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MANAGEMENT OF RADIOACTIVE WASTE GASES FROM THE NUCLEAR FUEL CYCLE — VOLUME I COMPARISON OF ALTERNATIVES

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GASES FROM THE NUCLEAR FUEL CYCLE —
VOLUME I, COMPARISON OF ALTERNATIVES**

NUREG/CR--1546-Vol.1

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ABSTRACT

Alternatives were compared for collection and fixation of radioactive waste gases released during normal operation of the nuclear fuel cycle, and for transportation and storage/disposal of the resulting waste forms. The study used a numerical rating scheme to evaluate and compare the alternatives for krypton-85, iodine-129, and carbon-14; whereas a subjective evaluation, based on published reports and engineering judgment, was made for transportation and storage/disposal options. Based on these evaluations, certain alternatives are recommended for an integrated scheme for waste management of each of the subject waste gases. Phase II of this project, which is concerned with the development of performance criteria for the waste forms associated with the subject gases, will be completed by the end of 1980. This work will be documented as Volume II of this report.

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1.0 INTRODUCTION AND SUMMARY

The Environmental Protection Agency (EPA) has adopted standards (40 CFR Part 190*) that will limit the release of krypton-85 and iodine-129 associated with the normal operation of the nuclear fuel cycle. The EPA limits for these radioactive gases, which are scheduled to take effect for commercial plant operation in 1983, are 0.005 Ci/GWe-yr for iodine-129 and 50,000 Ci/GWe-yr for krypton-85. While these standards do not contain limits on the release of carbon-14, EPA has announced its intention to consider setting appropriate levels of maximum environmental burden as information becomes available.

Current Nuclear Regulatory Commission (NRC) regulations permit the release of the radioactive gases by the method of dilution and dispersion, provided that certain concentration limits are not exceeded. Consequently, the Savannah River Laboratory (SRL) was engaged by the Probabilistic Analysis staff of the NRC to conduct a study to develop a data base for making decisions involving the management of these gaseous wastes.

The study is divided into two phases. Phase I involves a preliminary identification and comparison of the various alternatives for collection and fixation of the radionuclides and for transportation and storage/disposal of the resulting waste forms. The results of Phase I are documented in this report. Phase II of the study was originally intended to include a detailed evaluation and assessment of the most promising alternatives selected in Phase I. However, the objectives of Phase II have subsequently been narrowed to focus primarily on final disposition of the waste forms and the development of performance criteria for these waste forms. Work on Phase II is in progress and will be completed in the early calendar year of 1981.

* Code of Federal Regulations, Title 40: "Protection of Environment"; Chapter 1, Subchapter F, Part 190: Environmental Radiation Protection Standards for Nuclear Power Operations (40 CFR 190) (Revised July 1, 1977). (See Federal Register, 42 [No. 9], Part VII, pp. 2858-2861 [January 13, 1977] for comments on future standards.)

An examination of the sources of the radioactive gases indicates that most of these isotopes are released during the reprocessing of spent fuel. Thus the current study is focused on the treatment of the off-gas streams in a fuel reprocessing plant (FRP). Viable alternatives for collection, fixation, transportation, and storage/disposal of the iodine-129, krypton-85, and carbon-14 were identified and evaluated based on selection criteria. In the case of collection and fixation methods, a numerical rating scheme was devised for evaluation and comparison of the alternatives. For transportation and storage/disposal, a subjective evaluation was made based on previously published studies and engineering judgment.

Based on the numerical evaluation scheme for collection and fixation methods, the most promising alternatives are as follows:

- Iodine-129 is collected on silver-exchanged mordenite and stored as silver iodide in an interim storage facility. The eventual recovery of the silver is anticipated in this alternative. The ultimate disposal form will probably be lead iodide (HI loaded onto PbX).

A second alternative would be collection on beds of Ag NO_3 impregnated amorphous silica gel. No silver recovery scheme has been developed for this collection medium; thus the silver iodide would be the ultimate disposal form for this alternative.

- Carbon-14 is collected on a barium hydroxide bed and stored as barium carbonate encased in concrete.

Collection on molecular sieve beds followed by fixation on barium hydroxide beds is an alternative collection scheme. The resulting barium carbonate waste form is the same as that above.

- Krypton-85 is collected by the fluorocarbon adsorption technique followed by xenon separation utilizing cryogenic cold trap techniques. Pressurized cylinder storage of the purified krypton gas in a dry-well storage field is recommended until alternative fixation techniques are better developed.

Collection by cryogenic distillation is an alternative. Solid adsorption techniques may also be a viable alternative when further developed. For either method, pressurized cylinder storage would be used.

Ion implantation and zeolite encapsulation are presently the most promising fixation methods to obtain a waste form suitable for permanent storage/disposal for krypton-85.

Transportation can be by rail, truck, or barge. All three gases could be stored temporarily in onsite, above-ground facilities in the fixed waste forms previously described, although dry-well storage of krypton is preferred.

Due to the extremely long half-life of iodine-129 (~16 million years) permanent storage of iodine-129 should be limited to locations where isotopic dilution can be reasonably well assured when the storage package fails. Deep ocean sediment disposal of iodine offers the best assurance of isotopic dilution.

Carbon-14 (half-life of ~5700 years) could also be disposed of in deep ocean sediments, although geologic storage is a viable alternative for this waste gas.

In addition to dry-well storage, krypton-85 (half-life of ~11 years) can be buried in deep ocean sediments and in geologic repositories. Any of these three disposal options could be employed with ion-implanted or zeolite-encapsulated krypton-85.

Recommended processes for the collection, fixation, transportation, and storage/disposal together comprise the best integrated scheme for waste management of each of the waste gases in the judgment of the authors. For all three isotopes, the determining factor in developing the integrated management scheme was based on the evaluation of the collection/fixation method. This is the case because, given a method for the ultimate disposition of the waste form (e.g., isolation in terrestrial geologic media or in deep sea sediments), the overall risk of the integrated scheme becomes not so much a function of the waste form but rather of the effectiveness with which the gas can be collected and fixed. This is apparent for krypton-85 since each of the three processes considered produces the same end product, purified krypton gas. For carbon-14, the choice of CaCO_3 or BaCO_3 as a waste form was not as important as whether the method of collection was compatible with collection of the other gases. For iodine-129, neither the iodate salts nor the iodide salts of silver or lead can assure protection against ultimate release. Thus, minimizing risks at the collection/fixation stage and providing isotopic dilution for the storage form achieves the necessary population protection.

Based on these evaluations, a reference off-gas treatment system for a reprocessing plant was selected. The reference system is included as Appendix A of this report. The reference waste forms will be used to develop performance criteria for Phase II of this project.

2.0 SOURCES OF WASTE GASES

2.1 U. S. Nuclear Industry

Iodine-129 and krypton-85 are primary fission products in the nuclear fuel cycles based on uranium-233, uranium-235, and plutonium-239. Carbon-14 is produced by neutron interaction with isotopes of oxygen and nitrogen present in various reactor components and is not a fission product. More than 95% of the gaseous radionuclides present in irradiated fuel are released during the dissolving of the fuel elements.¹ Thus the major source of these gases in the normal fuel cycle is the reprocessing plant.

About 28% of the total carbon-14 produced in the nuclear fuel cycle is found in the reactor moderator as a result of activation of the isotope oxygen-17 (present in trace quantities in water). The carbon-14 is present as organic compounds or carbon dioxide depending on the reactor type (Pressurized Water Reactor - PWR, or Boiling Water Reactor - BWR). No technology is readily available for the recovery of this carbon, and this small source of carbon-14 is neglected in this study. Appreciable krypton-85 and iodine-129 may be released to the reactor containment building during a reactor accident, such as the incident that occurred at Three Mile Island Unit I. However, EPA regulations governing the release of these isotopes do not apply to the accident situation. In any event, the quantities released accidentally represent a small fraction of the total untreated releases from a fuel reprocessing facility, because such accidents are rare and because the quantity of fuel involved is small when compared to the anticipated fuel throughput in a reprocessing plant and subsequent normal, untreated releases.

Based on this consideration of the sources of radioactive waste gases, the emphasis in this study is centered on the collection, fixation, and eventual disposal of krypton-85, iodine-129, and carbon-14 released during nuclear fuel reprocessing.

For the gaseous fission products (iodine-129 and krypton-85), the quantity of each isotope present in irradiated fuel will depend on the fuel type, irradiation history, and cooling time after discharge from the reactor. For this study, Light Water Reactor (LWR) fuel is assumed with UO_2 fuel burnup of 35,000 MWD/MTHM (thermal MW) and a cooling time of one year. The FRP is assumed to have a 10 MTHM throughput capacity and operates 300 days/year or at a total capacity of 3000 MTHM/year. If a thermal efficiency of 34% is assumed for the typical LWR, the fuel processed represents 90 GWe-years of electrical energy production.²

Carbon-14 is formed from five reactions of neutrons with isotopes of elements that are normal or impurity components of fuel, structural materials, and cooling water of LWRs. These neutron induced reactions are as follows:

1. $C-13(n,\gamma)C-14$
2. $N-14(n,p)C-14$
3. $N-15(n,d)C-14$
4. $O-16(n,He-3)C-14$
5. $O-17(n,\alpha)C-14$

Reactions 2 and 5 are the only significant contributors to the carbon-14 inventory in LWR fuel.³ Oxygen-17 is present in trace quantities in the oxygen in the UO_2 fuel. Nitrogen is a trace impurity (as the nitride) in fuel and cladding materials. Total calculated production rates for carbon-14 in LWR fuels are summarized in Table 1.

TABLE 1

Carbon-14 Production Rates in LWR Fuel³

<u>Reactor Type</u>	<u>C-14 in Fuel, Ci/GWe-yr</u>
BWR	
Low*	9.0
Medium*	17.6
High*	46.3
PWR	
Low	9.6
Medium	18.8
High	49.5

* Low, medium, and high refer here to the range of measured levels of carbon-14 in specific operating reactors of each type.

The source terms used for the reference FRP are summarized in Table 2. These values were derived from several published reports on reprocessing and are briefly discussed in the following paragraphs.

TABLE 2

**Source Terms for Radioactive Gases in
the Reference Fuel Reprocessing Plant
(3000 MTHM/yr, 35,000 MWD/MTHM)**

	<u>Iodine</u>	<u>Carbon*</u>	<u>Krypton</u>
Inert Isotopes, kg	175	2733	970
Radioactive Isotopes			
Kilograms	724	1.3	82.2
Curies	126	1714	3.2×10^7
Total Waste, kg	899	2734	1052
Fixed Waste Form	AgIZ	BaCO ₃	Gas Cyl.
Fixed Waste Weight, kg	10230**	12260	1052†

* Weights for carbon are calculated as CO₂.

** This weight includes 899 kg of halides and 1860 kg of silver.

† Fixed waste would be stored in ~252 cylinders.

2.1.1 Iodine-129

Published values for the fission product halide content of typical LWR fuels were selected from References 4, 5, and 6. Corrections were made for differences in irradiation histories to obtain estimates of the total halide collection requirements for the reference FRP to obtain the values shown in Table 2.

Annual waste generation rates and silver consumption rates were based on the following assumptions:

- Reduced, silver-exchanged mordenite (AgZ) will be used in the collection beds (see Appendix A).
- Bed loadings of 100 mg I₂/g AgZ (Reference 5).
- 20% by weight of the AgZ is silver (Reference 5).

2.1.2 Krypton-85

Source terms for krypton-85 for the reference facility were obtained by scaling up published source terms for the Allied General FRP at Barnwell, SC (AGNS).² Complete separation of xenon from the krypton gas was assumed to reduce the required storage volume. The reference storage form for krypton-85 for this study (Appendix A) is bottled gas in pressurized cylinders housed in an interim storage facility. Each cylinder [modified Department of Transportation (DOT) Specification 3A-2015] will contain about 128 kCi of krypton-85 at 34 atmospheres of pressure (500 psi).⁷ The reference plant will recover about 108 kCi per day of krypton-85 in a total krypton volume of 978 L/day (STP). If xenon is not separated from the krypton, the total daily volume of noble gas collected would increase to 10,030 L/day (STP). One cylinder of separated krypton will contain about 1.2 days of production from the FRP.

2.1.3 Carbon-14

Davis³ uses a value of 0.030 GWe-yr/MTHM for typical LWR, so the median production rate of carbon-14 would be 1584 Ci/yr for BWR fuel processed in the reference FRP and 1692 Ci/yr for PWR fuel. Idaho National Engineering Laboratory⁶ (INEL) uses a value of 0.622 Ci C-14/MTHM processed, or a throughput rate of 1866 Ci/yr for a 3000-MTHM/yr FRP.

