encapsulated, silver-exchanged zeolites. Their results, shown in Fig. III-18, show that crush strength decreases with increasing zeolite concentration. The decline in crush strength drops almost linearly with zeolite loadings of less than 35 wt %; above this loading level, decline in crush strength with zeolite content becomes far more drastic.

These same researchers also tested the thermal resistance of zeolite-containing concretes with 31.8 wt % zeolite (sample 5, Table III-26) by two methods. The first was a modified IAEA test for thermal resistance of radioactive material. The normal test called for heating the material at 800°C in air for 10 min, but the researchers increased that time to 30 min. The samples tested lost between 18 and 25% weight by water and/or CO₂ and their surfaces were completely fissured. Carbon dioxide was formed from the thermal decomposition of CaCO₃ (CaCO₃ → CaO + CO₂). In the second test, the effect of localized thermal expansion was measured by placing the tip of a Bunsen flame to the concrete specimen for one minute. Two samples were tested. The first, a 65 g cylinder (sample 5), aged for 7 months prior to the test, lost 0.4 wt % during the test. The second, 190 g (sample 5), aged for 2 months before the test, lost 1.3 wt % during the test. Both samples showed fissuring near the area of contact with the Bunsen flame.

Beone and Di Stefano [BEONE] conclude from their results that loading limits for zeolite in concrete should not exceed 35 wt %. In addition, they feel that the use of additives to increase the concrete's strength and thermal resistance are necessary and should be studied.

Fig. III-16. Water Loss vs. Time for Various Loadings of AgX in Concrete [BEONE]. (The sample numbers correspond to those in Table III-26.)

Table III-26. Composition of Concrete Samples Prepared from Pozzolanic Cement and AgX Zeolite [BEONE]

Sample No.	Weight of Dry Cement, g	Weight of Dry Zeolite, g	Weight of Water Associated, g		Zeolite Content in Cured Concrete, wt %
			Cement	Zeolite	
1	900	150	450	45	10.1
2	900	300	450	90	18.1
3	1062	400	532	120	19.8
4	699	450	350	135	30.0
5	900	600	450	180	31.9
6	900	815	450	245	37.7
7	600	600	300	180	40.5
8	900	1100	450	330	44.8
9	450	750	225	225	49.5

Fig. III-17. Density of Concrete Prepared from Pozzolanic Cement and Silver-Exchanged Zeolite of Different Compositions [BEONE].

Fig. III-18. Crush Strength of Concrete Sample vs. the Zeolite Content [BEONE].

c. Alternative Iodine Waste Forms

Iodide sodalite has been the most studied alternative waste form for iodine and has been stated to be most promising [BURGER-1981A; CHIKALLA-1980, -1981B, -1981C; PLATT-1980 ; STRACHAN; VANCE; WINTERS; BARNES]. The chemical formula of sodalite is $\text{Na}_8\text{Al}_6\text{SiO}_{24}\text{X}_2$. The anions ($\text{X} = \text{Cl}^-$, Br^- , I^- , OH^- , etc.) reside in cages in the structure and are tetrahedrally coordinated to four Na^+ ions, which, in turn, are incorporated in the aluminosilicate network. The theoretical maximum loading of iodide ion is 22 wt %, and iodide sodalite has a theoretical maximum density of 2.6 g/cm^3 .

Iodide sodalite has been prepared by mixing, in the proper proportions, solutions of either (1) NaI and NaAlO_2 with an SiO_2 sol in aqueous ammonia or (2) NaOH , NaI , and kaolinite clay in an aqueous suspension. The gel product of this reaction must then be dried, ground into a fine powder, and hot pressed at $\sim 1000^\circ\text{C}$ and 34.5 MPa (5000 psi) for $\sim 1 \text{ h}$. Products of near theoretical density (2.4 g/cm^3) and iodide content ($\sim 19 \text{ wt } \%$) have been prepared that show good thermal and radiation stability [BURGER-1981A, VANCE].

The leach resistance of iodide sodalite was reported to be 100 times higher than that of $\text{Ba}(\text{IO}_3)_2$ in concrete and equivalent to that of AgI in concrete. Silver iodide was determined to be the most leach-resistant material encapsulated in concrete tested [BURGER-1981A].

There are two severe disadvantages to iodide sodalite production. The first is that sodium iodide is neither a conventional nor convenient product of any of the promising collection methods for volatile iodine. Some preliminary work has been done on mixing HI into a pot with excess NaOH to form NaI during sodalite production, but this process proved to be difficult to implement [CHIKALLA-1981C]; hydrogen iodide gas is the product of the regeneration of silver zeolite by hydrogen gas. Therefore, a great deal of initial research would be needed on the production of NaI from the collected radioiodine; this NaI production would later lead to a more complex operation for immobilization of radioiodine. Another disadvantage is the complexity of the iodide sodalite process itself when compared to the mixing of a solid with cement at ambient conditions. There are several steps to this process, one requiring high temperature and high pressure. This disadvantage is compounded because a loss of a few percent of iodine from the sodalite occurs during the hot-pressing step. The trapping and recycling of this iodine add even more complexity to the immobilization process.

d. Sorption of Iodide and Iodate by Minerals and Soil Components

Because the leach rates of I-129 from concrete are higher by several orders of magnitude than its decay rate, the migration rate of I-129 from the waste container to the environment is an important property of the disposal system. If geological burial is chosen for iodine, migration to an aquifer must be slow enough that dilution of I-129 in the groundwater makes it innocuous. If deep sea burial is the disposal option of choice, the migration rate must be low enough that isotopic dilution with natural iodine in the ocean makes I-129 innocuous. The requirement for isotopic dilution by seawater is a far less demanding criterion than that for dilution by groundwater. The following section is a literature survey of the retention properties of natural materials for iodine species, with emphasis on potential backfill-barrier materials that would inhibit the movement of iodine species from the waste canister.

In general, the anion-exchange capacity of minerals and soils is far less than their cation-exchange capacity. Table III-27 shows the anion-exchange capacity of some common minerals [ALLARD-1980]; in that same table, for comparison, are the cation-exchange capacities of some of the minerals [GARRELS]. Anionic I-131 has been tested and used as a means to trace water flow through the ground [KAUFMAN, HAMID]. The major criterion for picking a tracer for these types of studies is the low sorption of the tracer by the solid matrix. It was determined that sorption of I-131 could be a problem when used without a carrier, but that sorption could be eliminated as a problem if nonradioactive iodide was used with the tracer in the range of 10^{-4} to 10^{-3} M as NaI.

A great deal of work has been done in studying what constituents of soil are responsible for its retention of iodine [BOYARINOVA-1973; DE; RAJA; REINIGER; SUGAWARA; WHITEHEAD-1973A, -1973B, -1974A, -1974B, -1978; WILDING]. These studies of soils throughout the world all came to the same general conclusion: there are three variables in soil content that are primarily responsible for sorption of I^- and IO_3^- -- Al_2O_3 , Fe_2O_3 , and organic matter. The importance of each constituent is primarily dependent on the pH of the groundwater

Table III-27. Anion- and Cation-Exchange Capacities of
Some Common Minerals [ALLARD-1980, GARRELS]

Class	Mineral	Exchange Capacities, meq/kg	
		Anion ^a	Cation ^b
1. Sulfide	Chalcopyrite	0.5	
	Pyrite	<0.3	
2. Oxides, hydroxides	Magnetite	0.4	
	Limonite	0.6	
	Gibbsite	0.5	
	Hematite, corundum	<0.3	
3. Halides, Carbonates, Sulfates, Phosphates	Fluorite	0.2	
	Calcite, dolomite	<0.3	
	Anhydrite	<0.1	
	Apatite	<0.1	
4. Silicates			
a. Nesosilicates	Olivine	1.8	
	Zircon	0.7	
	Sphene	0.8	
	Garnet (almandine)	0.4	
b. Sorosilicates	Epidote	0.5	
c. Cyclosilicates	Beryl	1.4	
d. Inosilicates	Augite, hornblende, Attapulgite	<0.2 0.7	
e. Phyllosilicates	Kaolinite	0.5	30-150
	Serpentine	6.9	
	Montmorillonite	0.9	700-1000
	Halloysite	1.5	50-500
	Muscovite	0.6	
	Biotite, chlorite	<0.3	
f. Tectosilicates	Anorthite	0.7	
	Microcline, albite, bytownite, quartz	<0.3	

^aAnion exchange capacity for Br⁻ at pH 8.

^bCation exchange capacity at pH 7.

and on the contact time of the iodine-containing water and the soil. Figure III-19 shows a pH profile of the sorption of iodide by a typical British soil, Al_2O_3 , Fe_2O_3 , and organic matter. The general trend of the data suggests that sorption by organic matter is less pH-dependent than sorption by inorganic material. The soil exhibits a behavior expected from a weighted combination of all three of the soil constituents. Figure III-20 shows the capacity for sorption of I^- by two soils under the same conditions and the rate for that sorption.

Iodates were determined by [WHITEHEAD-1974A] to be, in general, sorbed less by soils, Fe_2O_3 , and Al_2O_3 than iodide or iodine was sorbed from 0.1 M calcium chloride or 1.0 M ammonium acetate solutions. The addition of composted organic matter slowly decreased the solubility of IO_3^- to that of I^- or I_2 ; this, the author speculated, was due to the reduction of IO_3^- by the organic matter to these species.

The differing sorption of iodine species was also witnessed by Wolfsberg [WOLFSBERG-1978] on studies of the sorption of iodine by Nevada Test Site alluvium from alluvium groundwater. The distribution ratio, K_d , for sorption from solution was measured to be 24.5 mL/g, while desorption produced a K_d of 400-900 mL/g. He attributed this difference to there being more than one iodine species in the groundwater; the use of paper chromatography showed the presence of three iodine species: $\text{IO}_4^-/\text{IO}_3^-/\text{I}^-$ in the ratio of 10%/40%/50%. This same author measured the sorption of iodine species by Nevada tuff minerals to be zero [WOLFSBERG-1980].

The high capacity for iodine retention of some organic matter has led researchers to test the use of charcoal and charcoal/clay mixes as backfill materials for disposal of I-129 [IKEDA, NOWAK]. The distribution ratios of iodide between a brine solution and three types of activated charcoals are listed in Table III-28. Based on the data of Table III-28 and assuming a backfill of three feet around the waste container, Nowak has calculated that a backfill of 30% bentonite and 70% activated charcoal (bulk density = $\sim 2 \text{ g/cm}^3$ and effective porosity = ~ 0.1) would have a time before "breakthrough" of 1000 y. "Breakthrough" is defined by the author as being the point when the concentration of I^- leaving the backfill barrier is 1% of that entering it from the waste form. Bentonite has no significant retention capacity for iodide. The mixture suggested by Nowak was made to sorb cationic species as well as anionic iodine and technetium species, based on the questionable assumption that all fission products would be disposed of together.

Ikeda and Tamaka [IKEDA] measured the distribution of iodide and iodate between aqueous phases of various pH values and activated charcoal. Both I^- and IO_3^- sorb on charcoal at pH values lower than 10 for iodide and 9 for iodate. Distribution ratios for this system are presented in Table III-29. The authors speculated that the iodate distribution becomes much like that of iodide at low pH values because iodate is being reduced to form iodide in this pH region.

Fig. III-19

The Effect of pH on the Sorption of Iodide by Soil (o), Root Compost (●), Ferric Oxide (Δ), and Aluminum Oxide (▲) from Solutions Containing Initially 40 µg I/100 mL [WHITEHEAD-1974B]

Fig. III-20. The Effect of Time on the Sorption of Iodide by Two Soils from Initial Iodide Concentrations of (i) 40 µg I/100 mL and (ii) 400 µg I/100 mL: (o) soil 1A; (●) soil 2A; pH = 5.5 [WHITEHEAD-1973A]

Table III-28. Distribution Ratio of Iodide between Activated Charcoal and Brine^a [NOWAK]

Solid Sorbent	K _d , mL/g	pH
Activated "Nuchar"	35	6.6
Activated Coconut Charcoal	53	6.7
Activated "Darco" G-60	18	6.5

^aBrine = 1.8 M NaCl, 0.8 M KCl, 1.4 M MgCl₂, 0.01 M bicarbonate, and 0.02 M borate (simulated brine in potash-containing horizons in the vicinity of the proposed WIPP site).

Table III-29. Distribution of Iodide and Iodate between Water of Differing pH Values and Activated Charcoal [IKEDA]

Iodine Species	pH	Distribution Ratio, mL/g
I ⁻	8.5	10 ²
	5	10 ³
	3.5	10 ⁴
IO ₃ ⁻	9	0.6
	6	10
	4	10 ²
	3	10 ³

In addition to charcoal, several minerals are possible sorbents for iodide and iodate [ALLARD; BIRD; BOVARINOVA-1972; COUTURE; HAQ; NEAL; STEINDLER-1978, -1979A, -1979B, -1979C; STRICKERT-1978, -1980]. The mechanism for these sorbents is not ion exchange but rather chemical reaction. In general, minerals that sorb iodine species are oxides and sulfides of heavy metals that have low solubilities of iodide and iodate salts.

Allard et al. [ALLARD-1980] tested several minerals of this type. In general, in the pH range studied (3-11), sorption of iodide was highest at low pH values and decreased at higher pH values; the exception was lead hydroxide, which showed increased sorption with pH. Bird and Lapata [BIRD] found PbO and Cu metal to be excellent sorbents for iodide. Lead oxide sorbed I⁻ ion at greater than 95% efficiency from mole ratios of 4.5×10^{-10} to 0.2 I⁻/PbO; at mole ratios between 0.2 and 3, the percent of I sorbed dropped dramatically. As I⁻ was removed from solution, the pH of the solution increased. The observed increase in pH is explainable by the following reactions:

X-ray diffraction confirmed that PbIOH exists on the surface of the solid when I^-/PbO mole ratio = 3. At lower ratios, Pb(OH)I was not found; instead, other unidentified species of variable composition were seen.

Haq et al. [HAQ] measured the kinetics and thermodynamics of the sorption of iodide by copper metal, Cu_2O and CuO . Both copper metal and cuprous oxide sorb I^- rapidly from solution of $>10^{-6}$ M I^- (50 mL of solution to 5 g of solid sorbent). The mechanism of sorption for both Cu metal and Cu_2O is the formation of $\text{Cu}^+(\text{I}^-)(\text{OH}^-)$, followed by crystallization of CuI and loss of hydroxide to the solution. Thus, the amount of iodide removed from solution is controlled by the solubility product of cuprous iodide ($K_{sp} = 1.1 \times 10^{-12}$). There is no significant sorption of iodide by CuO . Based on the results of this study, the authors recommend that the waste containers for disposal of ^{129}I be fabricated from copper; if the container is breached, the container material will still continue to limit the migration of iodine species into the environment.

Boyarinova et al. [BOYARINOVA-1972] have studied the coprecipitation of iodate ion with hydroxides. They have found that coprecipitation decreases the iodate concentration more substantially than sorption on previously precipitated hydroxides. The order of effectiveness of the removal of iodate from solution by the hydroxides studied was: $\text{Mn}^{4+} < \text{Sn}^{4+} < \text{Fe}^{3+} < \text{Al}^{3+} < \text{Sn}^{2+} < \text{Ni}^{2+}$. For example, at pH = 8.5, 96% of the IO_3^- present in solution coprecipitated with Ni(OH)_2 ; at this same pH, 75% coprecipitated with Sn(OH)_2 . Coprecipitation only occurred below this pH for tetravalent cations.

Work performed by Couture and Seitz [COUTURE; STEINDLER-1978, -1979A, -1979B, -1979C] showed that hematite (Fe_2O_3) was an effective sorbent for IO_4^- , IO_3^- , and I^- ; the effectiveness of sorption is in the order given. At any given pH, the sorption of periodate was determined to be ~ 4 times higher than that of iodate, and iodide retention was orders of magnitude lower. The data presented in Fig. III-21 show (1) the inverse relationship between solution pH and the degree of sorption of iodate on hematite and (2) the linear logarithmic relationship between the amount of iodate sorbed on the hematite and the aqueous phase concentration of iodate for a concentration range of almost eight orders of magnitude. The linear logarithmic relationship begins to level off when the surface concentration of iodate approaches a saturation value (10^{-4} mol $\text{IO}_3^-/\text{g Fe}_2\text{O}_3$). The sorption of iodate on hematite was found to be reversible by approaching the equilibrium condition from both high and low pH. This was accomplished by allowing the system to equilibrate at one pH before the pH was lowered by the consecutive additions of acids. These measurements were repeated by raising the pH again by consecutive additions of base. This experiment is depicted graphically in Fig. III-22.

These researchers also examined the sorption characteristics of two Pacific pelagic red clays that each contained ~ 5 wt % Fe_2O_3 . On the basis of their Fe_2O_3 content and the sorption behavior of hematite, these clays were

Fig. III-21.

Sorption of Iodate by Hematite (Fe_2O_3). (Γ is amount sorbed, mol/g Fe_2O_3 . $[\text{IO}_3^-]$ is in units of molarity. Squares indicate titrations, and circles indicate radio-metric measurement. [STEINDLER-1979B].)

Fig. III-22.

Adsorption of Iodate by Hematite as a Function of pH in 0.1 M NaClO_4 Solution. (Circles represent consecutive additions of base; squares represent consecutive additions of acid. Numbers are the time intervals after addition of the acid or the base. [STEINDLER-1979A].)

expected to sorb only about one half of the amount of iodate they actually did. This higher retention of iodate was attributed to the probable smaller particle size of Fe_2O_3 in the clays, hence, higher surface area-to-weight ratio of the sorbing species.

Neal and Truesdale [NEAL] studied the sorption of iodide and iodate from fresh and salt water by ferric hydroxide, peat, and kaolinite. Ferric hydroxide was determined to be the best of the three for sorbing iodide from salt water, followed by peat and then kaolinite. Peat differed from ferric hydroxide and kaolinite in that the amount of sorption did not vary between fresh and salt waters.