For the present study, an average of the three values, or 1714 Ci/yr, was assumed. This is the equivalent of 384 g/yr of carbon-14 or 1.26 kg of $^{14}\text{CO}_2$ /yr. The radioactive species will be diluted by atmospheric CO_2 present in the process gas streams. In this study, a combined flow of 400 cfm air is assumed (200 cfm each from the head end off-gas system and the dissolver off-gas system, see Appendix A). Thus the total CO_2 collected will be about 2734 kg/yr as shown in Table 2.

2.2 Global Perspective

Based on a survey of the world production of gaseous radioactive wastes,¹ the following conclusions with regard to the magnitude of the U.S. waste gas problem are apparent:

- Any reduction in carbon-14 releases from U.S. fuel cycles would have little impact on the atmospheric or biospheric inventory. The annual natural production (from nitrogen in the upper atmosphere) and the atmospheric inventory (about 50% from bomb tests) of carbon-14 are about 20 and 4000 times greater, respectively, than the untreated annual release of carbon-14 from the U.S. fuel cycle.

- The U.S. carbon-14 problem is insignificant on a global impact basis. The reactor system used by the USSR releases about 12 times as much per unit of power produced.
- Essentially all of the krypton-85 inventory is man-made and resides in the atmosphere. Currently, 88% of that inventory comes from foreign bomb tests and the world nuclear fuel cycle, 10% comes from U.S. bomb tests and weapons production, the remaining 2% are current U.S. releases. However, the amount of krypton-85 accumulating in the U.S. spent commercial fuel inventory is predicted to be 25% of the total. The total accumulated krypton-85 from the world nuclear electric power cycle is predicted to be five to six times the current inventory.
- The uncontrolled iodine-129 releases from the U.S. nuclear fuel cycle would exceed the natural and man-made inventory in one year if a commercial reprocessing plant was operating. Actual U.S. releases are about 2% of the current inventory in the biosphere. The amount of iodine-129 accumulating in U.S. spent commercial fuel rods is predicted to be about twice the current inventory by 1980.

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3.0 COMPARISON METHODOLOGY

3.1 Background

The present practice of atmospheric dilution and dispersion of the radioactive gases iodine-129, krypton-85, and carbon-14 may be accompanied by specific risks to present and future generations of mankind in the form of very low-level radiation exposure received by the global population. (It should be noted that the risk of small quantities of these gases in the atmosphere is speculative.) This report makes no assessment of the possible risks associated with the dispersion of the gases in the atmosphere; it simply identifies and compares possible means of reducing such airborne quantities. Reduction in the global population dose to the present generation can be accomplished by collection and fixation of the waste gases during the fuel reprocessing step of the nuclear fuel cycle. The fixed wastes must be stored for periods of time ranging from a hundred years to more than a hundred million years to reduce the global population dose to all future generations.

Several methods of collection and fixation of the gases have been studied or used in existing FRPs and other nuclear facilities in the U.S. and in other countries. Comparative analysis of these methods is a complex task since the waste form from a particular collection scheme often determines the available fixation options. The analysis is further complicated by the fact that no performance criteria have been established for the fixed waste forms, and the methods of storage and/or disposal have been the subject of considerable scientific and political discussion.

To reduce the complexity for the present comparative analysis, a number of assumptions had to be made with regard to desired features of the collection/fixation process and the storage characteristics of the resulting waste form. These assumptions were used in the development of certain bases for comparison of the alternatives as described in Section 3.2.

Using the bases of comparison as a starting point, specific criteria were then established for ranking the collection and fixation phases of the radioactive waste gas management process. A detailed numerical scheme (described in Section 3.3) relying on engineering judgment was developed for these criteria to facilitate the comparison. For the transportation and storage/disposal phases of the management process, previously published evaluations were used rather than attempting to develop a new methodology.

3.2 Bases for Comparison

The bases for comparing alternative methods for the collection, fixation, transportation, and storage/disposal of iodine-129, krypton-85, and carbon-14 are outlined below:

1. Compared to the current method of atmospheric dilution and dispersion, alternative processes must have the potential for reduction of risks to the world-wide population. Risk was considered in terms of radiation exposure to present and future generations of this population.
2. The potential risk from all phases of each alternative must be considered. These phases include collection, fixation, interim storage, transportation, and ultimate disposition.
3. In addition to risk, the other process characteristics considered were:
 - a. Demonstrated methods of recovery and fixation.
 - b. Adequate process efficiencies (DF's) during the collection phase.
 - c. Demonstrated technology for safe transportation and interim storage of the fixed waste form.
 - d. Adequate safeguards against undiluted release of the concentrated wastes in any disposal concept.
4. For cases where no demonstrated technology exists for any phase of the operation, the best developed conceptual processes are specified based on engineering judgments.
5. It was assumed that all packaged waste forms are stable to short term contact with water, brine, or other aqueous media which could be encountered following a transportation accident.
6. It was assumed that all nonradioactive constituents (such as nitrate, nitrite, mercury, silver, and lead) can be safely released to the biosphere as long as they are sufficiently diluted. Separate assessments were not made for potential chemical hazards of the waste after radioactive decay.
7. For iodine-129 and carbon-14, it was assumed that the highest risk uptake pathway (critical pathway) to man is via the atmosphere during the collection and fixation phases. The critical pathway is assumed to be drinking water during the transportation, interim storage, and ultimate storage phases.

8. The critical exposure pathway for krypton-85 is always via the atmosphere since this isotope is chemically inert and is not known to be biologically concentrated or retained.
9. Due to the extremely long half-life of iodine-129 (16 million years), it must be assumed that any geologic or ocean floor storage scheme for this isotope will provide only a temporary barrier against eventual release to the environment. Thus any earthbound disposal technique for iodine-129 must include assurances that isotopic dilution will take place to minimize doses to man in the distant future.

3.3 Criteria for Collection/Fixation Comparisons

As an aid in selecting the reference processes for collection and fixation of the gaseous wastes, a numerical rating system was devised. The rating system provides a basis for relative evaluation of the candidate processes. In this scheme, seven criteria were established for comparing collection techniques, and six of the same criteria were used to compare fixation techniques (the decontamination factor was omitted from the fixation evaluation).

For each criterion, candidate processes were assigned a "relative numerical rating" on a scale of 1 to 10, based on their ability to meet the criterion in question. A low rated process was assigned a value of 1, 2, or 3, medium was assigned a value of 4, 5, or 6, and high was given a 7, 8, 9, or 10, with 10 being the ideal or best rating. These assigned values, as well as the criteria themselves, are admittedly subjective, but were arrived by consensus among the authors and with the aid of other SRL personnel engaged in airborne waste management research.

Each criterion used in the evaluation was also assigned a weight factor in an effort to assess its relative importance in the overall immobilization and disposal process. These weight factors ranged in value from 1 to 3 (3 being assigned to the criterion judged most important). The product of the numerical rating value and the weight factor for each criterion is the relative evaluation factor for each process. The sum of the relative evaluation factors for each process formed the basis for selection of reference and alternative processes. A total of 266 separate judgments were required for the complete evaluation.

Criteria used and weight factors assigned are summarized below:

1. Simplicity (Weight Factor - 1.0)

The simplicity (or complexity) criterion deals with the number of operations, the amount and type of equipment required, and the ability to adapt the process to remote operation and maintenance. Large pieces of complex mechanical equipment that might require frequent or extensive maintenance should be avoided. The process should be easily understandable, easily controllable, and easily monitorable.

2. Reliability (Weight Factor - 1.5)

The processes to remove and to produce waste forms should be reliable. They must tolerate and recover from minor changes in operating conditions as well as more significant process upsets. They should produce a high yield with small recycle. The equipment should also be reliable; that is, it should be corrosion resistant, maintainable, replaceable, and require minimum redundant units. Quality assurance is also a part of reliability. Both the process and the equipment must be designed to facilitate sampling and analysis to determine an acceptable product and to assure minimum product loss.

3. Safety (Weight Factor - 2.0)

Process safety considers the use of high temperature, high pressure, and unstable chemicals. Lower ratings were assigned to processes which contain high potential energy because of the increased likelihood of an accident or release. Similarly, processes which require accumulation of large inventories of high energy-content products were given lower ratings because of the increased consequences of a release. Other specific factors considered in process safety are potential interaction with other processes, operating personnel exposure to radiation and toxic materials, and potential public exposure to hazardous materials.

4. Cost (Weight Factor - 1.5)

Detailed cost information is not available for many of the candidate processes because many have not reached the engineering stage of development. It was therefore necessary to rate each process on a relative basis using engineering judgment for the many factors that contribute to the overall cost of a particular collection or fixation method. Factors considered in the relative ratings included: capital equipment costs, equipment size and complexity, availability and cost of essential materials, chemical costs, manpower requirements,

energy consumption, materials handling costs, special shielding requirements, and other special construction requirements such as missile shields for high pressure operations or special ventilation designed to cope with potential pressure surges.

5. State of the Art (Weight Factor - 3.0)

State of the art refers to the state of development of the processes such as lab-scale, pilot-plant scale, or plant-scale. A large weight factor was applied to this criterion in recognition of the importance of proven technology. A lower numerical rating was given to alternatives only in the conceptual design stage. This was done because the lead time required to bring an untried design to the plant installation stage is sufficiently long that some candidate processes could not be available until well after the EPA regulations are implemented.

6. Material Handling (Weight Factor - 1.5)

Each process was rated on the basis of compatibility with preceding, parallel, and subsequent processing steps and the recoverability of essential materials. Ease of conversion of recovered waste to a suitable storage form is also a factor in the material handling criterion. Recovery/fixation schemes requiring the processing of powders or precipitates were rated lower than those based on fixed bed technology because the additional handling steps involved increase the likelihood of product loss or facility contamination.

7. Decontamination Factor (Weight Factor - 3.0)

The Decontamination Factor (DF) item rates each collection process on the basis of recovery efficiency, and is a direct measure of the chronic release of the waste gases to the environment. A large weight factor was assigned to this criterion to account for both present and potential future restrictions on radionuclide release. In addition, a high process DF (at least 1000) is necessary to achieve the desired system DF. Without this added safety factor, more costly redundant design would be necessary for the off-gas treatment facility to cope with such contingencies as system downtime and equipment malfunctions.

Evaluation of the various modes of transportation and long-term storage of the packaged waste gases were made using studies recorded in the literature. Numerical evaluations were not performed for the transportation phase of the study because such

analyses are already available. Insufficient data are available to permit numerical evaluation for all the storage/disposal options. Existing studies were used where available, and engineering judgments were applied in other cases to arrive at relative rankings.

The evaluations of the various modes of transportation used the following subjective criteria: risk, cost, surveillance and security, package integrity, leachability, and dispersibility. The criteria for ranking methods of long-term storage of the packaged waste gases used the following factors in addition to those used for transportation: state of the art, retrievability, facility stability, accessibility to future generations, and the ability of the storage scheme to ensure adequate isotopic dilution if isolation for at least 10 half-lives cannot be assured.

As indicated in the previous discussions of both the bases for comparison and collection/fixation criteria, consideration was given to the compatibility and integration of all necessary steps, from collection to the ultimate storage of the subject off-gases. When dissimilar but equally acceptable fixed waste forms were encountered for competitive processes, preference was given to the process requiring fewer and less-hazardous intermediate conversion steps to attain the final storage form. Additional attention was given to special storage requirements of the final waste form so that the integrated process (including all steps from collection to disposal) minimizes exposure to workers, the present generation of the general public, and future generations of the world-wide population.

Detailed descriptions of each alternative process and waste forms considered in this report and the evaluation of each are given in later sections.

4.0 COLLECTION AND FIXATION OF WASTE GASES

The various methods of collection and fixation of the gases iodine-129, krypton-85, and carbon-14 were compared using the numeric scheme outlined in Section 3.0. Results of the comparison are discussed in this section by isotope.

4.1 Iodine-129

4.1.1 Iodine-129 Collection and Fixation

Three generic methods are currently being considered for removal of iodine-129 from process air streams. These include:

1. Chemical reaction in a liquid scrubber system.
2. Chemical reaction in a granular solid bed.
3. Physical concentration in a liquid solvent system.

4.1.1.1 Chemical Reaction in a Liquid Scrubber System

Three recovery options for radioactive iodine from off-gas streams using liquid scrubbing techniques are the Mercurex, caustic and Idox processes.

4.1.1.1.1 Mercurex Process

The Mercurex process^{1,2} is a scrubbing technique which uses a solution of mercuric nitrate in nitric acid. The Mercurex process is described in the flowsheet for the off-gas system for the Barnwell Nuclear Fuel Plant's Separations Facility developed by Allied General Nuclear Services.³ The system is reported to be effective (99% recovery) in removing elemental iodine from off-gas streams, but is ineffective for organic species because of very slow kinetics for organic iodides. The Mercurex scrubbing process is a recovery process only, and additional process steps would be needed for immobilization. At present, there is no clear indication that the reagents can be recycled or that spent scrubber solutions containing inorganic salts can be converted to acceptable storage forms for iodine, although it may be feasible to develop methods for recycle of mercury salts (e.g., via electrolytic reduction to metallic mercury).

In the Mercurex process, the scrubbing solution normally is recirculated through a packed-bed scrubber with the removal of scrubber solution to waste when the mercury-to-iodine mole ratio drops from ten to four. This system is effective in removing elemental iodine from off-gas streams, but is ineffective for organic iodides. This system, like caustic scrubbing described below, is moderately inexpensive and is a well known technique. Disadvantages of the Mercurex process include: moderate DF, no known iodide recovery, consumes essential materials, and the toxicity of mercury.

4.1.1.1.2 Caustic Scrubbing

Caustic solutions of about 1 molar, with a reductant such as sodium thiosulfate added, remove elemental iodine from off-gas streams, but they are ineffective for organic iodides. Overall, caustic scrubbing has a low (50-90%) iodine-removal efficiency and therefore has limited applicability for iodine removal in the commercial fuel reprocessing industry. Application of a caustic scrubber on a dissolver off-gas stream where relatively large quantities of iodine are present could be expected to yield iodine retention factors on the order of 2 to 10. The caustic scrubbing media are typically employed in a packed tower through which the caustic solution is recirculated. Iodine containing caustic is held from the system, and fresh caustic is added to maintain caustic and carbonate concentrations within acceptable ranges. If sodium hydroxide is used as the scrubbing medium, plugging problems are frequently encountered in the column packing because of the low solubility of sodium carbonate. This problem is reduced by the use of potassium hydroxide as the scrubbing medium. Waste volumes generated are usually determined by the carbonate buildup and thus are proportional to the volume of air treated. Although this system is inexpensive and is a widely used technique, regeneration of the caustic for recycle from the nitrate salts in the caustic wastes appears infeasible. Disadvantages of caustic scrubbing include: low DF, sensitivity to acidic off-gases, additional waste volumes, and as mentioned above there is no known recovery process. In addition, the caustic scrubbing technique is not compatible with later carbon-14 recovery.

4.1.1.1.3 Iodex Process

The Iodex process uses concentrated nitric acid in the 20 to 22 molar range as the liquid scrubbing medium. The process, as demonstrated on a laboratory scale, effectively removes all iodine species from the off-gas (DF greater than 1×10^6). However, a

demonstration of system capability and operability over extended periods of operation is required. Also, long-term corrosion effects on applicable materials of construction need to be confirmed. The Iodox process, like the Mercurex and caustic scrubbing processes, requires additional steps for immobilization of the recovered iodine. Two main advantages of the Iodox process are the very high DF and that it removes all forms of iodine. Disadvantages of the Iodox process include: extreme corrosiveness of the scrubbing medium which can be pyrophoric, regeneration of the 22 molar nitric acid, and recovery of the iodate for immobilization.

4.1.1.2 Chemical Reaction in a Granular Solid Bed

One of the better techniques for both collection and fixation of iodine-129 in a single step is through the use of fixed beds of silver-exchanged or silver-impregnated granules of inorganic sorbents (activated carbon cannot be used because of the potential fire hazard). Three types of inorganic adsorbers have been evaluated extensively:

1. Silver-impregnated amorphous silicic acid (AC-6120 and AC-6120H1).
2. Silver-exchanged faujcrite (AgX).
3. Silver-exchanged mordenite (AgZ).

AC-6120 and AC-6120H1 have been used in the German reprocessing facility (WAK) for several years for the collection of iodine-131 and iodine-129.⁴ AC-6120H1, with its higher silver impregnation level, is the material in use at the WAK facility at present. It is highly efficient for the collection of all forms of gaseous iodine (both organic and inorganic iodine compounds). Silver utilization for Type H1 is reportedly better than for either AgX or AgZ. There is no demonstrated technology for silver recovery from either of these two German materials; however, use of these absorbers implies permanent removal of a valuable resource from the world economy. Both materials operate more efficiently in the presence of nitrous oxide (NO).

AgX is also highly efficient for collection of all forms of gaseous radioiodine. However, the inorganic substrate (Type 13X zeolite) is subject to attack by acids. Thus, any NO_x encountered during process upsets could damage or destroy the collection beds. Silver can be recovered from AgX by stripping the iodine off the bed with hot hydrogen (500°C). The original process used pure hydrogen gas,⁵ but a newer, safer technique has

been developed using a mixture of 4% hydrogen in argon.⁶ The HI generated during the recovery process is collected on lead-exchanged zeolite (PbX) very efficiently. PbX cannot be used directly in the off-gas collection system because the substrate is not acid resistant. Recovered beds of AgX could not be reused, because the efficiency for iodine trapping is lowered by the recovery process.

AgZ, particularly in the reduced form, is also highly efficient for collection of all forms of gaseous radioiodine. The loading capacity for methyl iodide has been found to be about 1/3 of that for elemental iodine in recent studies.⁶ The mordenite substrate is acid resistant so process upsets (failure of the NO_x destructor to operate properly) are less likely to damage the beds than if AgX was used. Silver recovery or bed reuse is possible with AgZ using the hot hydrogen techniques mentioned above. AgZ is more efficient in the absence of NO_x and in the presence of some moisture in the off-gas stream.

A decontamination factor of at least 1000 is expected with any of the above sorbents for the processing of spent LWR fuels. The advantages of this type of system are: it is simple, the adsorbent is nonflammable, elevated bed temperatures can be used (minimizing the need for cooling the process gas stream), the waste product is a dry solid easily handled and contained, the chemisorbed iodide is highly insoluble, and the adsorbent is highly efficient for the removal of both organic and inorganic iodides from off-gas streams. The disadvantages are: high cost of silver and the subsequent loss of a valuable resource by a once-used throw-away process, and sensitivity to fluoride, chloride, and oxides of nitrogen in the off-gas.

4.1.1.3 Physical Concentration in a Liquid Solvent System

Current investigations for the development of liquid phase physical concentration of iodine are limited to the ORNL fluorocarbon recovery system (FCRS). This system is reported to be capable of accumulating iodine, carbon, and krypton in a single process. Methods for recovery and fixation of iodine from the fluorocarbon solvent have not been developed, however.

4.1.2 Comparison of Alternatives for Iodine-129

A comparative summary of the collection schemes for iodine under consideration is presented in Table 3. The candidate processes were also analyzed using the evaluation criteria outlined in Section 3.0. The results of this analysis are summarized in Table 4. The analysis shows that the fixed bed chemical sorber system is the better of the candidates for iodine collection.

TABLE 3

Comparison of Iodine Collection Alternatives

Collection Alternative

1. Chemical Reaction in a Liquid Scrubber System

a. Iodox Process

- State of the Art
 - Lab/Pilot-Plant Scale
- Decontamination Factor
 - Greater than 1.0×10^6
- Waste Form
 - 22 Molar Nitric Acid-Iodate Slurry
- Advantages
 - High DF
 - Removes all Forms of Iodine
- Disadvantages
 - Very Corrosive Media
 - Can be Pyrophoric
 - Regeneration of 22 Molar Nitric Acid
 - Iodate Recovery Technology Not Developed

b. Mercurex

- State of the Art
 - Plant Scale
- Decontamination Factor
 - 1.0×10^2
- Waste Form
 - Mercuric Iodate Slurry
- Advantages
 - Moderately Inexpensive
 - Known Technology

TABLE 3 (Continued)

- Disadvantages
 - Toxicity of Mercury
 - Consumes Essential Materials
 - Iodate Recovery Technology Not Developed

c. Caustic

- State of the Art
 - Plant Scale
- Decontamination Factor
 - Less than 10
- Waste Form
 - Caustic Slurry
- Advantages
 - Inexpensive
 - Known Technology
- Disadvantages
 - Low DF
 - Sensitive to Acidic Vapors
 - Adds Waste Volume
 - No Known Recovery Process
 - Not Compatible with Carbon-14 Recovery

2. Chemical Reaction in a Granular Solid Bed

a. Silver Zeolites & Mordenites

- State of the Art
 - Pilot-Plant Scale
- Decontamination Factor
 - Approximately 1.0×10^3 (with pretreatment)
- Waste Form
 - Fixed Solid

TABLE 3 (Continued)

- Advantages
 - Simple
 - Disposal Waste Form
 - Demonstrated Technology
 - Very Insoluble
 - Silver Mordenite (AgZ) regenerable
 - Silver Recovery Process Demonstrated
 - Disadvantages
 - Consumes Essential Material (Silver)
 - Hazardous Silver Recovery Process
 - Sensitive to the Presence of Chloride, Fluoride, and Oxides of Nitrogen in the Off-Gas
- b. Silver Silica Gel
- State of the Art
 - Plant Scale
 - Decontamination Factor
 - 1.0×10^3
 - Waste Form
 - Fixed Solid
 - Advantages
 - Simple
 - Disposal Waste Form
 - Demonstrated Technology
 - Very Insoluble
 - Better Silver Utilization
 - Disadvantages
 - Consumes Essential Material (Silver)
 - No Silver Recovery Process
 - Sensitive to the Presence of Chloride, Fluoride, and Oxides of Nitrogen in the Off-Gas

TABLE 3 (Continued)

3. Physical Concentration in a Liquid Solvent System

a. Fluorocarbon

- State of the Art
 - Not Demonstrated for Iodine Recovery
- Decontamination Factor
 - Approximately 1.0×10^4
- Waste Form
 - Unknown
- Advantages
 - Compatible with Krypton Recovery
 - Good for Organic and Inorganic Iodides
- Disadvantages
 - No Demonstrated Recovery Method
 - High Energy Consumption

TABLE 4**Evaluation of Iodine Collection Methods**

<u>Criteria</u>	<u>Mercurex</u>	<u>NaOH</u>	<u>Iodox</u>	<u>FCRS</u>	<u>Silver Zeolite</u>	<u>Silver Mordenite</u>	<u>Silver Silica Gel</u>
Simplicity	9.0	7.0	2.0	7.0	8.0	8.0	8.0
Reliability	10.5	9.0	3.0	6.0	10.5	13.5	13.5
Safety	14.0	16.0	6.0	10.0	18.0	18.0	18.0
Cost	13.5	13.5	4.5	7.5	9.0	9.0	7.5
State of the Art	30.0	30.0	6.0	12.0	30.0	30.0	30.0
Material Handling	6.0	6.0	3.0	4.5	13.5	13.5	12.0
Decontamination Factor	<u>9.0</u>	<u>3.0</u>	<u>30.0</u>	<u>24.0</u>	<u>24.0</u>	<u>24.0</u>	<u>24.0</u>
Total	92.0	84.5	54.5	71.0	113.0	116.0	113.0
Percent of Maximum	68.1	62.6	40.4	52.6	83.7	85.9	83.7

A comparative summary of the fixation schemes for iodine under consideration, with advantages and disadvantages, is presented in Table 5. The candidate fixation processes were also analyzed using the evaluation criteria given earlier. The results of this analysis are summarized in Table 6. The analysis shows that silver mordenite is the preferred fixation scheme (in conjunction with collection) primarily because of the ability to recover the silver loaded beds for reuse as a collector or for recovery of the silver in the beds.