Strickert et al. [STICKERT-1978, -1980] studied the sorption of I^- and IO_3^- by a long list of minerals and laboratory-grade chemicals. Some of their results are presented in Tables III-30 and III-31. The aqueous phases in these experiments were pre-equilibrated in most cases with the minerals studied. In a few experiments, solutions other than pre-equilibrated aqueous phases were tested. This procedure made little impact on sorption except when simulated WIPP brine was used; as can be seen from the data in Table III-32, the saturated NaCl solution did significantly lower the sorption of IO_3^- on all the minerals tested.

Figure III-23 shows that sorption of iodide ion by sulfide minerals, as for soils, is not an instantaneous process. The difference between soil and cuprous sulfide is the much higher retention of iodide by Cu_2S . These authors found, as did others, that a mineral's anion-exchange capacity is not the important criterion for effective sorption of I^- and IO_3^- ; rather, this capacity was due to coprecipitation or to incorporation of iodide or iodate into the mineral lattice.

The conclusions to be drawn from the literature survey presented above are:

- I-129 will migrate freely in most soils and groundwater once it is leached from the waste form unless special precautions are taken.
- Because the ion-exchange capacity of most materials is very poor, conventional backfill materials such as clay minerals are nearly useless.
- Effective backfill materials for retention of I-129 species can be of two types: (1) organic materials such as activated charcoal or composted organic matter and (2) heavy metals and heavy metal oxides and sulfides that sorb iodide by chemical reaction. The effectiveness of the second type of materials is primarily based on the low solubility products of their iodide and iodate salts.

e. Burial Options for I-129

Some authors [ALTOMARE] do not favor a geological repository because the extremely long half-life of decay for I-129 makes it impossible to predict that the disposal site will remain water free for even a small fraction of the time needed for extensive decay of I-129. This fact, the relatively high predicted leach rates of iodine from concrete, and the low retention of iodine species in most minerals and soils (see above) makes the certainty of

Table III-30. Distribution Ratio (K_d) Values for Various Rocks and Minerals

Material	Major Constituents	K_d , ^a mL/g	
		I ⁻	10 ³ -
Apatite	$\text{Ca}_5(\text{Cl},\text{F})(\text{PO}_4)_3$	<1	-
Anhydrite	$\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$	-	4
Witherite	BaCO_3	-	1
Columbia river basalt	Feldspar, diopside	2	<1
PNL basalt	Feldspar, diopside	-	<1
Chalk	CaCO_3 , clay	<1	5
NTS dolomite	$\text{CaMg}(\text{CO}_3)_2$, SiO_2	<1	<1
Granite	Feldspar, KAlSi_3O_8	<1	<1
PNL granite	Feldspar, SiO_2	-	<1
Low temperature metamorphic	Chlorite, feldspar	-	3
Pierre shale	SiO_2 , feldspar	<1	1
PNL shale	SiO_2 , feldspar	-	9
Mottled siltstone	Montmorillonite	<1	5
LASL tuff	SiO_2	<1	<1
NTS tuff	SiO_2	<1	<1
PNL limestone	CaCO_3 , SiO_2	-	16
Bornite (with molybdenite)	Cu_5FeS_4 (MoS_2)	30-790	10,1000
Bournonite	CuPbSbS_3	200-1000	960
Chalcopyrite	CuFeS_2	190	400-1300
Chrysocolla	$\text{CuSiO}_3 \cdot 2\text{H}_2\text{O}$	3-12	40-75
Enargite	Cu_3AsS_4	130,150	70,120
Galena	PbS	26	3,30,45
Pyrite	FeS_2	300-3000	>1000
Tennantite	$(\text{Cu},\text{Fe})_{12}\text{As}_4\text{S}_{13}$	100-350	10-20
Tetrahedrite	$(\text{Cu},\text{Fe})_{12}\text{As}_4\text{S}_{13}$	1100,1500	1000-3000
Tetrahedrite (argentiferous)	$(\text{Cu},\text{Ag},\text{Fe})_{12}\text{Sb}_4\text{S}_{13}$	4-20,150-680	32

^aThe distribution ratio, K_d , is defined by the following equation

$$K_d = \frac{F_R}{1-F_R} \cdot \frac{V_T}{M_R}$$

where V_T is the total volume of the solution in mL, and M_R is mass of the rock in g; F_R , the fraction of activity sorbed by the rock, is defined by the relationship:

$$F_R = 1 - \frac{A_a}{A_T} \cdot \frac{V_T}{V_a}$$

A_a and V_a are the activity and the volume of the aliquot removed from the solution, and A_T is the total activity put into the solution. For the conditions used in these experiments, a K_d of <1 indicates that <5% was sorbed from the solution; a K_d of 10 indicates ~50% and a K_d of 100 indicates that 79% of the tracer was sorbed from the solution.

Table III-31. Fraction of Radiotracers Sorbed on Various Chemical Reagents

Chemical	F_R^a	
	I^-	IO_3^-
CdS	0.97	-
Cu ₂ S	0.99+	0.99+
CuS	0.99+	0.99+
FeS	-	0.14
HgS	0.70	-
PbS	0.94 ± 0.05	0.95 ± 0.04
ZnS	0.10	-
CuCl	0.56	0.59
CuCO ₃ ·Cu(OH) ₂ ^b	0.56	0.59
CuO	0.07	0.7 ± 0.2
Fe	0.01, 0.4	0.09
FeCl ₂ ·4H ₂ O	0.18	-
Fe ₂ O ₃	0.61	-
FeSO ₄	0.09	-
Pb	0.97	-
PbO ₂	0.01	-
Ag powder	0.99+	-

^aSee footnote on Table III-30.^bFor runs of shorter duration (few hours), greater sorption for iodide (0.97) was observed.

Table III-32. Sorption of Iodate by Several Materials from Groundwater and Simulated WIPP Brine

Material	Equilibrated Solution	Rock Mass, g	F_R^a IO_3^-
Bornite	Groundwater	0.717	0.98
	WIPP solution	0.712	0.53
Chalcopyrite	Groundwater	0.370	0.97
	WIPP solution	0.454	0.28
	Groundwater	0.522	0.99
	WIPP solution	0.647	0.41
Enargite	Groundwater	0.873	0.97
	WIPP solution	1.283	0.79
Galena	Groundwater	0.813	0.50
	WIPP solution	0.819	0.04
Tetrahedrite	Groundwater	1.062	>0.99
	WIPP solution	1.268	0.82

^aSee footnote on Table III-30.

Fig. III-23. Sorption of Iodide Tracer Activity on Cu_2S and Bournonite as a Function of Time [STRICKERT-1980].

isolating I-129 from the natural environment very low. The impact of I-129 entering the biosphere is, of course, dependent on the rate of its release. If the rate of release can be assured to be low enough that equilibration with a large body of water is assured, the impact will be small.

Wood and Richardson have done a calculation for deep geologic disposal, based on (1) a "typical" concrete waste-form cylinder (3 m long by 0.3 m in diameter) containing 6 wt % I-129 and (2) the ORNL leaching rate [WOOD]. If the total radioiodine waste for 20 y of operation from a 1500 MTHM/y reprocessing plant (600 kg of iodine/y) were buried in a repository in such concrete cylinders, the amount of iodine leached the first year would be 4.9×10^5 g (88 Ci). If this material finds its way into an aquifer with a typical flow rate of 200 cfs (or 16×10^{13} cc/y), the concentration of I-129 would be 5×10^{-7} mCi/L. This concentration is somewhat greater than the maximum permissible concentration (MPC) of soluble I-129 set by 10 CFR 20 of 6×10^{-8} mCi/L [BROWN-1980].

Seabed disposal is an attractive alternative to continental disposal, primarily because of the effect of isotope dilution of I-129 with natural iodine I-127 in sea water [MORGAN, BURGER-1980]. The concentration of natural iodine in the ocean, 4.9×10^{-5} g/L, is 145 times greater than

the MPC of I-129 (6×10^{-8} mCi/L = 3.4×10^{-7} g/L) and, therefore, would reduce the specific activity of I-129 even at a concentration as large as the MPC by a factor of 145. The importance of lowering the specific activity of I-129 can be seen by looking at the distribution of iodine in the body and the maximum permissible body burden of I-129. The major collection point of iodine in the body is the thyroid gland, which contains ~ 7 mg of iodine. The maximum permissible burden of I-129 is 0.6 μ Ci or 3.4 mg of iodine. Thus, for a person to receive a maximum permissible burden of I-129, approximately 50% of the iodine content in his body, which for all practical purposes is 50% of his thyroid iodine content, would need to be I-129. Diluting I-129 with natural iodine, even as little as by a factor of two, makes it impossible to exceed the maximum permissible burden. Dilution by a factor of 145 would make the maximum possible burden only 1.4% of the maximum permissible burden. Isotopic dilution of all the I-129 collected from nuclear fuel reprocessing through the year 2020 by all the natural iodine in the oceans (dilution by seven to eight orders of magnitude) would make the specific activity of iodine so low as to be inconsequential.

To assure mixing of I-129 with natural iodine, low leaching rates of iodine into the sea must be assured. The principal barriers to transport of iodine from the waste form would be about the same as with dry land burial [WOOD]:

- (1) the waste matrix,
- (2) the container,
- (3) the geological isolation (basement rock or sediment),
- (4) soil adsorption.

Because of the possibility of isotopic dilution by natural iodine in sea water, however, the criteria for leaching retardation are not as stringent.

E. Technologies of Carbon Waste Forms

1. Decay Characteristics of C-14-Containing Material

Carbon-14 has a half-life of 5730 ± 40 y by β^- decay. The maximum β^- decay energy is 156.08 ± 0.24 keV; the average β^- decay energy is 49.30 ± 0.10 keV [MARTIN]. Table III-33 shows the specific activities and thermal outputs of various chemical forms of C-14. As shown, the thermal output of C-14 is 13×10^4 W/g (2.91×10^{-4} W/Ci).

Because the collection system for $^{14}\text{CO}_2$ in the LWR-reprocessing plant also collects natural CO_2 in the plant atmosphere, there will be a great deal more natural, unradioactive CO_2 than radioactive $^{14}\text{CO}_2$ collected. The ratio of $^{12}\text{CO}_2/^{14}\text{CO}_2$ collected was estimated by [ALTOMARE] to be in the range of 10 to 100. Brown et al. [BROWN-1980] estimated that a caustic scrubber would produce 1.6 MT of total calcium carbonate to trap the 1.8 kg of $\text{Ca}^{14}\text{CO}_3$ produced in a typical reprocessing plant (C-12/C-14, ~ 900). Once the slurry from the scrubber is mixed with cement and formed into concrete, the initial 200 g of C-14 is diluted to 65 MT of concrete, a dilution factor of $\sim 32,000$ times. The specific activity of the final waste form would be ~ 13 Ci/MT or 22 Ci/m³.

Table III-33. Specific Activities and Thermal Outputs of ^{14}C Chemical Species

Chemical Species	wt % C-14	Density, g/cm^3	Specific Activity		Thermal Output ^a		Amount to Contain 850 Ci	
			Ci/g	Ci/cm ³	$\frac{\text{W/g}}{\times 10^4}$	$\frac{\text{W/cm}^3}{\times 10^4}$	kg	dm ³
C-14	100	--	4.46	--	13.0	--	0.191	--
$\text{Ca}^{14}\text{CO}_3$	14	2.74 ^b	0.624	1.72	1.82	4.99	1.82	0.494
$\text{Ba}^{14}\text{CO}_3$	7.0	4.33 ^c	0.312	1.35	0.91	3.94	2.72	0.644
$^{14}\text{CO}_2$	30.4	--	1.36	--	3.96	--	0.62	--

^aThe estimated amount of ^{14}C produced from a 1500 MTHM/y reprocessing plant is 850 Ci of $^{14}\text{C/y}$ [ALTOMARE].

^bBased on the density of the more stable form, calcite ($d = 2.711 \text{ g/cm}^3$) [DEAN], and the higher molecular weight of $\text{Ca}^{14}\text{CO}_3$. (The less stable form, aragonite, has a density of 2.93 g/cm^3 .)

^cBased on the density of witherite ($d = 4.29 \text{ g/cm}^3$) [DEAN] and the higher molecular weight of $\text{Ba}^{14}\text{CO}_3$.

2. Collection of Carbon

A critical review of collection methods has been presented by Brown et al. [BROWN-1981]. These methods are also discussed by Altomare et al. [ALTOMARE]. They will be briefly discussed below, with an emphasis on the amount and form of the collection product. Other references pertinent to each collection method are listed after the subsection headings below.

a. Caustic Scrubbing [CROFF-1976; BRAUN-1980, -1983; NOTZ-1980A, -1980B]

Absorption of CO_2 by a sodium hydroxide solution in a packed scrubbing column is a common industrial process. Although this process is untested as a means to trap $^{14}\text{CO}_2$, there is no obvious reason why it should not work effectively. Braun describes this process in detail in a recent publication [BRAUN-1983]. Figure III-24 is a schematic of this process. As the sodium hydroxide is converted to soluble sodium carbonate, the scrubbing solution is removed to a mixing tank where it is contacted with calcium hydroxide. Precipitation of the insoluble calcium carbonate regenerates the sodium hydroxide, which is returned to the scrubbing column. The carbonate cake is incorporated in cement for long-term disposal.

The potential difficulties of this method are as follows: (1) multiple and difficult steps would be required from collection to packaging; (2) decontamination from the scrubber solution or filter cake of other airborne radioactive wastes would be difficult; (3) NO_x species would deplete the caustic solution; and (4) approximately 200 g of C-14 would be disposed of in 65 tons of concrete (a great deal of waste generation for 850 Ci of radioactive material) [BROWN-1980].

b. Caustic Slurry Scrubbing [CROFF-1976; NOTZ-1980A, -1980B; HOLLADAY-1979; CHEW; PATCH; SHEPPARD]

Caustic slurry scrubbing is similar to the above process in that it involves caustic scrubbing with CaCO_3 as its product, but it is different in that $^{14}\text{CO}_2$ is contacted directly with an aqueous slurry of calcium hydroxide (or barium hydroxide) in a stirred cell contactor. Moreover, no NaOH would be present in the CaCO_3 cake; $\text{Ca}(\text{OH})_2$ would be present instead. Figure III-25 is a schematic of the caustic slurry scrubbing process. Advantages and disadvantages for this process are similar to those of caustic scrubbing. The amount of CaCO_3 produced would be essentially identical for both processes.

c. Molecular Sieve Adsorption [NOTZ-1980A, -1980B]

A molecular sieve bed would collect both CO_2 and Kr-85 from an NO_x -depleted, dry off-gas stream. Experiments under way to separate Kr-85 from $^{14}\text{CO}_2$ are based on the fact the CO_2 is more energetically bound to the Linde type 5A molecular sieve used in the sorption bed. Once the CO_2 is separated from Kr-85, it will be removed from the gas stream by caustic scrubbing and solidified as CaCO_3 . Use of a molecular sieve results in the concentration of CO_2 in the off-gas stream being much higher than that of the original gas stream; this condition may allow a scaledown of the caustic scrubbing process.

Fig. III-24. Schematic of the Double Alkali CO_2 Fixation Process [CROFF-1976].

However, the molecular sieve complicates the collection process further. The amount of CaCO_3 waste generated would be the same as for the processes described above. A more detailed description of this process can be found in Section III.A.2.d.

d. Fluorocarbon Absorption [STEPHENSON-1976, -1977; LITTLE]

Fluorocarbon absorption was primarily designed for the capture of Kr-85 , but does effectively trap CO_2 simultaneously, with lesser amounts of Xe , Ar , O_2 , and N_2O also being collected. These gases are absorbed from the off-gas stream at pressures of 6 to 20 atm. When the pressure is dropped to 1 atm, the desorbed gases are separated into high-boiling fractions (CO_2 , Xe , N_2O) and low-boiling fractions (Kr , Ar , O_2); the CO_2 is collected from the high-boiling fraction by molecular sieves and is subsequently converted to a CaCO_3 waste form. The amount of CaCO_3 waste collected from the process is similar to that for the collection processes described above. A more detailed description of this process can be found in Section III.A.2.c.

e. Adsorbent Bed Fixation [HAAG-1981A, -1981B, -1983; FERGUSON; NOTZ-1980A, -1980B; HOLLADAY-1978]

The major advantage to adsorbent bed fixation is its simplicity. As CO_2 passes through a bed of $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ chips, it is simultaneously sorbed

and immobilized as BaCO_3 :

The process is capable of decontamination factors >1000 and $>99\%$ transformation of $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$ to BaCO_3 .

Fig. III-25. Schematic of the Direct CO₂ Fixation Process [CROFF-1976].

Because the molecular weight of BaCO_3 is twice that of CaCO_3 , the process will develop twice as much (by weight) solid carbonate waste. However, because the density of BaCO_3 is higher than that of CaCO_3 and because there is no aqueous slurry to contend with, the amount of concrete waste produced would be 5-10 times less than that produced by caustic scrubbing; BaCO_3 is even more thermally and chemically stable than CaCO_3 and has a somewhat lower aqueous solubility.

From laboratory-scale investigations, Haag *et al.* [HAAG-1983] recommend process conditions that involve a "window" bounded by a maximum relative humidity of 60% and a minimum relative humidity determined by the dissociation pressure of $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$. Perhaps a good choice of conditions, based upon the data presented by [HAAG-1983], is a temperature of about 20°C and a relative humidity of about 45%.

The removal of CO_2 from mixtures with gaseous nitrogen has been investigated for possible application to the cleanup of CANDU reactor cooling systems [CHEH]. This process involves reaction of CO_2 with a packed bed of solid $\text{Ca}(\text{OH})_2$. The results were concluded to be promising for application. Sufficient information to allow comparing $\text{Ca}(\text{OH})_2$ -adsorbent-bed fixation against the other collection processes described here is not available, however.