TABLE 5

Comparison of Iodine Fixation Alternatives

Fixation Alternative

1. Iodox Process

Iodic Acid in Concrete

- State of the Art
 - Lab Scale
- Disadvantages
 - Solubility

Barium Iodate in Concrete

- State of the Art
 - Lab Scale
- Advantages
 - Relatively Insoluble

2. Mercurex

Mercuric Iodate in Concrete

- State of the Art
 - Lab Scale
- Advantages
 - Insoluble
- Disadvantages
 - Toxicity of Mercury

TABLE 5 (Continued)

3. Caustic Scrub

Iodine Crystals in Concrete

- State of the Art
 - Plant Scale
- Advantages
 - Known Technology
- Disadvantages
 - Large Waste Volume
 - Very Soluble Solid

4. Silver Zeolite

Fixed Bed of Iodine Loaded Silver Zeolite

- State of the Art
 - Pilot/Plant Scale
- Advantages
 - Simplicity
 - Known Technology
 - No Additional Processing Required
 - Silver Recovery Process Demonstrated
- Disadvantages
 - High Cost of Silver

5. Silver Mordenite (Without Regeneration)

Fixed Bed of Iodine Loaded Silver Mordenite

- State of the Art
 - Lab Scale
- Advantages
 - Simplicity
 - Known Technology
 - No Additional Processing Required
 - Silver Recovery Process Demonstrated
- Disadvantages
 - High Cost of Silver

TABLE 5 (Continued)

6. Silver Mordenite (With Regeneration)

Fixed Bed of Iodine Loaded Lead Mordenite

- State of the Art
 - Lab Scale
- Advantages
 - Reduced Costs
- Disadvantages
 - Uses Hot Hydrogen
 - PbI_2 More Soluble than AgI

7. Silver Silica Gel

Fixed Bed of Iodine Loaded Silver Silica Gel

- State of the Art
 - Plant Scale
- Advantages
 - Known Technology
 - No Additional Processing Required
 - Simplicity
- Disadvantages
 - High Cost
 - No Silver Recovery

8. Fluorocarbon

Not Demonstrated

TABLE 6

Evaluation of Iodine Fixation Methods

Criteria	Fixed in Concrete			NaOH Scrub as Iodate Crystals	Stored as Fixed Bed of Iodine Loaded on			
	Iodox as Iodic Acid	Iodox as Ba Iodate	Mercurex as Hg Iodate		Silver Zeolite	Silver Silica	Silver Mordenite	Lead Zeolite*
Simplicity	6.0	4.0	5.0	5.0	10.0	10.0	10.0	7.0
Reliability	6.0	4.5	6.0	6.0	12.0	13.5	13.5	12.0
Safety	8.0	8.0	10.0	12.0	18.0	18.0	18.0	10.0
Cost	6.0	6.0	9.0	10.5	10.5	9.0	10.5	12.0
State of art	12.0	12.0	15.0	21.0	27.0	30.0	27.0	15.0
Material handling	<u>7.5</u>	<u>7.5</u>	<u>9.0</u>	<u>7.5</u>	<u>13.5</u>	<u>10.5</u>	<u>13.5</u>	<u>13.5</u>
Total	45.5	42.0	54.0	62.0	86.5	91.0	92.5	69.5
Percent of Maximum	43.3	40.0	51.4	59.0	82.4	86.7	88.1	66.2

* Iodine stripped from Silver Zeolite and loaded on Lead Zeolite.

4.1.3 Iodine References

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4.2 Krypton-85

4.2.1 Krypton-85 Collection

Three processes are currently under development for the separation of krypton from process air streams. These processes are:

1. Cryogenic extraction and distillation.
2. Selective absorption in a fluorocarbon solvent.
3. Selective adsorption on silver mordenite solids.

4.2.1.1 Cryogenic Extraction and Distillation

This process is based on the separation of gases due to differences in their relative volatilities at low temperatures.^{1,2,3,4} The process equipment includes:

- A pretreatment train.
- A regenerative heat exchanger.
- A primary distillation column.
- A secondary distillation column for product purification.

The pretreatment train composition depends on the previous gas treatment before the krypton-85 removal step. Materials which can solidify in the cryogenic column and plug the system (if they are not removed to the few parts per million level) include water, carbon dioxide, iodine, oxides of nitrogen, and trace organics. Oxygen remaining in the gas stream will form potentially explosive ozone as a result of radiolysis when the krypton becomes concentrated.

It is assumed that all contaminants except oxygen have been removed in prior gas treatment steps. To remove oxygen, hydrogen is added to the off-gas stream, then heated to 400-500°C and passed through a catalytic recombiner. The oxygen and hydrogen combine to form water. Any trace nitrogen oxides will be reduced by the hydrogen to nitrogen and water. The water is removed by a condenser and by selective adsorption in packaged silica gel or molecular sieve beds.

The pretreated gas is first precooled in the regenerative heat exchanger before entering the primary recovery column. The gas is then contacted with downward flowing liquid nitrogen. The higher boiling components (xenon and krypton) are condensed and absorbed in the column. The remaining gases, including a slight excess of hydrogen, are released from the top of the column.

The absorbed krypton and xenon are concentrated in the bottom of the primary column and are transferred to the product column for purification by distillation. The overheads from the product column are fractionated into a krypton-rich fraction that is collected for storage. The less concentrated portion is recycled back into the system feed for rework. Relatively high purity xenon is recovered from the product column bottoms. The xenon can either be released or further purified for commercial sale.

The major advantage of the cryogenic absorption and distillation process is its advanced state of development. Liquid air plants using the same basic technology have been used for a number of years. A noble gas recovery plant of a design similar to the process described above has been operated at Naval Reactor Test Station (NRTS) for several years. The NRTS system was not designed for total krypton recovery, however, and the DF of that system is only about 10.

A total recovery cryogenic unit was installed in West Germany and operated for about two years. While a DF of 100 was obtained in their unit, xenon pluggage problems in the final product purification step along with other system reliability problems have resulted in unit redesign.⁵ The Germans are currently investigating use of a fluorocarbon recovery unit. The Japanese are having a cryogenic unit built which is to have a proprietary xenon removal step ahead of the cryogenic concentration stage. Anticipated krypton contamination of the xenon is high enough in this process to necessitate a separate xenon storage facility.⁶

The disadvantages of the cryogenic process include:

- A required high purity feed stream.
- Hazardous oxygen removal system.
- Relatively high energy consumption and operating costs.
- High energy-content system because of the low temperatures employed.
- Need for further process development for complete xenon removal.

4.2.1.2 Fluorocarbon Absorption System

The original design of the FCRS used four packed columns. The first three columns, the absorber and two strippers, perform the gaseous separation. The fourth column is used to purify the solvent for reuse. Oak Ridge has designed a new column which combines the first three columns into a single unit, thus saving considerable floor space. The single column system has been demonstrated over a two-year period on a pilot-plant scale.⁷

In the Oak Ridge single column FCRS system, prepurified gas is compressed to about 100 psi pressure and cooled to -30°C before entering the absorber section. Feed gas is passed countercurrently to the downflowing R-12 solvent. The noble gases, as well as any other gases present, are absorbed according to their respective solubilities. The stripped gas, containing mainly oxygen and nitrogen, is vented from the top.

The gas-laden solvent then enters the first stripper stage. The first stripper stage is also operated at 100 psi pressure, but with a temperature gradient ranging from -30°C to about 32°C (-22 to 90°F). In this stage, the loaded solvent is passed downward through an upward flowing vapor stream from a reboiler to further reduce the concentration of less soluble gases.

The product stream is extracted from the column after the first stripper stage. The more soluble species, including iodine, nitrogen dioxide, and water (if these gases are present) remain in the solvent and are removed from the bottoms of the solvent purification stage. The noble gas product stream contains up to 50% solvent along with CO_2 and nitrous oxide. Solid sorber beds are used to recover the solvent for reuse. Carbon dioxide can be removed in a solid absorbent bed (after solvent removal), while the krypton and xenon are further separated using cryogenic cold trap techniques.

The principal advantage of the FCRS system is its apparent tolerance for a wider variety of contaminant gases (such as NO_x , iodine, and CO_2) than the cryogenic system. However, the final concentration step still relies on cryogenic methods. While the FCRS has not been demonstrated on a full plant scale, sufficient experience has been gained in pilot plant operations to permit design of a full sized system.

The disadvantages of the FCRS system include:

- A high bay to house the column (up to 24 feet of vertical clearance is required).
- Fairly high energy requirements for solvent compression.
- Moderately high energy-content system because of the use of a pressurized, low boiling solvent (the German system will use subatmospheric pressure and lower temperatures to circumvent this objection).
- Lack of operating data for large scale units.
- Need for further process development for complete xenon removal.
- Potential fluorocarbon releases to the atmosphere.

4.2.1.3 Silver Mordenite Adsorption System

A new noble gas separation method is currently under development which utilizes the selective sorption characteristics of solid adsorbents. Studies to date show that activated, silver-substituted mordenites can be used at ambient pressure and temperature to separate xenon from mixtures of air and krypton.

The same material (or hydrogen mordenite) can also be used to separate oxygen from nitrogen and krypton at reduced temperatures.⁸ The product stream at the end of the oxygen separation step contains a mixture of about 1% krypton in pure nitrogen. The krypton can then be separated from the nitrogen in a cryogenic separation step utilizing liquid nitrogen as a coolant.

The separation technique utilizes multiple columns of solid adsorbent operated alternately so that one column is collecting the desired gas while the other(s) are being regenerated with a reverse flow of heated nitrogen. The number of columns needed is determined by the retentivity of the gas being collected. Laboratory studies show that only two columns are needed to separate xenon from air, while three columns are needed to effect the oxygen separation.

The basic technique has been used extensively for moderately sized air reduction plants. The adaptation of the technique for noble gas separation has only been demonstrated on a laboratory scale. Some column heating problems may be encountered in the regeneration steps when scale up to a full sized unit is attempted.

The advantages of the silver-mordenite separation technique include:

- Good separation of xenon.
- Minimum inventory of krypton-85 until the final concentration step.
- A low energy system operated at ambient pressure.
- Smaller space requirements than the cryogenic or older design FCRS systems.

The disadvantages of the silver-mordenite separation technique include:

- Incomplete development.
- Complex instrumentation required for process control.
- High cost of the silver mordenite.
- High purity feed stream required (iodine, nitrogen oxides, and carbon dioxide must be removed first).

4.2.1.4 Comparison of Alternatives for Krypton-85 Collection

A comparative summary of the krypton collection processes under consideration, with advantages and disadvantages, is shown in Table 7. The selection criteria methodology was applied to the krypton collection processes under study. The results are shown in Table 8.

TABLE 7

Comparison of Krypton Collection Alternatives

Collection Alternatives

1. Cryogenic Extraction and Distillation

- State of the Art
 - Plant Scale
- Decontamination Factor
 - Greater than 100
- Waste Form
 - Cold Liquid
- Advantages
 - Known Technology
- Disadvantages
 - Requires High Purity Feed
 - Hazardous Oxygen Removal Step
 - Xenon Separation Underdeveloped
 - High Energy Consumption
 - Large Space Requirements

2. Fluorocarbon Extraction

- State of the Art
 - Pilot Plant
- Decontamination Factor
 - 1.0×10^3 to 1.0×10^4
- Waste Form
 - Gas
- Advantages
 - High DF
 - Compatible with Iodine Recovery
 - Compatible with Carbon Recovery

TABLE 7 (Continued)

- Disadvantages
 - High Energy System
 - Requires Tall Column
 - Fluorocarbon Release
 - Underdeveloped Xenon Removal Step

3. Solid Adsorbent

- State of the Art
 - Laboratory/Semi-Pilot
- Decontamination Factor
 - 500 - 1000
- Waste Form
 - Cold Liquid
- Advantages
 - Low Energy System (except for the final step which is common to all Noble Gas recovery and separation systems)
 - Familiar Technology except for AgZ
 - Good Xenon Removal ($DF \approx 10000$)
- Disadvantages
 - New Technology (AgZ)
 - High Purity Feed Required
 - Requires Cryogenic Concentration for the Final Step
 - The System requires Complex Valving and Maintenance
 - Cost of Materials

TABLE 8**Krypton Collection**

<u>Criteria</u>	<u>Cryogenic</u>	<u>AgZ</u>	<u>FCRS</u>
Simplicity	5.0	5.0	6.0
Reliability	10.5	10.5	12.0
Safety	8.0	14.0	12.0
Cost	13.5	10.5	10.5
State of Art	24.0	9.0	21.0
Material Handling	6.0	7.5	7.5
Decontamination Factor	<u>18.0</u>	<u>21.0</u>	<u>27.0</u>
Total	85.0	77.5	96.0
Percent of Maximum	63.0	57.4	71.1

4.2.2 Krypton Fixation

For the chemically inert noble gases, the definition of fixation must be broadened to include physical confinement as well as chemical reaction. Thus the options for krypton-85 include cylinder storage of the pure gas with or without inert fillers, and physical entrapment in a metal or glass matrix. Storage forms evaluated in this study include:

- Storage of purified gas in pressurized cylinders.
- Co-storage in pressurized cylinders with zeolites.
- Zeolite encapsulation followed by cylinder storage.
- Ion implantation in an inert matrix plus packaging and storage.
- Encapsulation in glass followed by packaging and storage.

4.2.2.1 Pressurized Cylinder Storage

The most straight forward method for krypton storage is in a pressurized gas cylinder. A standard 50-L cylinder (DOT Specification 3A-2015) filled to 34 atmospheres (500 psi) could contain about 128,000 Ci of krypton-85 (326 g of krypton-85) along with about 5 kg of stable (nonradioactive) krypton, assuming removal of xenon is complete.⁹ The isotopic ratio in the total krypton mixture will depend on the fissile fuel type, the irradiation history and the decay time. The daughter product from the decay of krypton-85 is stable rubidium-85 so that up to 326 g of highly reactive rubidium metal will eventually accumulate in the cylinder.

Screening tests have been conducted in which candidate cylinder construction materials were exposed to rubidium metal and rubidium hydroxide at elevated temperatures.¹⁰ These tests showed that alloys are available which are potentially resistant to rubidium attack. Extensive additional testing will be necessary, however, to prove the efficacy of cylinder materials over the anticipated 50- to 100-year storage period.