3. Immobilization of Carbon

a. CaCO_3 and BaCO_3 in Concrete

(1) Solubility Data

The carbonate salts of barium ion and calcium ion have low aqueous solubilities. Solubility data for these two salts measured in ambient atmosphere by several researchers are presented in Fig. III-26; the lines through the points were drawn by eye to show the unusual retrograde temperature dependence of CaCO_3 solubility and the more-typical temperature dependence of BaCO_3 solubility. The scatter of data for solubility measurements by different experimenters may be explained by different aqueous concentrations of carbon dioxide. Because of the importance of the reaction

to the solubility of carbonate salts, the atmosphere in which their solubility is measured is very important. The effect of CO_2 partial pressure on calcite solubility is demonstrated in Fig. III-27.

Fig. III-26.

Solubility of CaCO_3 and BaCO_3
under Ambient Atmosphere vs. Temperature [GMELIN-1960, -1961; LINKE;
STEPHEN; SIDGEWICK].

Fig. III-27. The Solubility of Calcite vs. the Partial Pressure of CO₂ above the Solution at 25°C [LINKE].

(2) Thermal Stability

The thermal decomposition of both BaCO₃ and CaCO₃ occurs by the same process:

The temperature needed to initiate thermal decomposition to the oxide is, therefore, dependent on the partial pressure of carbon dioxide [DUVAL, SIDGEWICK, LIPTAY]. Dry CaCO₃, as calcite, begins to lose CO₂ at about 500°C and is converted to 100% CaO at a temperature between 680 and 810°C, depending on the rate of heating and the mass of the sample [DUVAL]. In a CO₂ gas flow, temperatures at the onset of decomposition can be increased to ~900°C.

At temperatures up to 1000°C, BaCO₃ is stable to decomposition. Its natural mineral form, witherite, began to show signs of CO₂ loss at 1075°C in air and 975°C in a N₂ stream [DUVAL]. If the witherite is kept under high CO₂ pressure to stop its thermal decomposition, a melting point of 1740°C is observed [SIDGEWICK]. According to Duval [DUVAL], BaCO₃ formed by precipitation from aqueous solution retains water tenaciously. Although 99.6% of the water was removed by heating to 400°C, the remainder was not removed until further heating to 681°C. There is no weight loss of dry BaCO₃ between 20 and 1000°C.

(3) Leaching of C-14 from CaCO_3 and BaCO_3 in Concrete

Two sets of leach tests have been performed at PNL by Scheele et al. [SCHEELE-1982, PLATT-1979B, -1979C; CHIKALLA-1982]. In the first set of experiments, the objective was to measure, by a Plackett-Burman experimental design, the effect of various parameters on the leach rate of $^{14}\text{CO}_3^{2-}$ from a concrete monolith. These parameters are listed below in the relative order of importance to C-14 leaching. The numbers in parentheses are "factor effects." The larger the magnitude of the factor effect, the greater is the influence of that parameter on the leach rate. A positive sign means an increase in this parameter increases the leach rate; a negative sign indicates an inverse relationship between the parameter and the leach rate. The parameters studied were:

- (1) initial carbon dioxide concentration in the leachate ($+7.45 \times 10^{-3}$),
- (2) concentration of the carbonate in the concrete ($+1.59 \times 10^{-3}$),
- (3) concentration of strontium nitrate added to the concrete (-1.44×10^{-3}),
- (4) concentration of barium nitrate added to the concrete (-1.37×10^{-3}),
- (5) solubility of the carbonate salt added to the concrete ([SCHEELE-1982], SrCO_3 , solubility = 10^{-5} M; K_2CO_3 , solubility = 8.1 M) ($+4.19 \times 10^{-4}$), and
- (6) addition of sodium resinate--an air entraining agent that changes the pore structure of the concrete--to the concrete ($+5.51 \times 10^{-5}$).

All of the factors above had a measurable effect on the leach rate of the carbonate except for sodium resinate addition, whose effect was considered negligible. The experimental conditions and fraction of carbonate leached after 25 d are presented in Table III-34.

In a second set of experiments, a static leach test was performed on CaCO_3 in portland type III cement (2.5 mmol CaCO_3/g dry cement) in different leachates (distilled water, river water, sea water). These data are shown in Fig. III-28. The drop in $^{14}\text{CO}_3^{2-}$ in solution with time for experiments #3 and #4 is attributed to isotopic dilution of $^{14}\text{CO}_3^{2-}$ by CO_2 from the atmosphere in those basic solutions.

In Fig. III-29 are the comparable IAEA leach studies for BaCO_3 and CaCO_3 in concrete (both at a concentration of 2.5 mmol $\text{CO}_3^{2-}/\text{g}$ dry cement). The initial leaching rate of the two barium carbonate samples was much higher than that of the two calcium carbonate samples. After 5 d, the ratio of the leach rate of BaCO_3 in concrete to that of CaCO_3 in concrete dropped to a factor of 2; the leach rates were determined to be 7.2×10^{-6} cm/d for BaCO_3 and 3.9×10^{-6} cm/d for CaCO_3 .

Table III-34. Summary of Carbonate Leaching Experiments [PLATT-1979C]

Trial	Car- bonate Salt Solubility, <u>M</u>	CO ₂ in Leachant	Addition, g/g Dry Cement				Cumulative Fraction Carbonate Leached After 25 d, cm
			Car- bonate	Stron- tium Nitrate	Sodium Resonate	Barium Nitrate	
1	10 ⁻⁵	Ambient	0.15	0.14	0.01	0.03	2.54E-5
2	10 ⁻⁵	Saturated	0.03	0.14	0.01	0	6.70E-3
3	8.1	Saturated	0.15	0.14	0	0	8.55E-3
4	10 ⁻⁵	Saturated	0.15	0	0	0	1.19E-2
5	10 ⁻⁵	Ambient	0.15	0	0	0.03	2.25E-4
6	10 ⁻⁵	Ambient	0.03	0	0.01	0	5.09E-4
7	8.1	Ambient	0.03	0.14	0	0.03	4.19E-4
8	8.1	Saturated	0.03	0	0.01	0.03	8.86E-3
9	8.1	Ambient	0.15	0.14	0.01	0	1.41E-3
10	10 ⁻⁵	Saturated	0.03	0.14	0	0.03	3.69E-3
11	8.1	Saturated	0.15	0	0.01	0.03	7.79E-3
12	8.1	Ambient	0.03	0	0	0	1.80E-4

Fig. III-28. C-14 Static Leach Studies of Concrete-Encapsulated CaCO_3 (2.5 mmol CaCO_3/g dry portland cement) [SCHEELE-1982].

Fig. III-29. C-14 IAEA Leach Studies of Concrete-Encapsulated BaCO_3 and CaCO_3 (2.5 mmol C/g dry Portland Cement) [SCHEELE-1982].

b. Alternative Carbon Waste Forms

No satisfactory alternatives to the waste forms described above are presently being considered for C-14 long-term disposal. Croff [CROFF-1976] looked at the possibility of disposal of CaCO_3 or BaCO_3 with no matrix in 55-gal steel drums or disposal of $\text{Ca}^{14}\text{CO}_3$ in grouts without containers, but rejected these options because of their low resistance to leaching of $^{14}\text{CO}_3^{2-}$.

c. Migration of Carbonate in Geological Formations

Allard et al. [ALLARD-1981] studied the sorption of $\text{H}^{14}\text{CO}_3^-$ from simulated granitic groundwater (pH = 7.2-8.5) by various common minerals and cement. Most groundwaters have concentrations of carbonate plus bicarbonate in the range of 1-6 mM (60-100 mg/L) and are slightly undersaturated in respect to $\text{MgCO}_3(\text{s})$ and $\text{CaCO}_3(\text{s})$. The following equilibria are important to the concentration of carbonate and bicarbonate in groundwaters and to the sorption of C-14 from these groundwaters:

where K is the equilibrium constant for the reaction of the aqueous soluble species, and K_s is the solubility product. The sorption of HCO_3^- on the materials listed in Table III-35 is explainable by their calcium ion content. The K_d values measured for these materials were far smaller than were measured for the cationic sorption of Cs^+ and actinides. These results can be explained by (1) an increase of pH as basic materials (e.g., cement) dissolve to precipitate CaCO_3 or (2) cationic exchange of Na^+ ion in solution for Ca^{2+} ion in minerals (e.g., bentonite), thereby increasing the Ca^{2+} ion availability and precipitating CaCO_3 . The results suggest that a container of concrete around the concrete-encapsulated CaCO_3 or BaCO_3 would greatly decrease carbonate migration from the waste form.

Table III-35. Distribution Coefficient (K_d) vs. Contact Time

Solid	K_d , mL/g							
	0.5 h	2 h	6 h	24 h	3 d	1 wk	5 wk	6 mo
Stripa granite					0	0	0	0
Na-montmorillonite					0	0	0	0
Bentonite/quartz (10/90 wt. ratio)					2.8	8.6	7.6	7.8
Sandy moraine	1.0	0.2	0.7	0.5	0.8	1.1	2.6	2.2
Clayish moraine	1.9	0.7	1.3	1.4	1.3	2.0	3.0	2.3
Calcite	1.0	1.0	1.5	1.7	3.7	3.5	9.0	83
Concrete					5.3	1600	>10 ⁴	
Cement paste					7.4	1600	>10 ⁴	

IV. SYSTEMS FOR PACKAGING, TRANSPORTATION, AND DISPOSAL

In the preceding sections, a review was presented of the processes for collection of H-3, I-129, C-14, and Kr-85 from streams of fuel reprocessing plants and the processes for conversion of the collected wastes into forms with physical and chemical properties that promise to be suitable for final disposal. The following sections review the possibilities for packaging these waste forms to meet the requirements of transport regulations and the technical criteria for final disposal.

A. Shallow-Land Burial (SLB)

1. SLB Practices

Conventional modes of LLW disposal are examined here for later analysis of whether they would be suitable for disposal of the four airborne wastes. The conventional mode of disposing of solid LLW in the U.S. is shallow-land burial, which is distinguished from geologic disposal by the proximity of emplaced waste to the surface of the earth. In brief, the method usually consists of excavating to create a series of trenches and placing the packaged waste into them. When a trench is filled with waste, it is back-filled with soil and capped to enhance water runoff.

Commercial SLB facilities exist at six sites: West Valley, NY; Morehead, KY; Sheffield, IL; Beatty, NV; Barnwell, SC; and Richland, WA. Only the last three are receiving wastes now [DOE/NE-0015]. In addition to these commercial facilities, several facilities are operated by DOE to accommodate wastes generated by DOE and its contractors. Information on site, location, and status of each of these sites is listed in Table IV-1 [DOE/NE-0015].

This method of disposal has generally been practiced with no intention of ready retrievability of the waste at a later date. Furthermore, before the drafting of 10 CFR 61, the responsibility of containment was assumed to be that of the soil, not the vessel surrounding the waste form [WHEELER]. More recently, a task force has recommended that an evaluation be done of packaging as containment barriers [NUREG-0217]. Wastes in shallow-land burial are subject to water infiltration and movement, erosion, plant root uptake, animal penetration, and human intrusion. Of all these potential radionuclide-transport mechanisms, water has been traditionally considered to be the principal vehicle by which radioactivity can migrate away from the trenches. More-recent investigations, however, indicate that the chief contribution to radioactivity exposure in the biosphere may be from (1) diffusion of radionuclides to the surface and resuspension of dust, which is ultimately inhaled [HUNTER], or (2), at particularly humid and arable sites, plant root uptake and ingestion of the contaminated food [HOOKER]. Both diffusion and plant root uptake are functions of the presence of water and the degree of saturation in the soil regions surrounding the waste. Good site selection and design practices should avoid buried waste standing in contact with underground water and producing a leachate that will later migrate away. It is particularly important to avoid the entry of water into the trench and to minimize the possibility of subsurface flow away from the trench, especially along fractures. A number of criteria or site characteristics are considered in the selection of a site and in the design of an effective disposal facility.

Table IV-1. DOE Sites for LLW Disposal

Site	Location	Status
Los Alamos National Laboratory ^a	Los Alamos, NM	Active
Brookhaven National Laboratory ^a	Brookhaven, NY	Inactive
Sandia National Laboratories ^a	Albuquerque, NM	Active
Idaho National Engineering Laboratory ^a	Idaho Falls, ID	Active
Nevada Test Site ^a	Las Vegas, NV	Active
Feed Materials Production Center ^a	Fernald, OH	Active
National Lead Company of Ohio	Niagara Falls, NY	Inactive
Oak Ridge Gaseous Diffusion Plant	Oak Ridge, TN	Active
Oak Ridge National Laboratory	Oak Ridge, TN	Active
Oak Ridge Y-12 Plant ^a	Oak Ridge, TN	Active
Paducah Gaseous Diffusion Plant	Paducah, KY	Active
Portsmouth Gaseous Diffusion Plant	Piketon, OH	Active
Weldon Springs	St. Charles County, MO	Inactive
Richland Site ^a	Richland, WA	Active
Savannah River Plant ^a	Aiken, SC	Active
Lawrence Livermore National Laboratory ^a	Livermore, CA	Active

^a DOE sites with significant national security responsibilities [DOE/NE-0015].

A desirable feature of all sites would be a deep water table. Soil regions that have either high, low, or alternating high and low permeability may be desirable, depending on topography and climatic conditions at a site. Some sites in the humid eastern U.S. may require a low-permeability (e.g., bentonite) cover [HOOKER] or may require the trench or pit to be sloped at one end, with water collection facilities at the low end. Thus, SLB water control techniques are important, and site selection relies greatly on information about the hydrogeology of the site.

In a DOE study [DOE/NE-0015], a typical SLB facility is described as consisting of shallow trenches from 30 to 240 m long, 6 to 12 m wide, and 6 m deep. After a trench is filled with waste, it is covered with 1 to 3 m of earth and topped with a mounded cap to encourage water drainage. Most trenches also contain a sump to monitor water levels and permit pumping if necessary. After the trench is covered, vegetation is usually planted to control erosion. Permanent markers at each trench indicate its boundaries and record the total volume and radioactivity of buried waste, as well as the date on which the trench was closed. The site is enclosed by a security fence and is usually surrounded by a buffer zone in which access is controlled and recorded. Records on wastes received, personnel exposure, and disposal operations are maintained at each site. After a site has been filled, stabilized, and closed, an extended-care plan with regard to environmental monitoring and maintenance is put into effect. Environmental monitoring is used to determine whether the site is performing as predicted and is safely containing the radioactivity in the waste. Corrective measures would be applied if necessary. Maintenance includes various measures to ensure that the integrity of trench caps is maintained. Site security is also maintained for the duration of the extended care.

An NRC study [NUREG-0782] also describes a reference SLB facility. The reference facility is sized to accept a relatively large quantity of waste, i.e., $50,000 \text{ m}^3$ of waste per year over a 20-year operating life, or a total volume of 10^6 m^3 . This volume corresponds to approximately one-quarter of the total volume of LLW projected in the United States to the year 2000. Disposal of 10^6 m^3 of waste in the reference facility will require about 150 acres of land, which corresponds to the approximate upper bound of the land area for current commercial disposal facilities. The disposal area at the reference facility includes 58 disposal trenches with dimensions of 180 m (591 ft) long, 30 m (100 ft) wide, and 8 m (26 ft) deep. Support facilities and structures at the site include (1) an administration building, (2) a health physics/security building, (3) a warehouse, (4) a garage, (5) a waste activities building,* and (6) a storage shed. All structures at the site are one-story metallic structures on concrete pad foundations. Shipments of radioactive waste arrive by truck and are processed at the site on a first-come, first-serve basis. Arriving shipments are inspected for compliance with applicable Federal regulations and waste acceptance criteria established as conditions in the disposal facility license. Waste is randomly emplaced in the trench, sometimes using cranes and forklifts, and the trench backfilled with dirt removed during trench excavation. Random waste emplacement results in a trench-volume-use efficiency of about 50%. Waste is emplaced to within

* Presumably for handling wastes generated at the site and any repackaging that may be necessary.

one meter of the top of the trench. Dirt is then backfilled into the trench until the trench cover approximately corresponds to the original grade of the site surface. A 1-m-thick earthen cap is then heaped upon the backfill in the shape of a mound. The earthen cap is covered with natural overburden material as necessary to provide good drainage characteristics and to comply with the final contours planned for the site surface. The overburden is reseeded to promote growth of a short-rooted grass cover. After a 20-year operating period, closure (decommissioning) of the facility, which is assumed to require approximately one to two years, involves dismantling and decontamination of site buildings, disposal of wastes produced during dismantlement and decontamination operations, and final site seeding and contouring. The licensee also makes a final survey of the disposal area to make sure that direct radiation levels are at essentially background levels. Following closure, the disposal license is terminated and control of the site is transferred to the site owner. For this reference facility, the site owner is assumed to be a state agency which carries out an active institutional control program of surveillance, monitoring, and maintenance for 100 y.

Costs for shipping and disposal of LLW have been estimated to be [LEVIN-1981] \$200-300 per 55-gal drum ($\sim 27\text{-}40$ \$/ft³). Fees for disposal alone are estimated [LEVIN-1982] to be \$425-700/m³ ($\sim 12\text{-}20$ \$/ft³), with additional surcharges for handling and disposal of packages with external radiation levels in excess of 200 mrem/h.

2. Local Regulatory Authority for LLW Disposal

In the past, NRC regulated the activities at all LLW disposal sites, except in cases where NRC had relinquished its authority to "agreement states" [ERDA-76-43A]. Among these states, regulations varied greatly. The current DOE philosophy on management of wastes at DOE facilities, expressed in [DOE-5820], is to provide general guidance for the implementation of the requirement that DOE field organizations develop site-specific guidelines for disposal operations under their control.

3. Federal Regulation of LLW Disposal

Probably the most significant recent developments in Federal actions concerning LLW disposal are the Low-Level Radioactive Waste Policy Act [PL-96-573] of December 22, 1980, and the addition of part 61 to Title 10 of the Code of Federal Regulations [10 CFR 61].

a. Low-Level Radioactive Waste Policy Act

The LLRWPA instructed the Secretary of the DOE to carry out four actions: (1) define the capacity of existing facilities; (2) review the adequacy of all commercial LLW disposal facilities and make any recommendations necessary to ensure public health and safety; (3) estimate regional transport requirements for LLW shipments, make an inventory of expected types and quantities of LLW, and judge the ability of generators, shippers, and carriers to meet requirements; and (4) evaluate the capabilities of DOE LLW disposal sites to provide interim storage for commercial wastes.

The LLRWPA declares that each state must provide for the disposal of commercial LLW generated within its borders. It suggests that wastes can be most safely and efficiently managed on a regional basis.

The response of the DOE to the instructions of the LLRWPA is as follows [DOE/NE-0015]. Storage of the LLW expected through the year 2000 will require additional disposal capacity. Indeed, within 10 years, there will be a need for 5-7 sites, in addition to the three that are now in operation. The technology for safe disposal by SLB is now available, and lessons learned from practice in SLB since 1962 will enhance its safety. The use of regional sites should reduce distances between waste source and final disposal site, costs of waste management, and risks of radiation exposure to the public up to 50%. Temporary storage of commercial LLW at DOE LLW disposal facilities is possible, at costs of up to \$2000/m³, depending on measures needed to control environmental impacts and to assure protection of national security. Of course, the passage of legislation that would authorize such disposal is required.

Although NRC has issued technical guidelines for SLB in 10 CFR 61, the task of setting a standard for exposure from SLB falls under the purview of EPA. The progress of EPA in developing a standard for the environmental impact of LLW disposal was reviewed by Galpin [GALPIN]. He acknowledged that a number of Federal and State agencies are anticipating the EPA standards and that the EPA has been given the responsibility for setting them. The EPA sees several issues that must be resolved in the development of the standard: (1) determination of a limit of exposure to the general public; (2) determination of whether to set a de minimus* level, below which disposal does not require isolation from the environment; (3) determination of what form the standard should be expressed in, e.g., annual maximum individual exposure, lifetime dose commitment, release rate, or total release; and (4) determination of whether the standard should apply to all facilities or only to new facilities. To help EPA make these determinations, ORNL is making a risk assessment for near-surface disposal and TRW Inc. is making a risk assessment, under contract to EPA, of eight methods for disposal of LLW. The EPA plans to investigate the question of how technology can reduce the risk of LLW and what this technology will cost. The agency also plans to determine the economic impact of whatever standard it recommends. All of these analyses are to be presented in a draft EIS to support the proposed standard. Following public hearings on this matter, the EPA will issue a final standard.

* Presumably a shortening of the Latin phrase, "de minimus non curat lex," meaning "the law is not concerned with trifles".

b. NRC Regulations [10 CFR 61]

Wastes addressed by 10 CFR 61 are low-level wastes containing source,* special nuclear,[†] or by-product material[‡] acceptable for disposal in a land-disposal facility. Furthermore, low-level waste has the same meaning as that described in the Low-Level Radioactive Waste Policy Act [PL 96-573], i.e., material not classified as high-level radioactive waste, TRU waste, spent nuclear fuel, or by-product material as defined in Section 11e(2) of the Atomic Energy Act.

The main items contained in 10 CFR 61 include: (1) procedures, terms, criteria, and conditions upon which NRC will issue a license for a near-surface disposal facility and also a number of additional rules not directly related to the question of whether airborne wastes would be accepted or appropriately disposed of in this type of facility; (2) performance objectives for a near-surface disposal facility, including protection of operators, protection of general population, and preservation of the stability of the disposal site; (3) technical requirements intended to assure achieving the performance objectives listed below. The information most important to the analysis of the acceptability of airborne wastes is contained in the last two items--performance objectives and technical requirements.

The performance objectives are (1) protection of operators through compliance with the standards for radiation protection set out in [10 CFR 20]; (2) protection of the general population through compliance with the rules that an annual dose, from releases, for any member of the public must not exceed 25 mrem to the whole body, 75 mrem to the thyroid, and 25 mrem to any other organ; and (3) preservation of the long-term stability of the site by eliminating the need for ongoing active maintenance after closure. This last stipulation will have a direct bearing on the choice of packaging for any waste to be disposed of in a near-surface facility.

The important technical requirements are expressed in a waste classification system based on radioactive concentrations and on a listing of acceptable waste characteristics.

The classification of wastes in 10 CFR 61.55, by radioactivity concentration, is intended to provide criteria for deciding the treatment to

* Source material is (1) uranium or thorium, or any combination thereof, in any physical or chemical form or (2) ores that contain by weight one twentieth of one percent (0.05%) or more of uranium, thorium, or any combination thereof. Source material does not include special nuclear material.

[†] Special nuclear material is (1) plutonium, uranium-233, uranium enriched in the isotope 233 or in the isotope 235, and any other material which the NRC, determines to be special nuclear material, or (2) any material artificially enriched by any of the foregoing.

[‡] By-product material is the tailings or wastes produced by the extraction or concentration of uranium or thorium from any ore processed primarily for its source material content, including discrete surface wastes resulting from uranium solution extraction processes.

be given a waste, namely, its burial depth, location within the facility, waste form, and packaging. For fuel cycle wastes, economic considerations are likely to result in the production of wastes with high specific activity; therefore, the most important items for the present discussion are the specifications of maximum concentration, reproduced in Table IV-2. Since the regulation states that concentrations can be averaged over the volume of the package, the concentration limit can be multiplied by the volume of the package to determine the allowable total quantity of radioactivity per package. In the case of a 55-gal drum, a package commonly used in near-surface disposal (volume $2 \times 10^5 \text{ cm}^3$), the specific activity values, in units of $\mu\text{Ci}/\text{cm}^3$, can be multiplied by 2×10^5 to estimate the total quantity of an isotope permitted per drum. Furthermore, estimates of the production rate of the isotopes, in units of $\text{Ci}/\text{GW}(\text{e})\text{-y}$, can be used with the specific activities in this table to calculate the amount of disposal space, or number of packages of chosen volume, required annually if airborne wastes are routed to a near-surface disposal facility and if all isotopes are available at acceptable concentrations. For I-129 and C-14, however, concentrations in the waste forms developed by R&D programs may be higher than the acceptable levels set by disposal facilities. Recent information (see footnote a of Table IV-2) indicates that specifications for Kr-85 from fuel reprocessing may be set at some future time.

Table IV-2. Maximum Concentrations of Airborne Wastes in Near-Surface Disposal Specified by 10 CFR 61

Isotope	Concentration Limit Defining Class A Waste, Ci/m^3	Concentration Limit Defining Class B Waste, Ci/m^3	Concentration Limit Defining Class C Waste, Ci/m^3
Kr-85	a	a	a
I-129	≤ 0.008	-	0.008-0.08
H-3	≤ 40	$> 40^b$	b
C-14	≤ 0.8	-	0.8-8

^aThe limits listed in [10 CFR 61] are not intended to apply to the millions of curies of Kr-85 that would require management from fuel reprocessing; the NRC intends, at some future time, to assess krypton-disposal technology and establish disposal requirements for this source of Kr-85.

^bNo maximum established; the permissible quantity is to be determined by radiation and heat generation.

The waste characteristics listed in 10 CFR 61.56, as minimum acceptable requirements, intended to facilitate handling and provide protection of health and safety to personnel at the disposal site, can be summarized as follows:

- (1) Waste cannot be packaged in cardboard or fiberboard boxes.

- (2) Packages containing liquids must also contain absorbent materials sufficient to absorb twice the volume of contained liquid.
- (3) Solid wastes containing liquid must contain as little free-standing and noncorrosive liquid as is reasonably achievable, but in no case must the liquid exceed 1% of the volume.
- (4) Wastes must not be capable of detonation, explosive decomposition, or explosive reactions in exposure either to normal temperatures and pressures or to water.
- (5) Waste must not be capable of generating quantities of toxic gases.
- (6) Pyrophoric materials must be packaged, treated, or prepared to render them nonflammable.
- (7) Wastes in gaseous form cannot exceed pressures of 1.5 atm at 20°C.
- (8) Wastes in gaseous form cannot exceed 100 Ci per container.
- (9) Wastes containing biological, pathogenic, or infectious materials must be treated to reduce the hazard as far as practicable.

Additional rules are given to provide stability of the disposal site by requiring stability of the waste forms:

- (1) Wastes must have structural stability provided either through stability of waste form or container.*
- (2) Where stability is provided by the container, liquids must not exceed 1% of the volume. Where stability is provided by the waste form, liquids must not exceed 0.5% of the volume.
- (3) Void space, within the waste and also between the waste and containers, must be minimized.

B. Greater Confinement Disposal (GCD)

1. Development of GCD

a. Need for a Greater Confinement Concept

The status of the development of the GCD concept was reviewed in recent conferences [LARGE-1981, -1982] on LLW disposal. The GCD concept is being explored for several reasons: (1) near-surface disposal may be inadequate for some types of LLW, (2) depth is likely to enhance the safety of storage, and (3) GCD may offer an economic and safe alternative to deep geological

* For Class B and Class C wastes, either the waste forms or the containers must maintain gross physical properties and identity for 300 y.

disposal for high specific activity wastes. Low-level wastes not considered suitable for SLB, even though they are non-TRU wastes, include those with specific activities that are too high. These wastes are thought to present significant risks if disposed of by SLB as a result of potential for exposure through erosion, transport by groundwater or vaporization, intrusions by plants and animals, and inadvertent human intrusion [HUNTER]. In the past, deep-geologic repositories have been viewed as the best destination for these types of wastes, but recently, it has been postulated that a more-economic, safe alternative is required.

In the waste classification system proposed by programs devoted to the development of near-surface disposal regulations, a class of waste that requires some sort of confinement greater than SLB was identified [MEZGA].* Of the three classes of wastes considered by NRC to be compatible with SLB, class A wastes are acceptable for routine disposal in conventional SLB facilities without additional confinement. Class A wastes have low specific activity and include transuranic wastes having alpha contamination less than 10 nCi/g. Class B waste requires confinement in a stable matrix or waste form. Class C waste, also called "intruder waste," requires the use of at least 5 m of cover, or the use of engineered barriers, or packaging in high-integrity containers. In addition, the NRC has a category of waste not generally acceptable for SLB at depths less than 15 to 20 m [MEZGA]. This category, while considered by many to encompass only a small volume of waste, has come to be called GCD wastes. The NRC classification and disposal scheme identifies two broad categories of disposal: (1) near-surface disposal including conventional SLB (depths up to 20 m) and greater depth disposal with at least 5 m of cover (depths of up to 15-20 m) and/or the use of engineered barriers to reduce intruder potential and (2) disposal at depths greater than 15 or 20 m. The DOE identifies burial at depths greater than conventional SLB (*i.e.*, 20 m) and the use of engineered barriers, containment, and solidification as options for achieving GCD [MEZGA]. Once the waste classification system has been completed, the DOE will be able to identify which technological options are the most appropriate for achieving GCD for specific wastes [MEZGA].

b. Definition of GCD

The DOE low-level waste management program has defined greater confinement as "the disposal of LLW in such a manner as to provide greater containment of radiation, reduced potential for migration/dispersion of radionuclides, and greater protection from inadvertent human and biological intrusions in order to protect the health and safety of the public [MEZGA]."

Greater confinement disposal is the emplacement of radioactive waste, with no expectation of retrieval, below grade level at sufficient depth that (1) surface effects (*i.e.*, floods, erosion, *etc.*) on waste containment are minimized and (2) the consequence of any penetration of the waste by plant roots, burrowing animals, or normal use of the land by humans is within acceptable limits [CARD].

One study derived a recommended optimum depth for GCD. This optimum depth was about 35 m and assumed an arid site with an aquifer depth of

*Presumably those that will not fit into any of the classes of 10 CFR 61.

170 m [HUNTER]. Humid sites or sites with more shallow aquifers may be expected to have an optimum burial depth approximately midway between the aquifer and the surface.

c. Comparison of the GCD and SLB Concepts

From an operational point of view, GCD is quite similar to SLB. The GCD concept involves burying the waste within unconsolidated material near the surface, but at greater depth than practiced in SLB. The advantages and disadvantages of GCD as an alternative to SLB or deep geologic disposal are summarized below [CARD].

Advantages:

- GCD positions the waste farther from plant roots and animals than does SLB, minimizing the possibility of human intrusion (by definition, GCD is designed to minimize the inadvertent reclaimer scenario^{*}).
- The increased depth may prevent, or reduce, infiltrating water from reaching the waste, thereby reducing the amount of waste that reaches the surface from upward migration.
- Stable temperatures at greater depths may be an important consideration where the formation of gases containing radionuclides is a concern.
- GCD provides greater protection against uncovering the waste through surface erosion.
- GCD, particularly at arid sites, may be a technically feasible and cost-effective alternative to deep geologic disposal for certain types of high specific activity wastes and wastes having long-lived nuclides, including transuranic wastes in very low concentrations.

Disadvantages:

- Increased costs are incurred for excavation and handling.
- The greater depth reduces the distance between the waste and the water table, increasing the potential for release to potable water supplies if water reaches the waste. This disadvantage, however, may not be significant, depending on the absorptive characteristics of the underlying strata, if the water table is sufficiently deep (>100 m).

* Presumably the radiation exposure of a person engaged in some future attempt to reclaim the site for purposes other than radioactive waste disposal.

Other waste disposal considerations for GCD, such as transporting, stacking, and health and safety requirements, are similar to SLB considerations.

d. DOE Program to Develop the GCD Concept

After a preliminary screening of several alternatives to SLB, the option selected for further study was burial at greater depths (GCD), with or without some combination of solidification, containment, or engineered barriers [MEZGA]. Solidification, containment, and engineered barriers were included as aids to confinement because in some geological environments depth alone may not provide greater confinement than SLB [MEZGA].

The purpose of the DOE GCD program is to explore whether GCD disposal of LLW provides greater containment, reduces the potential for migration/dispersion of radionuclides, and provides greater protection for inadvertent human and biological intrusions in comparison with SLB [HUNTER]. The main goals of development of the GCD concept are [HUNTER]: (1) the environmental impact of GCD should not exceed that of SLB and should conform to the guidelines of 10 CFR 61 and 10 CFR 20, (2) the increased cost (presumably over that of SLB) should be justified by a cost/benefit index, defined as the product of the facility cost and the maximum individual dose in rem (after release of institutional control) divided by the inventory in total Curies placed into storage. A recent estimate [HUNTER] of this index, for typical sites in the arid West, gave a value of 1.73 for SLB and 0.31 for GCD, a reduction by a factor of nearly six.

Four sets of factors are considered important in the performance of a GCD facility and are to be investigated in the GCD program: (1) the geologic medium, including structural, thermal, mechanical and hydrologic properties, the transport of contaminants, and homogeneity; (2) the facility design, including emplacement depth, waste volume, dedicated land surface, backfill operation, monitoring system, and the waste canister; (3) facility operations, including the sequence of operations, waste-emplacement methods, waste handling and transfer methods, site maintenance requirements, and decommissioning requirements; and (4) waste characteristics, including leachability, radionuclide mobility, radioactivity inventory, thermal characteristics, packaging, and handling [HUNTER].

e. Plans for Demonstrating the GCD Concept

Demonstration of one GCD concept at an arid site is being planned for the Nevada Test Site. The first objective of this demonstration is to evaluate the adequacy of geologic containment of waste at below near-surface depths. To accomplish this objective, three types of experiments have been defined: measurements of movement of moisture from the test hole, determination of the thermal response of the geologic medium to heat-generating waste, and observations of nuclide migration. The second objective is to define equipment and procedures required for GCD operations. The third objective is to establish costs. The GCD demonstration will consist of six subtasks: (1) systems analysis and evaluation, (2) design, (3) construction, (4) operation, (5) monitoring, and (6) decommissioning. This demonstration

was planned for FY 1982. The construction was planned for FY 1982-83, and operation is expected to be carried out during FY 1983-84 [MEZGA].

Demonstration of the intermediate-depth concept at a humid site is planned at Savannah River Laboratory [MEZGA], which plans to (1) operate a small-scale facility, (2) develop site-selection methods, (3) develop a reference design and operating procedures, and (4) monitor the behavior of the emplaced waste. Three initial designs have been suggested for an intermediate-depth facility at the SRL site: (1) a facility with a clay-lined bottom and a sloped clay cap--a layer of gravel would be positioned on the clay bottom from which any collected water would be periodically pumped out, requiring preparations for long-term maintenance; (2) a facility with a clay cap but no clay bottom and no provisions for long-term maintenance; and (3) a facility consisting of deeply placed upright sections of concrete culvert that encloses the wastes [HOOKER]. The SRL is looking ahead to the possibility of introducing intermediate-depth waste burial when the need for additional LLW burial facilities arises. This need may be felt in 7-11 y. Preparations for this possibility will include site-selection work, the development of containment criteria, radionuclide-migration studies for selected facility designs, and the estimation of cost estimates [HOOKER].

2. Criteria Proposed for GCD

It is expected that criteria for GCD will be developed during 1983 [LEVIN-1982]. At this time, the only available indications of possible criteria for GCD are suggestions given in a report on proposed criteria [CARD], which are summarized below. Only those criteria that seem likely to be of interest to organizations planning to send wastes to a GCD facility are of interest in the present study and are reported in some detail below. The criteria not discussed here include (1) radiation exposure protection to operating personnel, (2) radiation exposure protection for the general public, (3) documentation, (4) disposal-site waste handling, (5) inspection of waste received, (6) subsurface hydrology, (7) surface hydrology, (8) geology, (9) weather, (10) erosion, (11) tectonic activity, (12) archeological considerations, (13) recreational considerations, (14) natural resources, (15) flora and fauna impacts, (16) monitoring, (17) disposal depth, (18) buffer zone, (19) stratified disposal, (20) engineered barriers, (21) ownership, (22) demography, (23) land use, (24) resource commitment, (25) surface facilities, (26) personnel training, (27) occupational safety, (28) site security, (29) fire protection, (30) personnel and equipment decontamination, (31) equipment and facility maintenance, and (32) access roads.