4.2.2.2 Co-storage with Zeolites

One method of minimizing the effect of rubidium buildup in the storage cylinders would be to incorporate a 'getter' in the container to absorb the corrosive material. Zeolites would serve this function well since the rubidium would be incorporated into the crystal structure of the minerals rather than attack the metal cylinder. Some of the krypton may slowly diffuse into the zeolite crystal structure, also, thus reducing pressure in the cylinder slightly. At the temperatures and pressures anticipated in a krypton storage facility, this 'encapsulation' phenomenon will be minimal, however.

The principal disadvantage to co-storage with zeolites is the potential fouling of cylinder valves by fines when the krypton is recovered for fixation. In addition, the heat transfer through the zeolite-filled cylinder is somewhat poorer than that through the pure gas.

4.2.2.3 Zeolite Encapsulation

A third storage method utilizes the encapsulation phenomenon in which the krypton atom is selectively diffused into the zeolite cage structure. In this technique, activated sodalite is exposed to high pressure, high temperature krypton (or a mixture of krypton and xenon). The molecular diameter of the krypton is such that it will diffuse into the zeolite cage and become trapped. INEL tests show that the leakage rate back out of the crystal structure should be less than 0.1% in ten years.⁹

Because of the preferential encapsulation of the krypton into the zeolite structure, a mixture of krypton and xenon can be used as a process feed gas. INEL reports that up to 50% Xe (v/v) can be present in the gas mixture without an appreciable reduction in encapsulated krypton. The lower purity feed gas requirements should simplify the noble gas separation step in the off-gas cleanup system.

Conceptual design of a zeolite encapsulation facility calls for a fairly high inventory of krypton-85 (up to 1.4 MCi) in the pressure vessel during the encapsulation process. The process itself requires pressures of 1200 to 2000 atmospheres and temperatures of 400 to 575°C. Thus, stringent design requirements are necessary to preclude large releases of radioactivity in the event of an accident.

4.2.2.4 Ion Implantation

Krypton can be ionized in an electric field and be incorporated into an inert metal matrix using ion sputtering techniques. The resulting gas-loaded metal can then be further encapsulated to minimize the small amount of krypton-85 which could be released by diffusion out of the metal.

The process is carried out under vacuum and with a low volume feed stream. Thus any process line breaks should result in leakage rather than product loss. The low volume flow would also minimize product losses in the event of piping ruptures. Specific loadings of 9 to 10 atom % have been achieved in laboratory experiments. Reported krypton release rates from the matrix are as low as 0.07% in 100 years at a temperature of 200°C and 12% in 100 years at 300°C.¹¹⁻¹⁴

The ion sputtering technique is a high energy-consumption process, however, both because of the required vacuum system and the electrical energy required to sputter the host metal matrix. In addition, the process requires a high purity krypton feed stream. Since xenon has a lower ionization potential than krypton, any xenon present will be preferentially deposited in the matrix. Pacific Northwest Laboratory (PNL) experiments with the ion sputtering technique indicate that alloys of yttrium or zirconium may be required. Two alloys showing the most promise are:

1. 85% iron, 12% yttrium, 2% minor components.
2. 75% zirconium, 25% iron.

To date, the metal alloy ion sputtering process for krypton encapsulation is only a laboratory-scale process. Glass matrix ion sputtering techniques are also under investigation at PNL.

4.2.2.5 Glass Encapsulation

Laboratory experiments at PNL have also shown that krypton can be incorporated into glass, either as a solution or into a heat treated sintered material.¹¹⁻¹⁴ PNL has reported krypton gas loadings of 6 cc/g of SiO₂ (STP) when loaded at 5000 psi and

calculated loadings of 25 cc/g when loaded at 25,000 psi.^{15,16} The sintered glass technique requires both high temperature and high pressure. Thus this method, like the zeolite encapsulation technique, involves risk of a high potential release in the event of an accident. The glass encapsulation technique is still in the laboratory stage of development.

4.2.2.6 Comparison of Alternatives for Krypton-85 Fixation

A comparative summary of the krypton fixation processes under consideration, with advantages and disadvantages is shown in Table 9. The selection criteria methodology was applied to the krypton fixation processes under study. The results are shown in Table 10.

TABLE 9

Comparison of Krypton Fixation Alternatives

Fixation Alternative

1. Bottled Gas

- State of the Art
 - Plant
- Advantages
 - Known Technology
 - Simple
- Disadvantages
 - Rb Corrosion
 - Moderately High Pressure
 - Acceptance Questionable

2. Bottled with Zeolite

- State of the Art
 - Laboratory
- Advantages
 - Known Technology
 - Simple
 - Reduced Release if Bottle Ruptured
 - Rb Trapping and Less Corrosion

TABLE 9 (Continued)

- Disadvantages
 - Undeveloped Technology
 - Large Volume of Waste to Handle

3. Ion Implantation

- State of the Art
 - Laboratory
- Advantages
 - Low Volume
 - Immobilized Form
 - Low Pressure and Temperature
 - No Rb Interference
- Disadvantages
 - Undeveloped Technology
 - High Cost
 - Xenon Interference
 - Vacuum Required

4. Zeolite Encapsulation

- State of the Art
 - Laboratory
- Advantages
 - Immobilized Form
 - Preferential Kr Encapsulation
(Complete Xe removal not required)
- Disadvantages
 - High Pressure
 - High Temperature
 - Potential for Large Release in an Accident

TABLE 9 (Continued)

5. Glass Solution

- State of the Art
 - Laboratory
- Advantages
 - Immobilized Form
- Disadvantages
 - High Pressure
 - High Temperature
 - Potential for Large Release in an Accident

TABLE 10

Krypton Fixation

<u>Criteria</u>	<u>Bottled</u>			<u>Ion Implant</u>	<u>Glass Solution</u>
	<u>Gas</u>	<u>Zeolite</u>	<u>Encap.</u>		
Simplicity	9.0	8.0	6.0	5.0	6.0
Reliability	12.0	12.0	10.5	10.5	10.5
Safety	12.0	12.0	10.0	16.0	10.0
Cost	13.5	10.5	9.0	7.5	9.0
State of the Art	30.0	18.0	15.0	12.0	12.0
Material Handling	13.5	13.5	12.0	12.0	12.0
Total	90.0	74.0	62.5	63.0	59.5
Percent of Maximum	85.7	70.5	59.5	60.0	56.7

The reference storage form for krypton-85 in this study is bottled gas in pressurized cylinders housed in an interim storage facility. Because of the high cost of permanent storage in an above-ground engineered facility, storage of krypton-85 in such a facility is only an interim storage scheme. Dry-well storage of the pressurized cylinders, either onsite or in a central repository location, is a viable alternative for long-term storage. If a fixed waste form is required by regulatory agencies, ion implantation of krypton in a metal matrix appears, at this time, to be the safest encapsulation technique to produce a solid krypton-85 waste form. The ion implant technique does not require the high temperatures and pressures necessary for zeolite encapsulation or glass encapsulation. Cylinder storage was chosen for this study because the ion implantation technique is not fully developed.

4.2.3 Krypton References

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4.3 Carbon-14

4.3.1 Carbon-14 Collection

Four generic methods are currently being considered for the collection of carbon-14 (as CO_2) from process air streams. These include:

1. Chemical reaction in a liquid scrubber system.
2. Chemical reaction in a granular solid bed.
3. Physical concentration in a liquid solvent system.
4. Physical concentration in a granular solid bed.

4.3.1.1 Liquid Scrubbing

Liquid chemical scrubbing [with NaOH , or a mixture of NaOH and Ca(OH)_2] is a commonly used industrial process for CO_2 control. The method requires minimum pretreatment of the gas stream being scrubbed (acid and nitrogen oxide removal would be necessary to prevent both acidification of the scrubber solution and the accumulation of nitrogen salts in the scrubber liquid). Chemicals required are relatively inexpensive.

Liquid chemical scrubbing with $\text{Ba}(\text{OH})_2$ has also been investigated at Oak Ridge National Laboratory (ORNL).¹ ORNL has abandoned the liquid system in favor of a solid bed sorber system.

Disadvantages of the liquid scrubber process include:

1. Relatively low DF.
2. Additional process equipment requirements for CaCO_3 recovery.
3. Generation of a large volume of waste.
4. Tritium (if the voloxidation process is used) will be collected in the scrubber and increase the HTO waste volume.
5. Fairly large space requirements to house both the scrubber tower and recovery equipment.

4.3.1.2 Solid Bed Scrubbing

A solid phase, chemically active system would require less process equipment space, both because a solid, granular bed is smaller than a scrubber tower, and (with the proper choice of adsorbent) because no additional equipment is necessary to recover the carbonate salt end product. The reaction products in the solid bed system can be limited to BaCO_3 and water. Thus no chemical wastes are generated which require extensive later treatment.

The cost of raw materials will depend on the solid absorbent chosen for the system. Candidate materials include Baralime*, Ascarite*, LiOH or LiOH-impregnated granules, and $\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$. Vendors' quoted capacities for CO_2 for representative sorbent materials are shown in Table 11.

* Trademark of A. H. Thomas Co.

TABLE 11

Carbon Dioxide Sorption Capacities

<u>Material</u>	<u>CO_2 Capacity, by weight</u>
Baralime	5%*
Ascarite	20%*
LiOH-C	12%**
$\text{Ba}(\text{OH})_2 \cdot 8\text{H}_2\text{O}$	14%†

* Data from A. H. Thomas Co., Philadelphia, PA

** LiOH impregnated carbon. Data from NUCON, Columbus, OH

† Calculated from formula weight

Both the Ascarite and LiOH materials produce soluble carbonate salts as end products, and are thus less than ideal sorbers. Additional processing would be required with either to convert the carbon-14 to an insoluble salt for storage. In addition, both materials are hygroscopic. Ascarite fuses into a solid when exposed to moisture. LiOH-impregnated carbon will wet to the extent that increased pressure drops will be encountered.

Barium hydroxide octahydrate will react directly with CO_2 to form BaCO_3 and H_2O , thus meeting the desired goal of minimum waste generation. Other hydrated barium hydroxide salts have been investigated, but they tend to dust on reaction with CO_2 , thus leading to pluggage problems. The octahydrate salt has the disadvantage of requiring a humid air stream to prevent crystal dehydration.^{1,2}

If tritium removal is included in the total off-gas treatment package, the HTO must be removed prior to CO_2 collection. Otherwise, highly mobile tritiated water will accumulate in the carbonate salt bed, and further treatment will be necessary before the bed can safely be placed in a storage repository. Any attempt to remove moisture in the presence of CO_2 , however, will result in some carbon-14 contamination of the HTO. Thus, the disposal form for tritium may be dictated by the carbon-14 content.

4.3.1.3 Liquid Phase Absorption

Current investigations for the development of liquid phase physical concentration of CO_2 are limited to the ORNL fluoro-carbon recovery system (FCRS). This system is reported to be capable of accumulating iodine and carbon dioxide as well as xenon and krypton in a single process. DF's of up to 1000 are reported for CO_2 using R-12 as the fluorocarbon solvent.³

The disadvantages of the FCRS system (as compared to an in-line fixed bed system) include:

1. Fairly large space requirements to house the unit.
2. Increased energy requirements for solvent compression.
3. Greater risk for accidental activity release because of the use of a pressurized, low boiling solvent.
4. CO_2 must be fixed in an additional, separate step after being stripped from the solvent.

4.3.1.4 Solid Phase Adsorption

The fourth generic method, physical concentration of CO₂ on a solid sorbent, is a commonly used industrial process. Synthetic zeolites, such as types 13X or 5A molecular sieves, are used in parallel beds to accumulate the gas. When one bed approaches breakthrough, flow is diverted to the second bed and the first bed is regenerated. Regeneration is accomplished by purging the bed in the reverse direction with a heated, CO₂-free gas (air or nitrogen). The CO₂-rich purge gas (containing up to 1% by volume CO₂) is routed to a bed of chemically active material [Ba(OH)₂] for fixation. As noted earlier, the CO₂ stream must be rehumidified if octahydrobarium hydroxide is used.⁴

Moisture is preferentially removed by most of the zeolites, so dehumidification of the gas stream is a prerequisite before CO₂ removal on zeolite beds. Type 3A molecular sieves can be used to dehumidify the air stream prior to CO₂ removal. Some recycling will be necessary, however, to maintain a low CO₂ content in the moisture.

4.3.1.5 Comparison of Alternatives for Carbon-14 Collection

A comparative summary of the processes under consideration, with advantages and disadvantages, is shown in Table 12. The candidate processes were also analyzed using the evaluation criteria outlined earlier. The results of this analysis are summarized in Table 13. The analysis shows that the fixed-bed chemical sorber system is the better of the candidates for CO₂ collection.

TABLE 12

Comparison of Carbon Collection Alternatives

Collection Alternative

1. Chemical Reaction in a Liquid Scrubber System

a. Caustic

- State of the Art
 - Plant Scale
- Decontamination Factor
 - Less than 100
- Waste Form
 - CaCO₃ Slurry
- Advantages
 - Simple

TABLE 12 (Continued)

- Known Technology
 - inexpensive
 - Disadvantages
 - Relatively Low DF
 - Sensitive to Acidic Vapors
 - Recovery Process
 - Complex
 - Adds Waste Volume
 - Increases Waste Handling Hazard
 - Tritium if Present will Collect in Scrubber
 - Large Space Requirements
2. Chemical Reaction in a Granular Solid Bed
- a. Barium Hydroxide Octahydrate
- State of the Art
 - Pilot Scale
 - Decontamination Factor
 - Greater than 100
 - Waste Form
 - Granular Solid
 - Advantages
 - Disposal Waste Form
 - Concrete Compatible
 - Disadvantages
 - Must Remove HTO First (if present)
 - Must Rehumidify Gas Stream Before CO₂ Removal
 - Must Use the Octahydrate
- b. Other Candidate Materials Include
- Baralime
 - Ascarite
 - Lithium Hydroxide
 - Lithium Hydroxide Impregnated Granules

3. Physical Concentration in a Granular Solid Bed

a. Synthetic Zeolites (13X or 3A molecular sieves) (Followed by fixation on barium hydroxide)

- State of the Art
 - Plant Scale (collection)
 - Pilot Scale (fixation)
- Decontamination Factor
 - About 100
- Waste Form
 - Concentrated Gas (approx. 1% CO₂ in air)
 - Collected and Fixed on Bed of Barium Hydroxide
- Advantages (in conjunction with fixation on bed of barium hydroxide)
 - Disposal Waste Form
 - Concrete Compatible
- Disadvantages
 - Must Remove Water Before CO₂ Collection
 - Must Rehumidify Air Before Fixation Step
 - Must Use the Hexahydrate Barium Hydroxide

4. Physical Concentration in a Liquid Solvent System

a. Fluorocarbon

- State of the Art
 - Laboratory/Pilot Plant
- Decontamination Factor
 - Approximately 1.0×10^4
- Waste Form
 - Gas [to be fixed in Ba(OH)₂ bed]
- Advantages
 - Compatible with Krypton Recovery
 - Compatible with Iodine Recovery
 - High DF
- Disadvantages
 - Requires Separate Fixation Step
 - Requires Rehumidification Before Fixation

TABLE 13**Evaluation of Carbon Collection Methods**

Criteria	Scrubbing NaOH-Ca(OH) ₂	Ba(OH) ₂ Solid	Ba(OH) ₂ Zeolite	FCRS
Simplicity	6.0	7.0	6.0	5.0
Reliability	10.5	13.5	10.5	10.5
Safety	12.0	18.0	16.0	12.0
Cost	13.5	13.5	10.5	10.5
State of the Art	21.0	18.0	18.0	15.0
Material Handling	6.0	12.0	10.5	9.0
Decontamination Factor	<u>12.0</u>	<u>15.0</u>	<u>15.0</u>	<u>24.0</u>
Total	81.0	97.0	86.5	86.0
Percent of Maximum	60.0	71.8	64.1	63.7

4.3.2 Carbon-14 Fixation

For carbon-14, the principle fixation methods under investigation all rely on incorporation of a low solubility carbonate salt into a concrete matrix. Calcium carbonate from a caustic scrubber collection system could be mixed with sand and cement to form concrete. Barium carbonate from a fixed-bed collection system could similarly be mixed directly into the grout to form concrete. A second alternative for the barium hydroxide fixed bed system is to cast the spent collection bed directly inside a block of concrete, thus eliminating several potentially hazardous handling steps. This alternative would offer the additional advantage of providing a secondary barrier (the canister shell) to delay leaching of carbon-14 from the waste package.

4.3.2.1 Comparison of Alternatives for Carbon-14 Fixation

Advantages and disadvantages of the proposed fixation schemes are summarized in Table 14. The candidate fixation processes were also analyzed using the evaluation criteria given earlier. The results of this analysis are summarized in Table 15. The analysis shows that direct incorporation of a fixed bed into concrete is the preferred process.

TABLE 14

Comparison of Carbon Fixation Alternatives

Fixation Alternative

1. Caustic Scrubbing

a. Calcium or Barium Carbonate in Concrete

- State of the Art
 - Plant Scale
- Advantages
 - Known Technology
 - Relatively Insoluble
- Disadvantages
 - High Volume Waste

2. Barium Hydroxide Octahydrate

a. Canned Granular Solid Fixed in Concrete (or Buried as is)

- State of the Art
 - Laboratory/Pilot Scale
- Advantages
 - Relatively Insoluble
 - Minimal Waste Handling
- Disadvantages
 - Incomplete Development

3. Zeolite plus Barium Hydroxide

a. Waste Form

- Concentrated Gas (approx. 1% CO₂ in Air)
- Collected and Fixed on Bed of Barium Hydroxide

b. Canned Granular Solid Fixed in Concrete (or Buried as is)

(See No. 2 above)

TABLE 14 (Continued)

4. Fluorocarbon Extraction

a. Waste Form

- Concentrated Gas
- Collected and Fixed on Bed of Barium Hydroxide

b. Canned Granular Solid Fixed in Concrete (or Buried as is)

(See No. 2 above)

TABLE 15

Evaluation of Carbon Fixation Methods

<u>Criteria</u>	<u>CaCO₃ in Concrete</u>	<u>Ba(OH)₂ in Concrete</u>	<u>Ba(OH)₂ Fix Bed Concrete</u>
Simplicity	8.0	7.0	9.0
Reliability	9.0	10.5	13.5
Safety	14.0	12.0	16.0
Cost	10.5	10.5	13.5
State of the Art	21.0	21.0	27.0
Material Handling	<u>9.0</u>	<u>10.5</u>	<u>13.5</u>
Total	71.5	71.5	92.5
Percent of Maximum	68.1	68.1	88.1

4.3.3 Carbon-14 References

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5.0 TRANSPORTATION OF SOLIDIFIED WASTES

5.1 Background

This section describes alternatives for the transportation of solidified waste gases associated with the back end of the nuclear fuel cycle. The principal transport modes for radioactive fuel and waste materials are by truck and rail. In special cases, shipments by barge have been made.^{1,2}

All shipments of packaged radioactive waste gases are regulated by DOT and NRC. Standards and criteria for packaging and transportation are contained in Title 49 and Title 10 of the Code of Federal Regulations.^{3,4} Solidified radioactive waste gases are expected to be shipped in casks that resemble those currently available for shipment of spent fuel. However, casks for this purpose could be simpler in design because neutron shielding would not be required and heat removal requirements would be much reduced. Shipment to the long-term storage facility or federal repository could be by truck, rail, or barge.

Shipments of spent fuel have been made commercially on a routine basis for many years. Non-high-level transuranium wastes are presently shipped by rail from the Department of Energy (DOE) facilities to interim storage at DOE sites. Waste containers and overpacks for rail and truck shipments of TRU wastes are commercially available. Shipping containers are also available for those radioactive elements that require little or no shielding and have a relatively low heat generation rate.

There is presently no commercial processing of spent fuel in the United States, and commercial shipments of solidified high-level waste, fuel residues, and solidified radioactive waste gases have not been made. No technical reasons exist which would prevent the design and construction of shipping containers for all wastes from the fuel reprocessing plant.

Radioactive waste shipments are expected to be made by commercial carrier. The principal transport modes are truck and rail. Because of legal weight restrictions on the nation's highways, truck shipments are limited to payload weights of about 22 metric tons. Rail shipments can accommodate payloads of about 100 metric tons.

5.2 Classification

Each type of radioactive material is classified for transportation purposes into one of several groups according to its potential hazard if released to the environment. Transport Group I is the most restrictive. Plutonium and other transuranium elements are in this transport group. Transport Group VII is the least restrictive. Material such as tritium gas, with a low potential for producing radiation exposure, are in this transport group. Krypton-85 and iodine-129 are in Transport Group III, and carbon-14 is in Transport Group IV.

For radioactive material belonging to a specific transport group, the type of packaging required depends on both the specific activity and the total quantity of materials (curies) being shipped. Table 16 lists the curie limits associated for krypton-85, iodine-129, and carbon-14.

TABLE 16

Quantity Limits for Shipment of Radioactive Waste Gases

Element	Activity Level, Ci			Quantity
	Exempt	Type A	Type B	
Iodine-129	$<1.0 \times 10^{-3}$	$>1.0 \times 10^{-3}$ to 3	>3 to 200	>200
Krypton-85	$<1.0 \times 10^{-3}$	$>1.0 \times 10^{-3}$ to 3	>3 to 200	>200
Carbon-14	$<1.0 \times 10^{-3}$	$>1.0 \times 10^{-3}$ to 20	>20 to 200	>200

5.2.1 Exempt Material

Shipments which pose a negligible risk to public health may be classified as low-specific-activity (LSA) or exempt material [10 CFR 71.4 (g)]. Exemption is based on quantity and on several other conditions outlined in 49 CFR 173.391.

Exempt material shipped in exclusive use vehicles is exempt from most of the packaging requirements of Title 49 of the Code of Federal Regulations [49 CFR 173.392 (b)]. Basically only strong, tight packaging that will not leak in normal transport is required. The radiation dose rate limits of 49 CFR 173.393 are still applicable, however.

5.2.2 Type A Material

If the specific activity of a shipment exceeds LSA limits, but the total number of curies is within Type A limits, then Type A packaging is required for the transport of the material. In accordance with DOT specification 7A (49 CFR 178.350), a Type A container must meet the standard construction requirements for packages in 49 CFR 173.24 and 49 CFR 173.393. The packaging must also prevent loss or dispersal of its radioactive contents and retain its radioactive shielding efficiency if subjected to a series of tests designed to simulate package stresses associated with normal transport. The Type A packaging test series is outlined in 49 CFR 173.398 (b) and includes impact, puncture, and compression tests, vibration tests, water spray tests, pressure tests, and thermal tests. Typical Type A packaging includes steel drums (for example, the specification 17C 55-gallon drum), wooden boxes, and steel boxes.⁵

5.2.3 Type B Material

Type B quantities of radioactive material must be shipped in Type B packaging. Type B packaging includes specification 6M containers,* packaging authorized by NRC, and packaging meeting the 1973 International Atomic Energy Agency (IAEA) requirements. In addition to meeting the standards for a Type A package, a Type B package must be able to survive a series of hypothetical accident test conditions with essentially no loss of containment and limited loss of shielding capability (10 CFR 71.36). The test sequence for Type B packages is designed to simulate the damage that might be expected in a severe accident situation. The damage test sequence includes:

- A free fall from a height of 9.2 m (30 feet) onto a flat, essentially unyielding surface, with the package striking in the orientation that does the most damage.
- A free fall from a height of 1 m (40 inches) onto a 15.2 cm (6 inches) diameter steel plunger that is long enough, and with the package in the correct orientation, to do maximum damage.
- Heat input from exposure for 30 minutes to a fire or other radiant environment having a temperature of 802°C (1475°F) and an emissivity of 0.9.

* The specification 6M container is described in 49 CFR 178.104. It consists of an inner container fixed within an outer shell by a prescribed solid centering medium.

- For fissile material package only, immersion in water at a depth of 91 cm (3 feet) for 8 hours.

5.2.4 Large Quantity Material

Large quantities of radioactive material are required to be transported in Type B packaging (10 CFR 71.32). Waste shipments in the nuclear fuel cycle will normally be made in exclusive-use vehicles. DOT regulations (49 CFR 173.393) set the following limits on radiation dose rates associated with exclusive-use vehicle shipments:

- 1000 mrem/hr at 0.91 m (3 feet) from the external surface of the package (provided the package is transported in a closed vehicle).
- 200 mrem/hr at the external surface of the vehicle.
- 10 mrem/hr at any point 2 m (6 feet) from the vehicle.
- 2 mrem/hr at any normally occupied position in the vehicle.

DOT regulations also require the following limits:

- The absence of significant removable surface radioactive contamination on the external accessible surfaces of packages when they are shipped [49 CFR 173.393 (e)].
- The temperature at any accessible surface of a shipping cask to not more than 50°C (122°F) at any time during transport [exclusive-use shipments, however, may have temperatures as high as 82°C (180°F)] [49 CFR 173.393 (e)].
- Prohibit the loading, transportation, and storage of radioactive materials together with Class A explosives.*

5.3 Alternatives

Based on results from previous studies, the three viable alternatives for the transport of recommended solidified radioactive waste gases are truck, rail, and barge.⁶⁻⁹ The relative cost and risk of each mode of transport is given in Table 17. The cost and risk of truck shipment is greater than rail shipment. The cost of barge shipment is less than rail or truck if the fuel reprocessing plant and the storage facility are ideally located on a connecting waterway.