The criteria and the associated suggested standards that are likely to have a bearing on the plans of the organization intending to send its wastes to a GCD facility are summarized as follows.

Waste Description Criterion: Waste accepted for disposal shall be identified as to its source, physical characteristics, and general chemical makeup.

Standard: The containers of all high-specific-activity or non-low-specific-activity* waste containing long-lived isotopes shall be marked with a durable label describing the contained waste, the date it was packaged, the generator of the waste, and any identification number related to accompanying paperwork which describes associated hazards, radionuclides, etc.

Waste Container Criterion: The waste container shall be adequate (1) to maintain its integrity during transportation, handling, and stacking at the disposal site, and (2) to permit decay of waste to levels that would not impose a public hazard if the container were breached at a reasonable period after disposal (i.e., 100 y). (The lifetime of the container may be less than 100 y if the integrity of the waste disposal cell is designed to be maintained for 100 y.)

Standard: A waste container shall be capable of supporting five times its own maximum loaded weight applied uniformly in the direction in which it will normally be stacked. For certain types of waste (e.g., tritium), the containers should be designed to be capable of a reasonable period of integrity (e.g., 100 y) to permit decay to nonhazardous levels by the end of the control period.

Volume Reduction Criterion: The volume of the waste material shall be minimized, consistent with contemporary engineering and economic factors.

Standard: Reducible (by volume) or combustible waste shall be reduced in volume to the extent practical through the use of incinerators or compaction equipment.

Waste Form Criterion: The physical state of the waste shall be solid.

Standard: Waste shall be in a dry solid form containing not more than 0.5 % by volume or 3.8 L (1 gal) of free neutralized liquid per container, whichever is less. Liquids shall be immobilized by absorption in a dry, freestanding, homogeneous, monolithic matrix which is not readily dispersible, friable, or soluble.

Standard: Liquids may be accepted for disposal provided (1) there is no other practical means to immobilize or solidify the liquid, (2) specific DOE permission is granted, and (3) reasonable assurance can be given that the liquid will be completely contained in the disposal facility throughout the time the liquid remains a radiologic hazard (e.g., reasonable assurance may be demonstrated by use of high-integrity containers).

Standard: The waste form shall not be liable to cause fires through friction, absorption of moisture, spontaneous chemical changes, or retained heat from manufacturing or processing, or when ignited, burn so vigorously and persistently as to create a hazard during handling, storage, and disposal operations.

* Not quantitatively defined.

Standard: The waste form shall not contain untreated oxidizing materials, including chlorates, permanganates, inorganic peroxides, nitrates, or any other material readily yielding oxygen in sufficient quantity to stimulate combustion.

Standard: Chemically reactive LLW for which there is no practicable way to reduce the reactivity may be, subject to specific DOE permission, accepted for GCD if isolated in boreholes. Reactive waste in boreholes may be considered isolated if the depth of burial and borehole interspacing is greater than ten meters or greater than the distance required to absorb adequately the shock of detonation of twice the material without affecting nearby holes. Such materials may include waste which

- Is normally unstable and easily undergoes rapid chemical change without detonating; reacts rapidly with water or forms potentially explosive mixtures with water; generates or decomposes to form sufficient explosive or toxic gases, vapors, or fumes to be a hazard; or is a cyanide- or sulfide-bearing waste which can generate toxic gases, vapors, or fumes when exposed to mild acidic or basic conditions.
- Is readily capable of detonation or of explosive decomposition or reaction at normal temperatures and pressures, or reacts explosively with water.
- Is an explosive, i.e., any chemical compound, mixture, or device which explodes with substantially instantaneous release of gas and heat.

Waste Concentration Criterion: The average concentration of radioactive nuclides in the LLW disposed in a GCD facility at a generic arid site shall be low enough that releases to the biosphere will be within acceptable limits.

Standard: The concentrations of the airborne nuclides shall not exceed the limiting-case area concentration limits shown in column 2 of Table IV-3. The area concentration limit represents the amount or inventory (Curies) of waste that can be disposed in a GCD facility per unit area (square meters) of site dedicated for disposal. The allowable concentration limits calculated for arid site conditions (expected case) are shown for the nuclides in column 3 of Table IV-3 for comparison. Where isotopic concentration limits are not listed, the concentrations of the isotopes are unrestricted, provided the radiation and thermal emissions of the waste package are within acceptable limits. Nuclide-containing wastes having a concentration below that shown in column 4 of Table IV-3 should be considered for disposal at SLB sites.

Packaging for Transport on Public Road or Railways Criterion:
The waste package, including any overpack container or shielding, shall be capable of maintaining its integrity through any credible transportation accident.*

* This criterion would probably require the type of packaging classed as Type B by DOT regulations in 49 CFR 171-179.

Table IV-3. Proposed Area Concentration Disposal Limits for GCD of Airborne Wastes [HUNTER]^{a,b}

Isotope	Concentration Limits, Ci/m ²		
	Humid Site ^c	Arid Site ^d	SLB ^e
H-3	940	6,500,000	40
C-14	2.3	3.3	0.8
I-129	0.0011	0.05	0.008
Kr-85 ^f	----	----	----

^aMinimum area disposal concentration limits for GCD represent the inventory of nuclides in the waste (Ci) that can be safely disposed per unit area (m²) at a mean depth of 35 m. The limits assumed that containers maintain their integrity for a period of 100 y and are based on pathway analysis developed in Appendix A of [CARD].

^bThe numbers in this table are currently being revised to take into account more realistic hydrogeological soils information for typical arid and humid sites. The area concentration limits presented may not be conservative. Tables of area concentration limits for a typical arid western site (viz. the Nevada Test Site) and a typical humid eastern site (viz. the Savannah River Plant) are expected to appear in the final draft of a report by Hunter, Documentation of the PATHWAYS Computer Model: An Addendum to NVO-234, a document currently being prepared by Ford, Bacon and Davis Utah for DOE Nevada Operations Office.

^cAssumes a vertical groundwater velocity of 5 m/y, dispersion of 100 m²/y, and aquifer depth of 170 m.

^dAssumes a vertical groundwater velocity of zero, dispersion of 0.1 m²/y, and aquifer depth of 170 m.

^eMinimum SLB concentration limits per unit site area were estimated from [10 CFR 61], assuming a waste thickness of 1 m.

^fNo suggestion given for Kr-85.

Standard: The waste container shall conform to the pertinent regulations of DOT, NRC, and any applicable laws of the state through which it may be transported.

Packaging for Off-Loading and Stacking Criterion: Waste containers shall be designed with adequate strength to maintain their integrity during normal off-loading and stacking operations.

Standard: All packaged LLW accepted for disposal shall at a minimum meet the water spray, free drop, compression, and penetration tests

of [49 CFR 173.398] for Type A packages without a substantial reduction of the effectiveness of the package, loss or dispersal of the contents, or an increase in the maximum radiation levels recorded or calculated at the external surface of the package.

C. Deep Geologic Disposal

1. Concepts of Deep-Geologic Disposal Technology

Several descriptions of deep-geologic disposal technology have been published over the past decade [ERDA-75-43B, DOE/EIS-0046F-5.3, STEARNS, KAISER, BECHTEL]; this technology has evolved to the design stage [DOE/NWTS-15]. Although several preliminary concepts for disposal of waste into deep geologic media have been proposed [BNWL, GHOVANLOU], the current national strategy is focused on placing waste in metal canisters into cavities created by mining and drilling [DOE/NWTS-15].

A brief characterization of deep geologic disposal, sufficient for the present purpose, can be made in terms of three main aspects: the facility, handling operations, and packaging. A generic repository would have the following main characteristics [CRANDALL]: (1) surface facilities capable of receiving and temporarily storing encapsulated nuclear waste, (2) shafts connecting the surface facilities with the underground storage horizon, (3) a storage horizon located underground in geologically stable strata, (4) a ventilation system providing fresh air underground and particulate filtration, (5) equipment to aid contact-handling* of TRU-contaminated LLW, and (6) specialized facilities and equipment to handle remotely HLW and ILW, including hot cells, casks, and shielded transporters.

Additional details of a repository created in a salt dome are suggested in [STEARNS]. Four types of waste are assumed to be accepted at such a facility: solidified HLW, solidified ILW, cladding hull waste (CHW), and TRU-contaminated LLW. These wastes are assumed to be delivered to the repository by truck or railcar. The TRU-LLW would be emplaced at a depth of 460 m (1500 ft), and the HLW, ILW, and CHW would be emplaced at a depth of 610 m (2000 ft). The HLW, ILW, and CHW are assumed to be delivered in steel canisters that would be placed in holes drilled in the host stratum. To facilitate retrieval, the holes could be fitted with carbon steel liners and concrete shielding plugs. The LLW incorporated into concrete is assumed to be received at the repository in the form of 55-gal drums that would be emplaced by using pallets and stacking operations.

The area occupied by a generic repository includes an 80-hectare, central, controlled area containing the surface facilities within a 240-hectare fenced area, with access roads and rail spurs. Workers at the surface facilities handle the incoming wastes and prepare them for transfer to the underground storage horizon. Surface operations include: (1) unloading of shipping casks and cargo carriers arriving by rail or truck, (2) transfer of waste packages to inspection stations, (3) decontamination of waste packages that are

* Manual handling permitted; does not require use of remote-handling apparatus.

contaminated upon arrival, (4) overpacking of contaminated waste packages as required, (5) transfer of waste packages to underground facilities, (6) radwaste treatment and handling of onsite-generated wastes, and (7) general repository support procedures.

Several buildings are necessary to carry out the surface operations. (1) A receiving building houses all facilities and equipment to receive, handle, decontaminate, and temporarily store canisters requiring remote handling; both normal and overpacked canisters are prepared for transfer to underground storage. A separate wing of this building is used to receive LLW, packed in 55-gal drums and handled by direct-contact methods. (2) An underground-operations building houses (a) the support operations for underground facility personnel, their equipment, and supplies, (b) a maintenance and repair area for servicing all noncontaminated underground equipment, and (c) a warehouse with storage space for equipment and supplies. (3) A compressor-and-chiller building houses the compressors and water chillers for the air conditioning and ventilation systems, potable water, instrument air, compressed air, chilled water, and steam condensate systems. (4) A waste shaft-ventilation building contains the equipment to supply the shaft with conditioned air and houses the intake and exhaust air fans, filter, air conditioning, and electrical equipment. (5) A man-and-materials building houses equipment supplying ventilation to the man-and-material shaft, including fans, filters, air conditioning, and electrical equipment. (6) A ventilation-supply-filter building contains the filters for the conditioned ventilation air flowing to the underground areas. (7) A development-exhaust-filter building contains the filters for the exhaust air from the underground areas during mine development. (8) A repository-exhaust-filter building contains the filters for the ventilation exhaust air from the underground repository operating areas. (9) A radwaste building houses facilities, control room, and monitoring equipment processing the liquid and solid radioactive waste generated on site. All waste generated on site is expected to be LLW. After treatment and encapsulation, the drums are conveyed to the LLW area of the receiving building in preparation for lowering to the underground repository area. (10) An administration building houses all activities necessary to the administration of the site. (11) Storage of the media excavated from the mine may require storage in a structure at the surface for protection from weather elements.

The generic repository has five shafts: (1) waste-handling shaft, (2) man-and-material handling shaft, (3) ventilation-supply shaft, (4) repository-operations ventilation exhaust shaft, and (5) the mine-development ventilation exhaust shaft. Each shaft extends some distance below the storage horizon and is lined with concrete from the surface to about 30 m into the storage horizon.

The layout of underground areas is based on the following design considerations. (1) All shafts are located in a central area. (2) The underground development area is 800 hectares. (3) The waste is always emplaced in the outermost available areas of the repository. (4) The repository is initially excavated to permit one year of operation before waste emplacement operations are started. (5) The salt pillar between the storage rooms is sized to provide an adequate heat sink. For the generic repository, these pillars

are 42 m wide to accommodate the maximum design heat load of 370 kW/hectare. The pillars also maintain the physical integrity of the storage rooms. (6) During operation of the facility, continuous mining is carried out to open new storage rooms while waste is emplaced in the existing rooms. (7) High heat-producing wastes (HLW and spent fuel) must be retrievable after the start of operations, for a period of time to be determined by criteria and regulation developments. (8) The ILW and LLW are emplaced without provision for retrievability.

During initial development, five main corridors accommodate mined salt conveyors, mining equipment, fork-lift trucks, personnel, and air supply and exhaust flow while mining operations are in progress. Support facilities for mining operations are located in the central area. This support area includes personnel accommodations, shops, tool room, storage areas, and maintenance areas. Excavated salt is hoisted to the surface for stockpiling. No backfilling is scheduled during the retrievability period.* Other concepts [ARBITAL], however, call for backfilling immediately following emplacement with an easily removed media, e.g., washed sand. The mine is laid out with the storage rooms connected at right angles to branch corridors and separated by salt pillars. These rooms may be entered from branch corridors at either end. Main corridors extend in two directions from the central core area and afford access to each branch corridor. The main and branch corridors are about 9 m wide and 4.5 m high, and each storage room is about 6 m wide, 4.5 to 7.5 m high, and 300 m long. The height of the room takes into account the movement of the ceiling and floor from salt creep thermally induced by HLW.

Several wells are drilled in each room for storing waste that must be handled remotely. The placement of the storage wells varies with the heat load or sizes of the waste-handling equipment, depending on the type of waste stored. After a retrievability period, the storage room is backfilled with salt from ongoing mining operations or from the surface salt stockpile.

Ventilation is separately provided to the waste storage area and to the continuing mining operation. The ventilation air flow is arranged so that air is always moving towards areas of potentially higher contamination. The air pressure through the mining area is greater than that in the storage area so that flow is always away from mining personnel and toward the waste storage areas.

The specifications for packaging that will be accepted for deep geological disposal have yet to be fixed in the development of criteria, regulations, and design. Preliminary packaging concepts, however, have been described, and preliminary criteria for acceptance of packages at a deep geological repository have been suggested [RHO-CD-568]. It has been assumed that spent fuel and HLW would arrive in specially designed canisters and that ILW and TRU-contaminated LLW would arrive in 55-gal drums. Proposed canister characteristics are summarized in Table IV-4. Proposed acceptance criteria are summarized by the data of Table IV-5.

* A time period to be set by disposal regulations, following emplacement of waste, during which the retrieval of wastes in containers is permitted.

Table IV-4. Proposed Waste Canister Characteristics [ARBITAL]

Characteristic	Commercial Waste					
	Spent Fuel		HLW		Defense High-Level Waste	
	EIS ^a	Kaiser ^b	EIS ^a	Stearns-Roger ^c	Hanford ^d	SRP ^e
Waste Form	1 PWR fuel assembly or 1 BWR fuel assembly	1 PWR fuel assembly or 2 BWR fuel assemblies	Glass or Calcine	Glass	Glass or Calcine	Glass or Calcine
Canister Shape	Cylinder	Cylinder	Cylinder	Cylinder	Cylinder	Cylinder
Canister Material	Stainless or Carbon Steel	Carbon Steel	Stainless or Carbon Steel	Stainless Steel	Carbon Steel	Carbon Steel
Canister Dimensions and Weight						
Outside Diameter, in.	14 (PWR) 10-3/4 (BWR)	14-1/2	12-3/4 (glass) 8-5/8 (calcine)	12-3/4	24	24
Length, ft	16 (PWR) 16 (BWR)	15-1/2	10 (glass) 10 (calcine)	10	12	10
Weight, lb	2380 (PWR) 1360 (BWR)	3500	1720 (glass) 720 (calcine)	~3500	~4800 (glass) ~3200 (calcine)	~5000 (glass) ~1500 (calcine)
Overpack Material	Stainless or Carbon Steel	Carbon Steel	Stainless or Carbon Steel	Carbon Steel	NS ^f	Stainless Steel
Overpack Dimensions and Weight						
Outside Diameter, in.	16 (PWR) 12-3/4 (BWR)	16	14 (glass) 10-3/4 (calcine)	14	NS	27
Length, ft	16-1/2 (PWR) 16-1/2 (BWR)	Variable	11 (glass) 11 (calcine)	10-1/3	NS	10-5/16
Weight (Canister + Overpack), lb	3120 (PWR) 1950 (BWR)	Variable	2150 (glass) 990 (calcine)	~4000	NS	~5500 (glass) ~2000 (calcine)
Canister Fill	Helium (5 psig)	Sand	Helium (5 psig)	Air	Air	Air
Canister Heat Generation Rate, kW/Canister	0.55 (PWR) 0.18 (BWR)	1.75 (PWR) ^g 1.12 (BWR)	2.8 (glass) 2.5 (calcine)	4.35	0.26	0.34 (glass) 0.21 (calcine)
Canister Radionuclide Content, Ci/Canister	1.9x10 ⁵ (PWR) 6.2x10 ⁴ (BWR)	3.9x10 ⁵ (PWR)	6.7x10 ⁵ (glass) 1.3x10 ⁵ (calcine)	8.0x10 ⁵	4.3x10 ³ (glass) 7.5x10 ³ (calcine)	5.9x10 ⁴ (glass) 2.7x10 ⁴ (calcine)

^a[Y/OWI/TM-44].^b[KAISER].^c[STEARNS].^d[ERDA 77-44].^e[ERDA 77-42].^fNS = not specified.^gThree-year-old spent fuel.