* Class A explosives include black powder, propellant explosives, initiating or priming explosives, blasting caps, certain types of explosive projectiles, and detonating fuses.

TABLE 17

Comparative Risks and Costs of Transportation

Mode	Casks	Risks*	Costs
Rail	None Approved	<ul style="list-style-type: none"> • Impact > Fire > Dissolution • Lower than Truck 	Truck = Rail > Barge
Truck	None Approved	<ul style="list-style-type: none"> • Impact > Fire > Dissolution • Greater than Rail 	Truck = Rail > Barge
Barge	None Approved	<ul style="list-style-type: none"> • No Data Available 	Truck = Rail > Barge

* Risk based on the shipment of high-level waste.
Ranking depends on specific isotope shipped.

Several factors influence the choice of rail or truck for the shipment of solidified radioactive waste gases. Rail casks have a significantly larger payload capacity than truck casks. On the other hand, shorter truck travel times tend to compensate for the payload to gross weight advantage of rail shipments. The time required to load or unload a rail cask is about twice as long as that required to load or unload a truck cask. However, considering that a rail cask can transport up to ten times as much spent fuel as a truck cask, the loading or unloading time per ton of material shipped is lowest for rail shipment. In summarizing reported experiences to date, it is noted that there has never been a significant case of a transportation accident involving the shipment of radioactive materials that represented a danger to the public. There have been cases of leaks and contamination and improperly shipped sources. However, there have been no injuries or deaths of a radiological nature to the transportation of nuclear materials.

5.4 Transportation References

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2. Technology for Commercial Radioactive Waste Management, Volume 1, USDOE Report DOE/ET-0028, U. S. Department of Energy, Washington, DC (May 1979).

3. Title 49, Code of Federal Regulations, Parts 170-189.
4. Title 10, Code of Federal Regulations, Part 71 (10 CFR 71).
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7. Shipment of Nuclear Fuel and Waste - Are They Really Safe?, USDOE Report DOE/EV-0004, U. S. Department of Energy, Washington, DC (October 1977).
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9. Final Environmental Statement on the Transportation of Radioactive Materials by Air and Other Modes, Volume 1, NRC Report NUREG-0170, U. S. Nuclear Regulatory Commission, Washington, DC (December 1977).

6.0 STORAGE - DISPOSAL

6.1 Concepts

A large number of alternative storage/disposal media have been suggested for the disposal of high-level wastes and spent fuels. No in-depth studies were found in the literature for the storage of radioactive gases. However, since (1) the radioactive gas waste forms would be considered high-level waste, and (2) all of the recommended radioactive gas waste forms are solids (except possibly for krypton-85 gas), any high-level waste storage facility would have the potential for storing radioactive gas waste forms. Of the many media suggested, the following have received the most attention and are considered to be the most likely candidates for disposal either in the near future or for eventual storage. A more detailed description of most of these concepts can be found in Reference 1.

6.1.1 Engineered Storage Facility

Since it appears that no geologic storage facility will be available in the near future, possibly not this century, above-ground storage in an engineered facility is the most likely temporary expedient for storage of radioactive gas waste forms. Spent fuels are already being stored in reactor basins, and additional storage in away-from-reactor storage facilities is being studied. Calcined high-level waste is presently being stored above ground, and current plans are to store any vitrified waste in above-ground facilities for up to five years and possibly longer.

Economic penalties for long-term storage in large-scale engineered facilities make above-ground storage unattractive. As discussed in Section 6.3.2, an above-ground engineered storage facility is up to 100 times more costly than geologic storage for krypton-85. The certainty of eventual destruction of such a facility by natural phenomenon makes the above-ground repository unacceptable for permanent storage of iodine-129 and carbon-14.

One engineered storage option that is attractive for krypton-85 is the Spent Unreprocessed Fuel (SURF) dry-well, or subsurface shaft storage concept.² The SURF configuration consists of bottom sealed, steel liners inside concrete encased wells. Krypton-filled cylinders would be stacked inside the well in a vertical array. The number of cylinders per hole and the

hole spacing will vary somewhat with the heat capacity of the soil where the facility is located. Thick concrete plugs would be used to seal the holes. Each plug could be equipped with a sample line to monitor for leaking cylinders. Use of the dry-well concept eliminates the need for the refrigeration units necessary for heat removal in an above-ground concept, thus reducing the operating costs for storage. Long-term stability of cylinders in the dry-well configuration is open to question, however, because of potential moisture-induced corrosion after the heat load is reduced by decay. Thus, this concept is more viable as an interim storage option than a permanent storage facility for bottled gas. Fixed forms of krypton (ion sputtered or zeolite encapsulated) could be stored indefinitely in the dry wells. The fixed waste forms are not pressurized and are relatively insensitive to moisture if the package is breached.

6.1.2 Mined Repositories

The most likely near-term geologic repository will be a mined repository in either salt or basalt. Storage in shale and granite has also been investigated. These four media are considered to be representative of all geologic media.

The proposed Waste Isolation Pilot Plant (WIPP) in New Mexico would be a mined repository in a salt dome. The proposed WIPPB would utilize basalt as the media for a repository in the state of Washington. Considerable studies have been made on mined repositories and several risk assessments are in progress.

The technology is available, but further research is required to resolve some deficiencies in the data base before repository performance can be confidently predicted.

6.1.3 Deep Ocean Sediment

Analysis of ocean sediment has shown that abyssal hill regions are the most appropriate areas for ocean sediment disposal. These abyssal hill regions are large, are remote from human activities, have few natural resources, are biologically unproductive, and have weak and variable bottom currents. These sediments are soft and pliable near the ocean floor and become increasingly rigid and impermeable with depth. Tests have shown that the sediment has a high sorption coefficient and low pore-water movement. Core analysis has shown them to be stable for millions of years. One emplacement method is to encase the waste in a bullet-shaped cask which would be propelled or dropped from a ship to penetrate into the sediment.^{1,3}

There are no known technical reasons why ocean sediment disposal would not be satisfactory, but much additional research is needed to assure safe permanent disposal. In addition, legal and political issues will need to be resolved.

6.1.4 Very Deep Holes

The placement of waste in very deep holes would delay the release and re-entry of radioactive material to the biosphere. The capability to excavate a narrow hole to depths of 35,000 feet and wide holes to 15,000 feet has been demonstrated. However, placing of waste in very deep holes may present some severe engineering problems. This concept cannot be fully evaluated until the characteristics of potential sites have been determined. Until then, how deep is deep enough cannot be defined.

6.1.5 Ice Sheet

Three modes of ice sheet disposal have been suggested, they are:

1. Retrieval surface storage.
2. Suspension in the ice sheet using anchored cables.
3. Deep emplacement permitting the decay heat to allow the waste to sink to the bottom of the ice sheet.

Disposal in the antarctic or Greenland ice sheets would offer the advantages of remoteness and potential isolation from the biosphere. The disadvantages are the long transport distances, the high cost and difficulties of operating in the polar areas, and the uncertainties in the long-range interactions between the ice sheets and the waste. Furthermore, ice sheet disposal would require new international initiatives, an amendment of the Antarctic Treaty, or a new treaty with Denmark.

6.1.6 Transmutation

Transmutation is the conversion of waste to nuclides with more desirable properties by means of irradiation. For example, iodine-129 might be converted to short-lived iodine-130 (12.4 h half-life). While the advantages are speculative, the disadvantages are much more certain. For example, increased costs, additional processing, and increased risk of release to the environment will result. Transmutation will require advanced technology and is not considered a near-term option.

6.1.7 Extraterrestrial Space

The attraction of space disposal is the prospect of permanent removal of the waste from the human environment. The currently favored concept is injection into a circular solar orbit halfway between Earth and Venus. The waste could be placed into earth's orbit by the space shuttle and then launched into solar orbit. Because of the cost of space flights, only the long-lived isotopes such as iodine-129 should be considered. However, because of the uncertainties of launch reliability and high costs, space disposal is a questionable option. The reduction in long-term risk may be more than offset by the increased short-term risk for the space disposal option.

6.2 Risks

There are three government agencies actively preparing risk assessments for the storage of high-level waste in a mined salt repository:

1. NRC - Sandia and Lawrence Livermore Laboratories are developing methodology for the preparation of a risk assessment of geologic storage.⁴
2. EPA - Arthur D. Little Company is preparing a risk assessment to determine whether a risk-based standard can be established. Risk will be expressed in curies released rather than dose-to-man.⁵
3. DOE - The Pacific Northwest Laboratory is preparing a risk assessment for the Office of Nuclear Waste Isolation in an effort to minimize the risk of operating a waste repository.⁶

In addition, Los Alamos Technical Associates is preparing a risk assessment for the storage of high-level waste in basalt. None of these risk assessments, nor any previously completed safety analysis has specifically addressed the storage of radioactive gas waste forms.

For comparative purposes, however, the DOE Environmental Impact Statement¹ gives a general assessment of risk. Assessment of their data results in the ranking shown in Table 18. A rating of 1 corresponds to a low risk and a rating of 10 to a high risk.

Most risk assessments do not estimate the risk of storage past 500,000 to 1,000,000 years; iodine-129, however, has a half-life of 16,000,000 years. Present methodology is inadequate to forecast the probability of natural events or human activity in terms of tens of millions of years.

TABLE 18**Risk of High-Level Waste Storage Modes**

<u>Mode</u>	<u>Relative Risk</u>
Mined Repositories	
Salt	1
Shale	1
Granite	2
Basalt	2
Very Deep Holes	2
Deep Ocean Sediment	3
Ice Sheet	3
Transmutation	3
Extraterrestrial Space	10

6.3 Costs

The cost of constructing and operating various repositories can only be estimated since many of the storage/disposal options have not been sufficiently defined, and all of the operating problems and construction criteria have not been determined. The literature survey revealed only three documents which attempted to compare the costs of several options. BNWL-1900⁷ and DOE/EIS-0046-D¹ compared the cost of storing high-level waste, and estimated costs of storing various radioactive gas waste forms are given in an NRC Cost Assessment.⁸

6.3.1 High-Level Waste

BNWL-1900 developed costs based on a reference design. Costs were compared as levelized unit costs. The levelized unit cost is the single charge that could be assessed over the entire life of a facility that would recover all operating expenses, capital expenditures, and interest charges per unit processed.

DOE/EIS-0046-D used a panel of experts to estimate the expected increase in electrical power generation costs for construction and operation of a disposal facility. The disposal modes were then rated; a score of 5 represented costs of less than 0.5 mills/KWhr and a 1 was unlikely to exceed 2.5 mills/KWhr.

The costs from these two documents are shown in Table 19. While the difference in costs from 1973 to 1979 dollars varies by a factor of 10 to 18, the relative ranking remains unchanged. Both reports concluded that the disposal costs are not a large factor in the overall power cost.

TABLE 19

Costs of Storing High-Level Waste

Mode	Cost, Mills/KWhr	
	BNWL-1900	DOE/EIS-0046-D
Mined Repositories		
Salt	—	0.24
Shale	—	0.31
Granite	0.024-0.064	0.33
Basalt	—	0.40
Very Deep Holes	—	1.0
Deep Ocean Sediment	0.052-0.063	<1.5
Ice Sheet	0.007-0.090	<1.5
Transmutation	0.15	<2.0
Extraterrestrial Space	0.15-0.34	<2.5

6.3.2 Radioactive Gas Waste

The NRC cost assessment⁸ determined the volume and/or weight of various waste forms for iodine-129, carbon-14, and krypton-85. The cost of storage in various media per unit volume or weight (as determined by others) was applied to estimate the cost of storage per unit of power generated. A summary of these storage costs is shown in Table 20. Except for engineered storage of krypton-85, the storage costs were minor when compared with the cost of collecting and fixing the gaseous wastes.

TABLE 20

Radioactive Gas-Waste Storage Costs⁸

<u>Media</u>	<u>Cost, Mills/KWhr</u>		
	<u>Iodine-129</u>	<u>Carbon-14</u>	<u>Krypton-85</u>
Geologic Repository	0.067	0.0027	4.13
Seabed Disposal	0.30	0.0030	0.975
Sub-surface Shafts	0.97	0.20	1.87
Engineered Storage	—	—	166

6.4 Comparison of Alternatives

Only the four near-term alternatives were chosen for the comparative study. Engineered facilities were selected because radioactive gas waste forms are almost sure to be stored above ground until geologic repositories are available. Mined repositories are the most likely geologic repository, and deep ocean sediment and very deep holes are more near-term than the other alternatives.¹ Table 21 summarizes the advantages and disadvantages of each storage mode.