Table IV-5. Proposed Canister Acceptance Criteria^a

Criteria	Spent Fuel		HLW	
	PWR	BWR	Glass	Calcine
Physical Dimensions and Weight				
Canister OD, in.	14	16	11.5	8.5
Canister Length, ft	17.5	15.5	14.0	14.0
Overpack OD, in.	16	18	13.5	10.5
Overpack Length, ft	18.5	16.5	15.0	15.0
Gross Weight, lb	10,000	10,000	5,000	5,000
Thermal Power				
Canister Heat Generation Rate, kW/canister	1.0	1.0	3.1	2.8
Radiation Limit				
Canister Dose Rate, rem/h	3×10^4 at surface	3×10^4 at surface	1×10^5 at 1 ft	1×10^5 at 1 ft
Internal Pressure Limit				
Canister Operating Pressure, psia	100	100	80	80
Surface Contamination				
Removable Surface Contamination ^b	--	--	--	--
Materials of Construction	Low Carbon Steel	Low Carbon Steel	Stainless Steel (304L)	Stainless Steel (304L)

^a[ARBITAL], abstracted from [RHO-CD-568].^bSee [49 CFR 173.397].

The packaging concepts have varied in several conceptual studies done for the NWTs project through the Office of Nuclear Waste Isolation (ONWI). A study by the Bechtel National Corp. [CRANDALL] contains suggestions on construction of packaging to be received at a deep geological repository. In particular, this study assumed that a mixture of off-gases--krypton, carbon dioxide, and xenon--would be stored as compressed gas in cylinders at the repository. This off-gas storage, however, would seem not to conform to the proposals on materials acceptable for geologic disposal that are in 40 CFR 191. These packaging concepts are summarized in the following paragraphs and in Table IV-6.

The HLW is put into a canister fabricated from carbon steel pipes (32.4 cm OD by 3 m long, with a wall thickness of 0.95 cm). A carbon steel pipe (3.4 m long, 35.6 cm OD) encases the canister as the overpack. Each canister contains approximately 0.18 m³ of vitrified waste and generates 22 kW of heat one year after discharge from the reactor. At ten years after discharge from the reactor, the heat generation rate is about 2.6 kW/canister.

Hulls, end fittings, or failed equipment are contained in a canister fabricated from carbon steel pipe (30-cm dia, 1-cm wall thickness, 3-m long). An overpack capability is provided in case the canister is damaged before it is stored in the repository. The overpack is also fabricated from carbon steel pipe (56-cm dia, 1-cm wall thickness, 3.3-m long).

The end-fittings canisters are identical to the hulls canisters and hold the severed end fittings of either three PWR or six BWR assemblies. The overpack is a carbon steel container identical to the hulls canister overpack.

Failed equipment, which is disassembled and reduced in size at the facility, is also packaged in hulls canisters.

The TRU wastes are sealed in 55-gal carbon steel drums (DOT 17C or 17H). Depending on the corrosive character of the waste, some of the drums may require plastic liners or stainless steel construction with 83-gal carbon steel drum overpacks.

The compressed gas cylinders used for the containment of krypton, xenon, and carbon dioxide have a nominal volume of 50 L. They are made from 23-cm (9 in.) dia pipe and are 1.40 m long. The cylinders are modified by the installation of a special valve designed for high-temperature service. The radioactivity of the gas-filled cylinders is (considered to be the equivalent of) high level. Groups of six gas cylinders are encapsulated in a 56-cm (22-in.) OD carbon steel canister.

2. Criteria and Regulations for Deep Geologic Disposal

Two definitions are presented to aid the interpretation of criteria and regulations for deep geologic disposal. "High-Level Radioactive Waste" (HLW) means (1) irradiated reactor fuel, (2) liquid 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

Table IV-6. Conceptual Packaging Characteristics for Deep-Geologic Disposal [CRANDALL]

Content and Type of Package	Package OD by length, m	Gross Weight, kg	Decay Heat, kW	Overpack OD by length, m	Remarks
Vitrified HLW canister	0.30 × 3.05	780	2.60	0.35 × 3.30	80% fill
Hulls canister	0.51 × 3.05	1,905	0.46	0.56 × 3.30	
End fittings canister	0.51 × 3.05	590	0.10	0.56 × 3.30	
Kr-Xe-CO ₂ canister	0.56 × 3.05	-	-	-	6 gas cylinders per canister
ILW Canister					
TRU and non-TRU wastes	0.51 × 3.05	1,497	-	0.56 × 3.30	
Failed equipment	0.51 × 3.05	-	-	0.56 × 3.30	
LLW (TRU) drum	(55 gal)	381	-	(83 gal)	

facility for reprocessing irradiated reactor fuel, and (3) solids into which such liquid wastes have been converted. "Geologic Repository" means a system which is intended to be used for the disposal of radioactive wastes in excavated geologic formations. A geologic repository includes (1) the geologic repository operations area and (2) all surface and sub-surface areas where natural events or activities of man may change the extent to which radioactive wastes are effectively isolated from the biosphere.

Wastes addressed in [10 CFR 60] and [DOE 5820] are HLW, as defined above, and the disposal area addressed is a deep-geologic facility as described in Section IV.C.1. The major features of the proposed rule cover performance objectives, siting requirements, design and construction (of the repository), waste packaging, and performance confirmation.

The code 10 CFR 60 (for HLW) is not as specific as 10 CFR 61 (for LLW), possibly because further development of 10 CFR 60 by the NRC must await issuance by the EPA of final standards for disposal of HLW. The "engineered system" comprises two major barriers--the waste package and the underground facility--each with its own performance objectives. In addition, independent requirements are for containment, controlled release rate, and 1,000-y ground-water transit time.

The specified technical criteria which appear to be most relevant to our study are given in Table IV-7.

The waste-form and waste-package requirements, addressed in 10 CFR 60.135, must be weighed relative to the proposed waste forms for the airborne wastes under evaluation in this program. The effects of the site on the waste package, the waste package on the site, and the waste form on the waste package must also be evaluated so that the given items are not compromised. Some design considerations are: solubility, oxidation/reduction reactions, corrosion, hydriding, gas generation, thermal effects, mechanical strength, mechanical stress, radionuclide retardation, radiolysis, radiation damage, leaching, fire and explosion hazards, thermal loads, and synergistic interactions.

The waste-form requirements are: a solid, all particulates to be in a consolidated form, any combustibles converted to a noncombustible form, non-explosive, nonpyrophoric, not chemically reactive, and no free liquids in an amount to impair waste integrity.

The waste package design requirements are that the container maintains waste containment during transportation, emplacement and retrieval, is handleable, and has a unique identification. The final version of 10 CFR 60 was recently published.

The DOE has initiated a "National Waste Terminal Storage (NWTS) Program," which is aimed at development of geological disposal; criteria for such disposal are described in [DOE/NWTS-33(1)], which discusses program objectives, and performance criteria; in [DOE/NWTS-33(2)], which presents site performance criteria; in [DOE/NWTS-33(3)], which presents repository performance criteria; and in [DOE/NWTS-33(4)], which presents waste package performance criteria.

Table IV-7. Technical Criteria for Deep Geologic Disposal [10 CFR 60]

	Engineered System	Repository	Waste Package
Time period during which packaging must contain waste after emplacement, y	≥ 1000		≥ 1000
Time period during which waste is released after package breach, y	≥ 1000	≥ 1000	
Time required for waste to reach nearest groundwater, y		≥ 1000	
Retrievability period, ^a y	≤ 110		≤ 110
Minimum depth, m		300 m	
Release rate, ^a fraction per y	$\leq \frac{1}{100,000}$		

^a Does not apply to radionuclides whose contribution is less than 0.1% of the total Curie release.

Performance specifications have been proposed for the waste form [DOE/NWTS-19] and the waste package [DOE/NWTS-20]. These are to be followed by more-detailed specifications. Interim specification for the Defense High-Level Waste form for geologic disposal is proposed in [N/DFT-35]. A second set of documents is being generated covering wastes sent to the WIPP for disposal of TRU wastes [WIPP-DOE-114; WIPP-DOE-069 (Rev 1); DOE 5820 (draft)].

Information from some of the above criteria, requirements, and product specifications in response to proposed and existing regulatory requirements has been collated. This information is presented for the overall general disposal and site requirements in Table IV-8, for the waste form in Table IV-9, and for the waste package in Table IV-10. The bulk of the available information on contractor response to regulations is in the area of defense TRU waste, with little or no information for commercial TRU or HLW.

Based on review of the information in the tables, a summary of some of the required physical, chemical, thermal, and mechanical properties of the

Table IV-8. Criteria and Characteristics of Deep-Geologic Isolation

	[10 CFR 60]	(EPA) [40 CFR 191]	[WIPP-DOE-114]
Waste Type	HLW	Nuc. Fuel, HLW	Defense TRU
Site/Repository	Geologic	Any except direct to oceans	Geologic
Barriers	Multiple	Multiple	Overpack
System Type	Engineered	Engineered	
Waste Package			Type A [49 CFR 173]
Retrieval	110 y	Future, Yes	15 y
Containment	1000 y		
Design Life			15 y
Site			
Controlled Release	1000 y		
Thermal Spike Time	10,000 y		
Release Rate, Fraction/y			
<1000 y	none		
>1000 y	<1/100,000		
Ground Travel (Time or Distance)	≥1000 y	≥10 km	
Active Control (Institutional)		100 y	
Radioactivity to Biosphere			
Time		10,000 y	
Radionuclides Excluded		t _{1/2} < 20 y	
Dose (max.) ^a			
whole body		25 mrem/y	
thyroid		75 mrem/y	
other organs		25 mrem/y	
Interim/Long Term Storage		<10 y (minimize)	
Cumulative Release for			
10,000 y			
¹⁴ C		200 Ci/1000 MTHM	
¹²⁹ I		500 Ci/1000 MTHM	

^aThese limits apply only to management and storage.

Table IV-9. Waste Form Requirements for Deep-Geologic Disposal

Waste Form Requirements	[10 CFR 60] HLW	[10 CFR 60] TRU	[49 CFR 173 .389-.395]	[10 CFR 71]	[WIPP-DOE-114] Defense TRU	[N/DFT 35] Defense TRU	[EVANS]
Form	Solid	Solid		Special form		Solid glass	
Particulatea	Consolidated	Consolidated		>0.5 mm			
Combustibles	Reduced to noncombust.	Reduced to noncombust.				Limited	
Explosive	None	None			None	Limited	
Pyrophoric	None	None			Chemically stabilized	Limited	
Chemically Reactive	None	None				Transformed to Chem. Stable	
Free Liquids	None	None			None	Controlled-None	
Compressed Gases					None		
Toxic and Corrosive					None	Transformed to nontoxic	
pH					>4.0		
Fissile Isotopes					<200 g/55 gal drum	$K_{eff}+3\sigma \leq 0.95$	
Isotope Quantity						F.P., 10^5 Ci/L glass; T-A T-B Q actinide, 10^3 Ci/L	1:3 Ci 200 Ci >200 Ci Kr:3 Ci 200 Ci >200 Ci C:20 Ci 200 Ci >200 Ci
Special form (Spec 55)			300 Ci/pkg				
Low Specific Activity			10^{-6} Ci/g ^a				
Immobilization of Powders							
<10 μ m size					>1 wt %		
<200 μ m size					>15 wt %		
Radiolysis						Control to reduce leach rate	
Isotope Group; Transport							
III			^{129}I ; ^{85}Kr	^{129}I ; ^{85}Kr			
IV			^3H ; ^{14}C	^3H ; ^{14}C			
Oxidizers ^a						Limited	
Leach Rates							
Solubility				<0.005 wt %			
Surface Area for Dissolution							
Strength to Withstand Hydrostatic or Lithostatic Pressure						18 MPa	
Temperature							
Melting Point, Ignition in air				>1000°F			

^a Non-TRU where activity from TRU <1% of total activity.

Table IV-10. Waste Package Requirements for Deep-Geologic Disposal

Waste Package Requirements	[10 CFR 60] HLW	[10 CFR 60] TRU	[49 CFR 173 .389-.39 ^c]	[10 CFR 71]	[WIPP-DOE-114] Defense TRU	[N/DFT-35] Defense TRU
Sealed	Yes	Yes	Yes	Yes		
Structural Strength for Transportation, Em- placement, Retrieval	Yes	Yes	Yes (T) ^a	Yes (T) ^a	Yes	
Combustible					Non	
Type Package				Special Form	A	
Specification					[49 CFR 173.398 (b)]	
Design Life					15 y	
Structural Strength for Gas Containment					vented at 7 psig	
Corrosion by Waste Form					Minimal	
Weight					<25,000 lb	
Size				>0.5 mm		
Maximum					12 x 8 x 8.5 ft	
Minimum					30 gal drum	
Gas Generation					<10 mol/m ³ y	
Sludges					internal corrosion protection	
Thermal Power					consider if >3.5 W/m ³	6.74 x 10 ⁻⁴ W/cc
Surface					200 mrem/h	
Dose Rate (max)					α 0.5 pCi/cm ²	
Contamination			α 10 ⁻⁴ μCi/cm ² β 10 ⁻³ μCi/cm ²		β 4.5 pCi/cm ²	10 ⁻⁴ μCi/cm ²
Temperature						
Regular Shipment			<122°F			
Exclusive Use Shipment			<180°F			
Normal Form Type "B"			<250°F			
Decay Heat			10 W			
Special Form Type "B"			2 W			
Centerline (max)						350°C
Melting Point, Sublimes or Ignition in Air				>1475°F		
Shielding			Yes			
Breach (ΔP = 1 atm)						10 ⁻⁵ atm cc/s
Quantity per Pkg.						
Type B				<200 Ci		
Large Quantity				>200 Ci		
Solubility				<0.005 wt %		

^a(T) Transportation only.

site, waste form, and waste package can be generated (Table IV-11). Our sole interest here is in the properties that might be applicable to the waste forms and containers associated with the airborne wastes under consideration.

D. Ocean Disposal

1. Practices in Ocean Disposal of LLW

a. Present Status

Opinions on the propriety of using the ocean for nuclear waste disposal have been set forth by private environmental groups. The present study, however, is limited to a consideration of the expected performance of the disposal technologies and comparison of this performance to Federal regulations and criteria; therefore, this section of the report does not treat nonregulatory discussions and opinions.

Much of the information presented in this section has been obtained from the recent review by Holcomb on the history and practice of ocean disposal of LLW [HOLCOMB]. Ocean disposal of high-level wastes is prohibited at present. The ocean disposal of low-level wastes, although not presently practiced by the U.S., was not legally prohibited until the U.S. House of Representatives recently passed legislation establishing a two-year moratorium on EPA approval of applications for ocean disposal of LLW [NWN]. Amendments to the Marine Protection, Research, and Sanctuaries Act would allow EPA to consider permit applications following expiration of the moratorium, dependent on an impact assessment to be filed by the applicant. The assessment will be required to include a list of radioactive materials, number of Curies, exposure levels, in rems, at the exterior and interior of the container, and an analysis of environmental impact. Ocean dumping of LLW will continue to be attractive to countries whose geography and geology make shallow-land burial difficult. Several aspects of ocean dumping of LLW that should be considered in the question of applicability to disposal of airborne wastes are presented here. Probably the two most important considerations of ocean disposal, for the analysis to be done here, are regulations and acceptable packaging.

b. History of the Practice of Ocean Dumping of LLW

Several countries, including the U.S., practiced ocean dumping of radioactive materials during the years 1946-1972 without any international control. Beginning in the summer of 1967, several countries dumped LLW in the Northeast Atlantic under the supervision of the NEA. From 1967 to 1980, about 700,000 Ci of radioactivity, contained in materials weighing about 70,000 tons, were dumped in this area. Between 1946 and 1970, the U.S. dumped LLW at 50 locations in the Atlantic, the Pacific, and the Gulf of Mexico. These actions involved a total of 86,000 containers and a total of 94,500 Ci. These U.S. dumping activities were carried out under AEC authority by AEC contractors.

Since ocean dumping of low-level wastes was practiced for several years, it is considered that the technology has been amply demonstrated.

Table IV-11. Summary of Information That May Be Required on Wastes Destined for Geologic Disposal^a

Item	Waste Form	Container
Physical Properties		
Dimensions		X
Geometry		X
Weight		X
Density	X	
Structural Support		X
Particle Size Distribution	X	
Amount of Particles	X	
Impact Resistance	X	X
Surface Area for Leaching	X	
Chemical Properties		
Leach Rate	X	
Volatility	X	
Flammability	X	X
Flash Point	X	
Isotope Content	X	
Chemical Composition	X	X
Chemical Forms	X	
Phases	X	
Radiolysis	X	
Activity Level	X	

^aPooled from documents on criteria and proposed regulations listed in Tables IV-8 through IV-10.

Table IV-11 (Cont'd)

Item	Waste Form	Container
Mechanical Properties		
Tensile Strength	X	X
Compressive Strength	X	X
Shear Stress	X	X
Modulus of Elasticity	X	X
Shear Modulus	X	X
Bulk Modulus	X	X
Elongation	X	X
Charpy Impact	X	X
Thermal Properties		
Thermal Conductivity	X	X
Specific Heat	X	X
Coefficient of Thermal Expansion	X	X
Melting Point	X	X
Temperature Limits	X	X

2. Criteria and Regulations for Ocean Disposal of LLW

a. Philosophy

According to Holcomb two philosophies characterize the use of ocean dumping: dilution/dispersal and containment [HOLCOMB]. The current U.S. position favors containment. Revisions in 1979 on the policy of the NEA of the OECD recommend minimizing release to the sea.

The tone of IAEA recommendations is that, although it has produced, specifically for ocean dumping, definitions of waste classes and recommendations for their handling, this effort should not be construed as encouragement for ocean dumping. Current general expectations are that ocean dumping would be used only if it is shown to be the best alternative to other disposal possibilities.

b. History of Rules on Ocean Dumping of LLW

The regulations and laws related to LLW ocean dumping differ from those for all other modes of disposal in that multilateral, international agreements rather than U.S. laws only must be considered. The EPA has the authority to control the use of this method of disposal by U.S. organizations and is charged with the responsibility of ensuring compliance of U.S. activities with international agreements.

In the 1958 Conference on the Law of the Sea, the IAEA initiated moves to control dumping. During the 1960s, the NEA developed operational controls for the safe dumping of LLW. The London Convention on the Prevention of Marine Pollution by Dumping of Wastes and Other Matter, commonly referred to as the London Ocean Dumping Convention (LODC), was assembled in 1972. It produced a treaty, accepted by 43 countries including the U.S., that became effective in 1975.

The LODC gave the IAEA responsibility for defining high-level waste, i.e., waste not acceptable for dumping, and for making recommendations on disposal of other waste types. In 1973-1975, the IAEA issued initial recommendations but further work was required. The IAEA recommendations were based on estimated dose rates derived from information on seafood consumption, oceanography, and marine biology. In 1975-1977, the IAEA revised these recommendations, and the revisions have been effective since 1978.

c. Content of Rules on Ocean Dumping of LLW

Under the LODC, each member nation is required to establish national control over materials leaving its shores for dumping. The LODC rules include operational controls that address conditioning and packaging of waste, choice of a dumping site, and container design. Not only a number of prohibited substances (Annex I) but also a list of substances requiring special permits (Annex II) are listed by the LODC treaty.

Although the main interest in this method has been for disposal of LLW, international guidelines also allow the dumping, in limited concentrations, of alpha-radioactive wastes and also wastes that might normally be considered medium or intermediate-level radioactive materials.

In 1978, the IAEA produced a generic assessment of radiological consequences with the object of defining HLW unsuitable for ocean dumping. The basis for these definitions was the long- and short-term transport of radionuclides in the ocean, considering physical transport processes and possible pathways to man. The activity limits were based on dose limits established by the International Commission on Radiological Protection. The result is the definition of three classes of wastes and specific activity limits for each above which any waste would be considered unsuitable for dumping. The limits for specific activities of waste for ocean dumping are: for alpha emitters, 1 Ci/MT; for beta-gamma emitters with $t_{1/2} > 0.5$ y (except tritium), 10^2 Ci/MT; and for tritium and beta-gamma emitters with $t_{1/2} < 0.5$ y, 10^6 Ci/MT.

d. Administration of Rules

The LODC is administered by the Intergovernmental Maritime Consultative Organization (IMCO). Although the LODC authorized individual countries to supervise dumping operations, most OECD member countries favored continuing NEA surveillance. Therefore, NEA agreed to continue a supervisory role and issued a statement of consultation and surveillance that was adopted in 1977 by the OECD Council. The OECD role is consistent with the objectives and principles of the LODC and the IAEA. The important aspects are:

- (1) establish and update standards, guidelines, and recommendations,
- (2) provide consultation,
- (3) provide international surveillance,
- (4) report to the OECD,
- (5) assess site suitability,
- (6) monitor previous dumping sites, and
- (7) provide notification of proposed disposal actions.

The U.S. is committed to application of the LODC to protect the marine environment and, as a participant in NEA-OECD agreements, to cooperate in surveillance of ocean disposal. The U.S. commitments to the LODC are enforced by Public Law 92-532, the Marine Protection Research and Sanctuaries Act of 1972. This law also gives EPA the authority to regulate ocean dumping of LLW by permit. With this authority, the EPA issued initial regulations and criteria in 1973. These rules stated the philosophy that U.S. objectives would be remote containment, not dilution/dispersal. Final regulations were issued in 1977, reiterating the philosophy that properties of packaging should permit decay to innocuous levels within the expected lifetime of the container or matrix. Since assuming this authority, the EPA has issued no permits for ocean dumping of radioactive materials.

e. Position of the U.S. on Ocean Dumping of LLW

Ocean dumping by the U.S. was discontinued for two main reasons. (1) The AEC and the nuclear industry decided to use shallow land burial; among other reasons, it was less costly [LEVIN-1982]. (2) According to [HOLCOMB], the Council on Environmental Quality issued a report to the President recommending the prohibition of ocean disposal for HLW and any other radioactive material unless no other alternative offered less risk to man and the environment.

After accepting the authority to regulate ocean dumping, the EPA began a program to determine the status of ocean sites in which dumping had been done. Between 1974 and 1978, the EPA conducted surveys on the condition of waste containers in both Atlantic and Pacific disposal sites. The EPA surveys of the Pacific sites in 1974, 1975, and 1976 were done from samples of

sediment, water, biological materials, and photos. The EPA also recovered a drum for corrosion observations. Some drums were observed to have imploded and low concentrations of radioactivity were found near some containers. It was concluded, however, that no risk to man or marine environment had been incurred.

Additional EPA surveys off the coasts of Maryland and Delaware found Cs-137 in sediment near drums, but also concluded that no risks to man or environment were involved. Some containers, retrieved from ocean disposal sites, were examined by BNL for corrosion, leaching, and degradation. Although the containers were corroded to varying degrees, the concrete matrix inside them had not degraded appreciably in 15 years of seabed residence [HOLCOMB]. Regardless of these favorable observations, the U.S. is presently neither encouraging nor discouraging ocean dumping, except for the present moratorium; if ocean dumping were allowed, its use would be strictly regulated

f. Packaging for Ocean Disposal of LLW

Since criteria for ocean disposal of LLW emphasize containment, packaging must have certain general attributes. The proper choice of corrosion-resistant materials and coatings for containers is expected to delay release to the ocean, permitting additional radioactive decay before exposure of waste or matrix to the ocean. Several factors could retard the release of packaged wastes to the ocean: radioactive decay; packaging integrity lasting several years; designs that include multiple barriers of bitumen, metal, plastic or concrete; slow leaching; and sorption by bottom sediments.

Packaging has commonly consisted of 55-gal drums or cylindrical vessels cast of concrete. Within these containers, the wastes are commonly incorporated into a matrix of concrete or bitumen. In some cases, pressure-equalization devices have been incorporated into the design. The low-level wastes have included a wide range of physical and chemical forms that can be incorporated into matrices. Although many matrix materials have been used to incorporate LLW, e.g., plaster of Paris, urea formaldehyde, bitumen, and organic polymers, cement is the matrix that has been used most in ocean disposal.

Guidelines for ocean disposal of packages of radioactive waste were prepared by NEA in 1974 [HOLCOMB]. The NEA convened another group in 1978 to review and revise these 1974 guidelines, setting forth general requirements for packaging design, radiological safety considerations, and instructions for packaging manufacture and assembly.

In 1979 the IAEA convened a group to assess requirements and practices of packaging radwaste for ocean disposal. The resulting report recommended IAEA guidance on licensing, operations, and administration [HOLCOMB]. Recommendations on packaging by IAEA are compatible with those of NEA guidelines on that subject. The major points are:

- (1) Waste should be cast in or absorbed in a matrix.
- (2) Liquids must be carried on an absorbent material in a primary container, which must be enclosed in a secondary container. The space between the containers must also be filled with absorbent.
- (3) Packages must provide containment during handling, transport, and descent to the bottom.
- (4) Specific gravity of packages must be >1.2 to promote descent to the bottom and resistance to movement in currents.
- (5) Package design must ensure that the inner container remains on the sea floor.
- (6) The container must remain intact under sea pressure or be equipped with a pressure-equalization device.
- (7) Buoyant materials must be excluded or be treated to prevent return to the surface or be innocuous and non-interfering in legitimate uses of the sea if they do return to the surface.

3. Concepts for Ocean Disposal of HLW

Disposal of HLW below the seabed has been identified, although presently prohibited, as an alternative to geologic repositories [ONWI-65]. Final EPA standards and NRC operating criteria are not yet available for sub-seabed disposal [SAND80-2384]; therefore, it was assumed by the Subseabed Disposal Program (SDP) that standards for subseabed would be the same as those for HLW in mined (geologic) repositories. The EPA is "... in the process of reviewing existing rules governing the ocean disposal of hazardous materials including radioactive waste. These were put into effect in 1977, as required under the Marine Protection and Sanctuaries Act passed in 1972" [RADEX]. Present policy appears to be that "disposal of high-level waste, defined in the 1977 regulations is absolutely prohibited" [RADEX].

The concept of emplacing I-129 wastes at a depth of about 30 m into ocean sediments has been under evaluation at Sandia [KLETT-1982]. This concept is not explored here because the regulations against which this practice could be compared have not been formulated.

E. Packaging

1. Current Federal Requirements on Packaging for Transportation of Radioactive Materials

a. Responsibilities of Federal Agencies

The responsibilities of Federal agencies in regulation of packaging of radioactive materials for transportation are summarized in Table IV-12. Since it is unlikely that airborne wastes from the nuclear fuel cycle

Table IV-12. Agency Responsibilities in Packaging of Radioactive Materials for Transportation

Agency	Area of Responsibility	Regulations
DOT	All radioactive materials in Interstate Commerce	Parts of 49 CFR 171-179
NRC	Radioactive materials of the nuclear fuel cycle	10 CFR 71
FAA	Transportation of radioactive materials by air	14 CFR 103
USCG	Transportation of radioactive materials by water	46 CFR 146, 149
USPO	Transportation of limited quantities of radioactive materials by mail	39 CFR 123-125
DOE	Transportation of radioactive materials by Headquarters divisions, offices, field organizations, and contractors	DOE 5480.1, Chap III

will be shipped by air or water, or through the mail, the regulations of the FAA, USCG, and USPO will be ignored here. Therefore, this discussion will deal with the regulations of DOT and NRC only. Since transportation is likely to be by exclusive-use shipment, regulations applying to common carriers are ignored.

The latest revision of DOT rules for transportation of radioactive wastes was issued recently [FEDREG 10218]. Revision of NRC rules on the same topic is expected to be issued also. The consistency of NRC and DOT regulations has been expressed in memoranda of understanding. The DOT regulations likely to have the most significance for waste management planning [49 CFR 173.389-173.398] are reviewed below.

The following discussion is a summary of the elements of DOT regulations that would be likely to apply to considerations of shipping the airborne wastes. The intent is to illustrate how the information in 49 CFR would be used to select packaging for these nuclides that would permit their transportation in compliance with the regulations. Although the regulations contain directions on a number of procedures to be followed, such as labeling of packages, placarding of vehicles, routing of shipments, controlling of surface contamination, and controlling of external radiation levels, probably the most important consideration for the present task is use of the regulations to extract an indication of what packaging would be permitted for the airborne wastes.

b. Determination of Acceptable Packaging

Several classification schemes are to be found in DOT regulations. The characteristics of the radioactive material to be shipped place it in a category or categories in these schemes. The categories, in turn, determine what packaging would be acceptable for the given material.

This indication of permissible packaging is probably one of the most important roles that transport regulations play in decision making for handling any radioactive waste.

Perhaps, the best place to begin in determining acceptable packaging for H-3, I-129, C-14, and Kr-85 is the question of whether the material will qualify as special form.

Special form material, because of the properties of either the waste form or its container, is essentially nondispersible in water, in a fire, and under severe impact conditions. The complete definition is found in 49 CFR 173.403. A practical significance of the special form definitions is that the rules permit a larger quantity of radioactive material in special form to be shipped in some packages because of the greatly reduced dispersibility. It is unlikely, however, that any of the airborne waste forms will qualify as special form material. Furthermore, it is unlikely that, for economy, airborne waste forms will be carried in containers that would qualify as special form. Therefore, it is assumed that the airborne waste forms would be classified as normal form according to DOT regulations. Accordingly, no further discussion on special form material is included here, and in the compliance analysis concerning transportation, properties of airborne waste forms and their packagings will be considered in the light of rules for normal form materials only.

A second point in choice of packaging is the A_2 value assigned to the nuclide by regulations. Each of the airborne nuclides is assigned a value of A_1 and A_2 , which play a role in determining acceptable packaging. (The assigned values of A_1 and A_2 represent the maximum number of Curies permitted in Type A packages in special form and normal form, respectively.) For the present task, only the A_2 value, relevant to materials in normal form, is considered. The A_2 values for the airborne nuclides are listed in Table IV-13.

A third point to be considered in the choice of packaging is whether the material to be shipped is exempt from specification packaging and labeling rules because of its small quantity or concentration. Two categories may qualify for exemption: limited quantity and low specific activity. The limited-quantity category is 10^{-3} A_2 per package for gases other than tritium and solids. It is unlikely that any of the airborne waste forms would be economically managed in such small quantities. The low-specific-activity category is a concentration of 3×10^{-4} Ci/g for the airborne nuclides, because their A_2 values are >1 Ci. Low-specific-activity material can be transported in DOT specification 7A, Type A packaging, or in any other strong, tight packaging that meets several other requirements listed in 49 CFR 173.425.

Table IV-13. Assignment of A_2 Values to the Airborne Nuclides by DOT Regulations [49 CFR 173.435]

Nuclide	A_2 , Ci
H-3(uncompressed)	1000
H-3(compressed)	1000
H-3(activated luminous paint)	1000
H-3(adsorbed on solid carrier)	1000
H-3(tritiated water)	1000
H-3(other forms)	20
I-129	2
C-14	60
Kr-85(uncompressed)	1000
Kr-85(compressed)	5

If the material to be transported is not exempt from specification packaging, then the next point to be considered is whether the amount of nuclide per package is less than or greater than A_2 . If the quantity is not greater than A_2 , Type A packaging can be used, or Type B packaging may be used if desired. If the quantity is greater than A_2 , then Type B packaging must be used.

The regulations divide Type B packaging into two sub-categories: B(U), which requires unilateral approval in international shipments, and B(M), which requires multilateral approval in international shipments. Since the present task considers only shipments of airborne wastes within the U.S., all Type B packagings are applicable.

In the choice of packagings, four general options seem to be available: (1) use of DOT specification packaging, (2) use of an NRC-licensed packaging that is already licensed specifically for the waste form of interest, (3) adaptation of an NRC-licensed packaging that has been licensed for some other cargo, and (4) design of a new packaging.

Table IV-14 lists the packagings that are permitted for carrying the radioactive material at $<A_2$ and $>A_2$. Six specification packagings are listed in Table IV-14: 7A, 6M, 20WC, 2R, 6L, and 6J. A brief description of these packagings is given in Table IV-15. Specification 20WC packaging is pictured in Fig. IV-1; Spec. 6M packaging, including the Spec. 2R container, is pictured in Fig. IV-2; Spec. 6J is illustrated in Fig. IV-3; and Spec. 6L is illustrated in Fig. IV-4.

Table IV-14. DOT-Authorized Packaging and Corresponding Quantities^a

Authorized packaging ^b for shipments of radioactive materials in normal form inside U.S.	Allowed Quantities of Radioactivity per Package for Normal-Form Materials	
	$\leq A_2$ Ci	$> A_2$ Ci
DOT Spec. 7A Type A general packaging (including Spec. 6L, Spec. 6J)	X	
Any foreign-made packaging that bears the marking "Type A" and complies with the regulations of the country of origin applicable to Type A packages	X	
Any Type B(U) or B(M) packaging that meets the applicable requirements in the regulations of the NRC [10 CFR 71] and that has been approved by that commission	X	X
DOT Spec. 6M metal packaging, only for solid or gaseous materials that do not undergo pressure-generating decomposition at temperatures up to 121°C and do not generate more than 10 W of decay heat	X	X
DOT Spec. 20WC wooden protective jacket, when used with a single, snug-fitting inner DOT Spec. 2R	X	X

^aDerived from DOT rules revised March 10, 1983 [FEDREG 10218].

^bAll packagings employing Spec. 55 containers are omitted here. Since only existing Spec. 55 containers can be used (the construction of new ones was prohibited after December 31, 1975), it is unlikely that they would be used in shipments of any airborne waste forms from fuel reprocessing operations.

Table IV-15. Description of Specification Packaging

Specification	49 CFR Part	Description Summary
20WC	178.194	A family of overpacks, 20WC-1 through 20WC-6, with permissible carrying capacity ranging from 500 to 6000 lb, depending on construction. General configuration is a cylindrical shell constructed by stacking rings of plywood or hardwood glued together and reinforced with steel rods, threaded at the ends and secured with washers and nuts. Some of the members of this family must be encased in steel liners. (See Fig. IV-1.)
2R	178.34	Inner container of metal; cylindrical configuration closed by screw-cap, welding, brazing, or flanged cover. Maximum I.D. of 30 cm, maximum length of 183 cm. (Examples of use are in 6M and 6L packaging; see Figs. IV-2 and -4.)
6M	178.104	Steel drum outer of 10-110 gal capacity. Inner container of no less than 1.24 L capacity. Inner container conforms to Spec. 2R with maximum I.D. of 13.3 cm and is fixed within the drum by solid centering media. Permissible gross weight, varying with volume, up to 640 lb for 110-gal drum. (See Fig. IV-2).
7A	178.350	A general specification that requires conformance to (1) the general requirements that all packagings must meet as described in 49 CFR 173.24, (2) the standards for Type A packaging in 49 CFR 173.389(j) and 173.398(b), and (3) package markings in accord with 49 CFR 178.350-3.
6J	178.100	Drum with rolling hoops of low-carbon steel with maximum capacity of 55 gal and maximum gross weight of 880 lb (See Fig. IV-3.)
6L	178.103	Outer drum of low carbon steel, with hoops rolled or swaged, with capacity of 55 to 110 gal and inner vessel of not more than 17.74 L capacity. Maximum weight limits are 350 lb for 55 gal size and 480 lb for 110 gal size. (See Fig. IV-4.)

Fig. IV-1. DOT Specification 20WC Packaging [49 CFR 178.194-7].

Fig. IV-2. DOT Specification 6M Packaging Including Spec 2R Inner Container [EDLING].

Fig. IV-3.

DOT Specification 6J
Packaging, Configura-
tion A [EDLING].

Fig. IV-4.

DOT Specification 6L
Packaging Including
Spec 2R Inner Container
[EDLING].

c. Use of Nonspecification Packaging

Acceptable packagings can be chosen from DOT specifications, but there are only a few from which to choose. One of the main questions is whether their volumes are practicable. Another question is whether they would be acceptable as terminal storage containers; although they are accepted by DOT for transportation, their acceptance by disposal-facility criteria must be determined. If DOT specification packaging is impracticable in the case of an airborne waste, the regulations can be used to indicate what tests would be required of other packaging that might be designed especially for this waste. The performance of nonspecification packaging in these tests can be evaluated either by engineering calculations or by physical tests on a prototype.

Table IV-14 includes, in addition to several specification packagings, the use of packaging that is approved by an agency or that meets regulatory requirements. Nonspecification packagings would have to be shown capable of meeting tests in licensing proceedings. Although the use of specification packaging would avoid the time and cost of design, testing, and licensing procedures, there are two important reasons why it might not be adequate for transportation of airborne wastes in the nuclear fuel cycle. First, the volume limits of specification packaging might require an unreasonably large number of packages. Second, the characteristics of DOT-specification packaging might not be compatible with acceptance criteria for terminal storage. In final emplacement, it would be convenient to use the container that the radioactive material was shipped in. Thus, it may be necessary to design a container that would conform to shipping regulations, provide a reasonably large capacity, and conform to acceptance criteria for final emplacement.

If a proposed design were to be used to ship airborne wastes in Type A packaging, it would have to be shown capable of meeting the general design requirements of 49 CFR 173.411 and the additional design requirements for Type A packaging that are summarized in 49 CFR 173.412. Furthermore, a proposed Type A packaging would have to be shown capable of withstanding the tests that are representative of normal conditions of transportation described in 49 CFR 173.465 (Table IV-16). On the other hand, a proposed Type B packaging would be required to meet the general design requirements of 49 CFR 173.411 and the Type B design requirements of 10 CFR 71. Furthermore, it must be demonstrated to be capable of withstanding the tests representative of normal conditions of transport described in 49 CFR 173.465 (Table IV-16) and also the tests representative of accident conditions described in 10 CFR 71 (Table IV-17).

A number of packaging designs, other than DOT specification packagings, are catalogued [WASH-1279, NUREG-0383]. These have been licensed by AEC, ERDA, or NRC. Some of these packagings may be adaptable for transportation of airborne wastes, but, since they are licensed for a specific cargo, their use for airborne wastes would require amendment. Thus, if packagings other than those specified in DOT rules are to be used, the adaptation of catalogued packaging could save the cost and effort of design and testing, though relicensing is unavoidable.

Table IV-16. Tests for Proposed Packagings: Normal Conditions of Transportation^a

• Water Spray	Must precede each test or test sequence; simulates exposure to rainfall of 5 cm/h for 1 h. Must allow maximum soak-in before applying succeeding test.
• Free drop	Is made onto target to cause maximum damage, distance and procedure specified according to design and weight of package. ^b
• Compression	Is applied for 24 h, at five times weight of package or 1300 kg/m ² times vertically projected area of package, whichever is greater; applied uniformly to two opposite sides, one of which is the base of the package.
• Penetration	Drop made through 1 m distance of 6 kg bar with 3.2-cm dia and hemispherical end onto package. ^c

^aFrom [49 CFR 173.465].

^bDistance for free drop test set at 9 m for Type A packagings designed to contain liquids and gases [49 CFR 173.466].

^cDistance for penetration test set at 1.7 m for Type A packagings designed to contain liquids and gases [49 CFR 173.466].

Table IV-17. Tests for Proposed Packagings: Accident Conditions^a

• Free drop	- A drop through a distance of 9.1 m onto an unyielding surface striking the surface in the position which would produce maximum damage.
• Puncture	- A free drop through a distance of 1 m onto a cylindrical steel bar (15.2 cm dia) with the edge rounded to a radius of 6.4 mm. The length of the bar is selected for maximum damage (minimum length is 20.3 cm). The bar is mounted on an unyielding horizontal surface and the package is dropped in a position expected to produce maximum damage.
• Thermal	- Exposure to the equivalent of an 800°C fire for 30 min with no artificial cooling for three hours after the exposure.
• Hydro	- Water immersion (fissile material packages only) at a depth of 91 cm for eight hours.

^aSubject to change in the revision of [10 CFR 71]. Tests applied sequentially to determine cumulative effect on the package.

A survey of available systems and equipment for transportation of radioactive materials was published in 1976 [JEFFERSON-1976]. Also, detailed descriptions of certified packaging have been issued periodically [WASH-1279, NUREG-0383, EDLING]. Recently, information on appropriate packaging for transportation of radioactive materials has been placed into computer storage at the SANDIA Transportation Technology Center (TTC). This information bank, the TTC Database of Certified Packagings for Radioactive Materials (RAMPAC Database), will output a description of appropriate transportation equipment in response to an input description of the waste [EMERSON].

It has been suggested [JEFFERSON-1982] that the RAMPAC Database might be searched to obtain descriptions of transportation equipment that could be applied to airborne waste forms. Plans for the analysis forming the second part of this program included assembly, for each airborne isotope, of a reference description of the most probable waste form and its packaging. Therefore, the RAMPAC Database will be used wherever possible to assemble these descriptions of a complete waste management system for each airborne isotope.

d. Requirements of the Licensing Process

Shipments of airborne wastes would require NRC licensing not only if nonspecification packaging were employed as discussed above, but also if shipments of Type B quantities or large quantities of radioactive materials even at low specific activity were involved. The general responsibilities of the shipper in the licensing process are to show that the packaging and vehicles involved conform to the Code of Federal Regulations, to describe the procedure that would be used in case of accident, to describe controls and precautions that will be used in handling packages, and to describe inspection and accountability procedures.

e. Limitation of External Radiation in Shipments

In addition to the concerns for containment, expressed in the specification of packaging construction discussed above, dose rates to the public and to shipping personnel are to be controlled through a transport index and also through a list of specifications on dose limits.

The transport index is a number placed on a package of radioactive material to designate the degree of control to be exercised by the carrier during transportation. For packages that do not contain fissile material, the transport index is the highest radiation dose rate in mrem/h at 1 m from any accessible external surface of the package.

Radiation dose limits for exclusive-use shipments are summarized in Table IV-18.

2. Packaging for Deep Geologic Disposal

The waste package for disposal of long-lived nuclear wastes is an engineered barrier system consisting of the waste form and a canister or container forming a protective sealed envelope for the waste form. Barriers between the container and the geologic site also play a role in the engineered

Table IV-18. Radiation Dose Limits for Exclusive-Use Shipments

-
- 1000 mrem/h at accessible external surface of package^{a,b}
 - 200 mrem/h at any point on outer surface of vehicle^b
 - 10 mrem/h at any point 2 m from vertical planes represented by outer lateral surfaces of vehicle^b
-

^aPackage secured so that it remains fixed during transportation, and no loading or unloading operations are undertaken between beginning and end of transportation.

^bClosed vehicle.

system. One concept for such a system being investigated at PNL [WESTERMAN] is shown in Fig. IV-5. The primary barrier in these concepts is the canister, and appropriate materials are being investigated for use in these containers [PITMAN]. The container must not be degraded by the contained waste or by interaction with the barriers or other components in the geologic site. Corrosion tests [PITMAN] were conducted on various proposed container materials when exposed to a potential groundwater or to a brine solution (salt dome). Results of these screening tests indicated that various nickel alloys, titanium alloys, and austenitic stainless steels exhibited low corrosion rates; however, the austenitic stainless steels could be subject to chloride stress corrosion cracking. Ceramic and polymeric materials are also being investigated as isolation barriers between the canister and the geologic repository environment [FULLAM]. The container material would have to be evaluated against any selected barrier material for corrosion or other reactions.

Several alloys were also investigated by Sandia [MAGNANI] for corrosion resistance suitable for a 300 y lifetime. Titanium-Code 12 was found to be a very corrosion-resistant material in bedded salt or seabed environments and worthy of further investigation.

Materials suitable in clay environments are being investigated at Centre d'Etudes de l'Energie Nucleaire, Mol, Belgium [CASTEELS]. In the clay environment, sulfur appears to affect reactions with the metals, and Hastelloy C provided the best corrosion resistance.

3. Packaging for Shallow-Land Burial

a. Definitions and Present Status

An EPA-sponsored survey describes the types of packaging most commonly used in the management of LLW [MILLER]. The conclusions of this survey are summarized here. For this discussion, it is understood that the term low-level wastes is used to describe any radioactive wastes other than (1) high-level (the raffinate from the first cycle of the reprocessing of spent nuclear fuel), (2) TRU (wastes containing ≥ 10 nCi/g of transuranium nuclides or U-233), and (3) mine or mill tailings.

- Number of engineered barriers, exclusive of waste form 4 (canister, concrete, polymer, backfill)
- Materials for canister: Cu, Cu-base alloys, Fe base, Ni-base alloys
- Materials for hole sleeve: Concrete, polymer concrete, ceramics, graphite

Fig. IV-5. Engineered Barrier System Compatible with Both Oxidizing and Anoxic Environments, Featuring Massive Non-Metallic Hole Sleeve

In the discussion of packages and packaging, the term "packaging" refers to all containment components, including wrappers, structural members, shielding, seals, and covers. The term "package," on the other hand, refers to the combination of packaging, plus cargo that is contained within it; in this case, the radioactive waste.

One of the main concerns pointed up by this survey is the lack of common basis of criteria and standards for packaging. This situation is expected to be remedied by the adoption of 10 CFR 61. The survey of [MILLER] is useful for its description of the types of containers most commonly used in SLB, *i.e.*, steel drums, larger steel containers, and non-metal packaging. A useful summary of such packaging is also provided in [ONWI-20].

b. Criteria, Standards, Regulations for Packaging of LLW

In the past, the packaging of LLW has been guided by tenets from three sources: rules of the waste-generating facility, Federal transportation regulations, and rules of the waste-accepting facility. Criteria for acceptance of packages at commercial burial grounds have generally coincided with the DOT construction requirements, although some additional requirements have been imposed by the generating or burial facilities, mainly to accommodate whatever drum-handling equipment is available. Transport regulations have been constructed to ensure container integrity during transport, without regard for performance in burial.

Although the packaging is considered responsible for containment of LLW in transportation, this is not the case at the burial grounds. At these sites, it is the soil rather than the packaging that has been considered to be the containing element. Although DOT specifications on

containers are intended to ensure their integrity during transportation, the containers are not strong enough to survive some operations used to place containers in SLB trenches.

The DOT spec. 17C or 17H drums are required to sustain a drop of 4 ft onto concrete. In the past, such containers have been dropped from heights greater than 4 ft into SLB trenches, in some cases onto other containers [MILLER]. Present-day burial operations treat containers with more care [LEVIN-1982].

c. Steel Drums

The container most often used for packaging of LLW is the steel drum. Steel drums are relatively inexpensive, easy to handle, and have been accepted at all SLB sites. Although 55-gal drums are most often employed, drums of 30-gal and smaller capacities have also been used.

Drums used to package LLW are usually coated on all surfaces with paint, but such coatings are not expected to significantly extend lifetimes of drums in contact with soil. Additional containment within the steel drum, consisting of plastic bagging or a rigid polyethylene drum, is commonly used. When shielding is required, liners of concrete or other material have been placed inside the drums, but limits on total weight are specified both by DOT rules and by acceptance criteria of burial grounds. Usually non-disposable, reusable exterior shields are used for transportation and handling.

For some cargoes, the steel drum offers the desirable combination of acceptability for transportation (DOT-approved drums) and also acceptability for burial. For other cargoes, however, steel drums qualify for burial but not for shipping. In such cases, the drums are shipped within an overpack that provides the required shipping qualifications. For example, the "Super Tiger" is an unshielded, Type B overpack that can contain up to 42 drums (55-gal) for transport by truck, rail, or barge [JEFFERSON-1976]. Shielded overpacks have smaller capacities. On arrival at the burial ground, the drums are removed from the overpack and buried. The overpack is returned for reuse. At present this is probably the most common methodology for shielding wastes in drums.

The 55-gal drum, typical of much of the packaging used for SLB, is often used with a concrete matrix containing up to 25 vol % waste [MILLER]. Excess water may be released from the matrix after the drum is closed, resulting in corrosion of the interior surfaces of the drum. The use of a concrete matrix gives structural support to the package so that the drum is less susceptible to being punctured. Moreover, in the event of drum puncture, the matrix retards dispersal of the waste to the environment.

d. Large Steel Containers

Containers larger than the 55-gal drum have come into use for low-density wastes that must be disposed of regularly in large volume. These containers have capacities many times that of the 55-gal drum. For example,

low-activity LLW has been shipped in containers of 1200-1400 gal* with shielding equivalent to 2 in. of lead. Also, LLW of higher activity has been shipped in containers of 500 gal with shielding equivalent to 4 in. of lead. These containers are constructed from 1/4-in. carbon steel, using welded seams. They are transported in a Type B overpack to prevent damage from impacts. With these large containers, a matrix such as cement is not used, probably because of weight limitations. Without the support of a matrix on inside surfaces, they are susceptible to damage from impacts. Because of their 1/4-in. walls, these containers can be expected to have a much longer lifetime in soil than steel drums which have much thinner walls.

e. Non-Metal Containers

In general, non-metal packaging has been more readily accepted at DOE burial sites than at commercial burial sites. Cardboard boxes are unacceptable at commercial burial sites, and other non-metal packaging has been limited to DOT-approved designs. At DOE burial sites, on the other hand, examples of non-metal packaging include (1) sealed plastic bags within cardboard boxes or fiberboard drums, (2) plastic-lined fiberboard boxes, and (3) liners of PVC or fiberboard within DOT 7A plywood boxes.

Fiber-drum packaging is representative of practices at DOE burial grounds where public transportation systems and, therefore, DOT regulations are not involved. Packages involving fiber drums are typically carried from a lab area and dumped directly into a burial trench.

F. Interim Storage

Interim storage is commonly understood to be an arrangement for temporarily storing a waste form after conversion of the primary form to a suitable storage form and packaging, but before emplacement in a terminal storage situation. In some cases, re-packaging may be planned between interim storage and terminal storage. The general system of options is diagrammed in Fig. IV-6.

Interim storage might be used for several reasons, only some of which may be useful for airborne wastes. (1) The radioactive isotopes may be allowed to decay to reduce the heat generation rate or the specific activity to levels necessary to meet transportation regulations or acceptance criteria at a desired terminal storage facility. This isotope decay would avoid distributing the waste among an unreasonably large number of packages or committing uneconomically large amounts of shielding to terminal storage. (2) Interim storage may be necessary to permit time for decisions to be made on selection of waste form and the process by which it will be produced. (3) It may be necessary to store a waste pending the R&D required to develop the process operations used to produce the terminal waste form. (4) It may be necessary to store a waste to await regulatory developments, including formulation of criteria and promulgation of regulations. (5) Interim storage may be necessary pending the design and construction of a terminal disposal facility.

^a Containers with volumes of up to 240 ft³ (~1800 gal) have been used [LEVIN-1982].

Fig. IV-6. Options Related to Interim Storage

At present, TRU wastes are being received for interim storage (or "retrievable" storage) at a facility at Hanford, operated by the Rockwell-Hanford Company [MCCALL] and also at the Radioactive Waste Management Complex, operated by the EG&G Co. on the INEL site [ANDERSON].

For Kr-85 and tritium, which have half-lives of only about one decade, interim storage until the radioactivity amounts are low enough to permit release, with dilution by air to the environment, has been suggested. A detailed description of a facility for storing krypton at an FRP site is designed with the intention of storage for 50 y, at which time controlled release to the atmosphere would be started [DOE/ET-0028]. Less-detailed concepts for interim storage of tritiated water [NEA, GRIMES] have similarly suggested storage for about the same time period, ending in discharge to the environment.

Although most of the technology of interim storage of non-high-level solid wastes is aimed at TRU-contaminated wastes, some technology seems adaptable to interim storage of airborne wastes. For example, the brief summary of packaging for interim storage of TRU wastes [ERDA-76-43A] might be applied to interim storage of solid airborne waste forms.

V. SUMMARY

In preparation for an analysis of compliance of technologies for waste management of the airborne nuclides (H-3, I-129, C-14, and Kr-85), an information base was assembled and is summarized here. The topics considered in this assembly include processes for the collection of airborne nuclides from streams of fuel reprocessing plants, conversion of primary waste to forms suitable for disposal, requirements on waste forms and packaging made by transportation regulations and by technical criteria for final disposal, and characteristics of disposal facilities and the procedures used at them in emplacement of waste packages. The disposal modes considered include shallow-land burial, greater confinement disposal, deep geologic disposal, temporary storage on the site of the fuel reprocessing plant, and ocean disposal.

Based upon this information, a complete waste management system that could be assembled for each nuclide from the technologies that have been developed to date will be briefly described and then the question of whether such a system could comply with the applicable regulations will be addressed.

The survey reported here indicates that much information relevant to management of airborne nuclides is available. It also indicates that, although the technologies have been tested at varying fractions of full-scale and at varying degrees of simulation of real systems, none has been tested in full-scale use.

In general, most of the development of collection processes and waste forms seems to have been done before the regulations that define the criteria to be met by the waste management technology were issued in final form. Furthermore, many of the concepts or models of expected performance of waste management technology in full-scale practice have not been developed in the detail or in the form that would permit ready comparison with the requirements of regulations, especially requirements that are expressed as quantitative limits on dose rates. The large body of information assembled here seems to be sufficient, however, to provide the material for predictions on where compliance may and may not be expected and indications on where development of either technology or regulations would contribute to yielding complete disposal arrangements for the airborne nuclides.

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