TABLE 21

Comparison of Storage Modes

1. Engineered Facility

a. Temporary Storage

- Advantages
 - Provides Monitorable, Retrievable Storage
- Disadvantages
 - Possible High Cost (lower for dry wells)
 - Subject to Natural Phenomena in Excess of Design Basis
- Adequacy
 - Iodine-129 (Good)
 - Carbon-14 (Good)
 - Krypton-85 (Good)

TABLE 21 (Continued)

b. Permanent Storage

- **Advantages**
 - Eliminates Cost and Risk of Transportation (if built at the FRP)
- **Disadvantages**
 - High Cost (lower for dry wells)
 - Finite Life of Facility too Short for Iodine and Carbon
 - Subject to Natural Phenomena in Excess of Design Basis
- **Adequacy**
 - Iodine-129 (Very Poor)
 - Carbon-14 (Very Poor)
 - Krypton-85 (Fair - good for dry wells)

2. Mined Repository

- **Advantages**
 - Retrievability Before Sealing
 - Low Cost
 - Technology Available
- **Disadvantages**
 - Not as Stable as Ocean Sediments
 - Subject to Eventual Breaching by Natural Phenomena
 - Political Resistance for Physical Location
- **Adequacy**
 - Iodine-129 (Fair)
 - Carbon-14 (Good)
 - Krypton-85 (Excellent)

TABLE 21 (Continued)

3. Deep Ocean Sediments

- **Advantages**
 - Low Cost
 - Isolated from Man
 - Very Stable Geologically
 - Low Porosity
 - Low Natural Resource Content
 - Low Biological Activity
 - Low and Variable Current Flow
 - Isotopic Dilution for Iodine-129
 - Natural Sink for CO₂ (carbon-14)
 - Temperature and Pressure Favorable for Krypton-85 Dissolution
- **Disadvantages**
 - Technology not Fully Developed
 - Irretrievable
 - Requires International Agreement
 - Greater Transportation Risks to Site
- **Adequacy**
 - Iodine-129 (Good)
 - Carbon-14 (Excellent)
 - Krypton-85 (Excellent)

4. Very Deep Holes

- **Advantages**
 - Isolated from Human Activity
- **Disadvantages**
 - Technology not Fully Developed
 - Irretrievable
- **Adequacy**
 - Iodine-129 (Fair)
 - Carbon-14 (Good)
 - Krypton-85 (Very Good)

6.4.1 Engineered Storage Facilities

The temporary use of above-ground engineered storage is almost a certainty if radioactive gases are collected during this century. An engineered facility can be constructed to resist design basis events and designed to contain escaping gases. The cost of such a facility, however, may be high.⁸ Iodine-129 may need to be stored above ground until processes are available for recovery of the silver adsorbent or until permanent disposal modes like deep ocean sediment are available.

Engineered storage facilities would be adequate permanent storage of krypton-85 since the krypton will have sufficiently decayed in about 100 years. However, it is unrealistic to consider an engineered facility for permanent storage of carbon-14 and iodine-129 because of their long half-lives. Even without human intrusion, an engineered facility would almost certainly be subject to natural phenomena in excess of the design basis event over a period of fifty thousand to several million years. Storage of krypton-85 in an engineered storage facility would eliminate the cost and risk of transportation to a disposal site. Increased storage costs in such a facility would, however, offset any small gain in transportation costs.

Interim storage of pressurized cylinders of krypton-85 would be less expensive in subsurface shafts (dry wells) than in a refrigerated vault type engineered facility. Long-term storage of pressurized cylinders in dry wells is less desirable, however. Corrosion of the cylinders and subsequent release of the contents of one or more cylinders could lead to local doses near the storage field in excess of the EPA guide. Krypton-85 in the fixed form (ion sputtered or zeolite encapsulated) could be stored safely in a dry well since package corrosion would not lead to excessive gas releases.

6.4.2 Mined Repositories

Repositories mined from salt, shale, granite, or basalt offer the advantages of low cost, retrievability, and available technology. While mined repositories are judged to be excellent for krypton-85 and adequate for carbon-14, they are probably insufficiently stable for storage of the long-lived iodine-129. If silver iodides were stored temporarily, retrievability would be advantageous so that the silver could be recovered in the future. On the other hand, retrievability could be a disadvantage because of possible unauthorized human intrusion to obtain the silver.

6.4.3 Deep Ocean Sediment

Deep ocean sediments offer the following advantages for disposal: are isolated from human activity, contain few natural resources, are biologically unproductive, have low pore-water movement, and have been shown to be stable for millions of years. On the other hand, burial in deep ocean sediment is irretrievable, requires much additional research to ensure that the nuclides would not re-enter the biosphere, and would require international agreements.

Deep ocean sediment is particularly attractive for iodine-129, carbon-14, and krypton-85. Even if the iodine casks were breached and a fraction of the iodine reached the ocean, the isotopic dilution with stable iodine would significantly reduce the effect of the release. Similarly, carbon-14 would be isotopically diluted with stable carbon. There is a considerable quantity of carbon in the ocean as carbonates and carbon dioxide. Environmental studies⁹ have shown that the carbon dioxide concentrations in the ocean and atmosphere tend to reach an equilibrium, and that about half of the carbon dioxide released from the burning of fossil fuels eventually finds its way to the ocean. With no carbon-14 recovery from the nuclear fuel cycle, the atmosphere retains only half of the carbon-14 released. The remaining half (along with half of the natural and bomb-produced carbon-14) is absorbed in the oceans at present. Burial of krypton-85 at great depths in the ocean would result in any krypton leakage being absorbed in the ocean due to the high pressure and low temperature. Because of the low current flow in the abyssal hill areas, most of the krypton would decay before reaching the surface. Should some of the krypton reach the surface it would do minimal harm because (1) abyssal hills are remote from man, (2) krypton is not ingested biologically by man, and (3) due to its short half-life, krypton-85 will not build up in concentration over a period of hundreds of years.

6.4.4 Very Deep Holes

Placement of waste in very deep holes would isolate these nuclides thousands of feet below depths normally excavated by man. The movement of nuclides from this depth to the surface would be exceedingly slow and, except for iodine-129, would probably permit significant decay of the nuclides. It would take an unusual tectonic event to resurface the nuclides in any significant concentration.

6.5 Conclusions

6.5.1 Iodine-129

As indicated in earlier sections of this report, special emphasis must be placed on the evaluation of disposal options for iodine-129. Technology is available to collect iodine-129 within the limits imposed by EPA regulations. The collected iodine can be isolated from man for several thousands of years and possibly several tens of thousands of years. However the very long half-life of the isotope (16 million years), the high mobility of the iodide ion, and the lack of packaging media resistant to corrosion and erosion for millions of years make it very unlikely that isolation from the general environment can be assured long enough to preclude eventual uptake by man. The potential dose for future man can be kept to very low levels, providing the iodine-129 can be diluted with sufficient stable iodine (iodine-127) to maintain a low isotopic ratio (the ratio of iodine-129 to total iodine) in any materials ingested by man.

Russel and Hahn¹⁰ have calculated that an equilibrium isotopic ratio of 0.0083 in the thyroid will result in an annual thyroid dose of 500 mrem. To maintain the dose to an individual below 75 mrem/yr (EPA regulation), the isotopic ratio in the thyroid must be less than 0.0013. If the isotopic ratio in any exposure medium is lower than this value, the thyroid dose will also be within the limit. Thus about 800 grams of iodine-127 must eventually be mixed with each gram of iodine-129 to be within the dose requirements for all future generations.

Premixing iodine-129 with iodine-127 to the proper ratio before disposal is an unattractive option. This alternative will significantly increase both the waste volume and the cost of treatment. A second alternative would be disposal in an iodine-rich mineral deposit to obtain eventual isotopic dilution. This option is also unattractive since such deposits are rare and are one of the primary sources of iodine for industrial and domestic use. Iodine-rich brine wells (such as those in California and Michigan) are similarly unattractive because of their use as sources of iodine salts for man.

Massive continental salt deposits may contain enough total iodine to provide potentially favorable isotopic ratios for iodine-129 buried within the deposit. The low specific iodine concentration (<0.05 ppm) and low iodine mobility within the solid bed may dictate careful spacing of disposal packages within the deposit, however, to assure adequate dilution should the salt deposit be breached by a flowing stream of water. Similar arguments apply to rock and shale disposal sites for iodine-129.

Dynamic flow of water through deep ocean sediment deposits (particularly those in the abyssal hill regions) is much less likely to occur than through continental rock or salt deposits. Thus movement of iodine-129 out of canisters buried in such deposits, and subsequent movement through the deposit would be limited to molecular diffusion. The bottom currents in such areas are weak and variable, biological activity is low, and the areas are remote from man's activities. In the absence of "streaming effects," the dilution volumes available to these sites can be measured in cubic kilometers. Thus, isotopic dilution is more likely for the ocean sediment disposal option than for the alternative disposal media.

Additional research is necessary to confirm adequate mixing for iodine-129 buried in these deep ocean sediments. In addition, international agreements for the sea disposal option are far from certain. Until such research is completed and international agreements worked out, fixed beds of iodine-129 can be placed in retrievable storage repositories to maintain isolation from man.

6.5.2 Krypton-85

Krypton-85 in the fixed form can be stored in dry wells, mined repositories, or ocean sediments with nearly equal safety. A dry well field above a mined high-level waste repository would probably be the least costly compromise since the land area above a mined repository would require surveillance for a long enough period of time to allow appreciable decay of the krypton. Thus both the near-surface soil and the deep rock or salt deposit would be utilized in the same purchased land package. Interim storage of pressurized cylinders before the fixation step in a dry well field would be less costly than construction and operation of a refrigerated storage vault.

6.5.3 Carbon-14

Mined repositories or ocean sediments are both acceptable alternatives for carbon-14 disposal. The large mass of nonradioactive carbon available in the ocean for isotopic dilution coupled with the uncertainty of stability of land based repositories over

a time span of 50,000 years (about ten half lives for carbon-14) tend to make the seabed option slightly more attractive. The necessity for collection and fixation of carbon-14 from U. S. reprocessing facilities is questionable, however, in the light of the minor impact such efforts will have on global dose from this isotope, and the added cost of such collection and disposal.

6.6 Storage References

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APPENDIX A. REFERENCE OFF-GAS TREATMENT SYSTEM

A.1 General Description

Based on an evaluation of the various alternatives for collection, fixation, transportation, and storage/disposal of the long-lived gaseous radionuclides, a reference system for off-gas treatment in a fuel reprocessing plant was selected for the detailed evaluation and assessment planned for Phase II. This section describes the reference processes selected. Detailed discussion of the selection process used in developing the reference system and the individual alternatives for each phase of the isolation process are presented in the following sections.

A fuel reprocessing facility is designed to process spent fuels from BWRs or PWRs. The spent fuel can be either UO_2 fuel or mixed oxide (MOX) fuel containing UO_2 and PuO_2 . A modified Purex (plutonium-uranium recovery by extraction) solvent extraction process is used to separate and isolate three streams: (1) plutonium alone or mixed with uranium, (2) uranium only, and (3) fission products. The combined plutonium and uranium are converted to a mixed oxide, blended with natural UO_2 , and fabricated into MOX fuel assemblies. The separated uranium is converted to UF_6 for shipment to offsite enrichment facilities. The high-level liquid wastes from processing are solidified into a leach-resistant borosilicate glass suitable for long-term storage in a Federal waste repository. Low-level wastes are solidified in concrete and transferred to Federal facilities. No liquid process effluents are released to the environment. Any process water is purified to, or below, the permissible regulatory limits and discharged to the atmosphere as water vapor. These facilities are designed to meet all existing and proposed Federal and State regulations and licensing criteria of the NRC.

Off-gases containing high concentrations of particulate matter, ruthenium, and oxides of nitrogen pass through filtration and scrubbing systems. Off-gases in vessel vents within the main processing building are collected, filtered, and treated for removal of iodine and organics as required before final filtration (sand or HEPA filters), and released to the environment from an exhaust stack.

Process gases from the shear, voloxidation, and dissolving steps, after some preliminary treatment, are sent to an off-gas treatment facility where gaseous and volatile nuclides are isolated. The design goal of the treatment facility is to limit releases of radioactive waste gases to values below all present and proposed allowable limits. A block diagram showing the origins of the radioactive waste gases is shown in Figure A-1.

The Off-gas Treatment Facility (Figure A-2) separates radioactive fission products (tritium, carbon-14, iodine-129, krypton-85, and residual ruthenium-103,106) from the voloxidation, shear purge, failed fuel vent, and dissolver off-gas streams and stores the fission products in appropriate forms to prevent their release to the atmosphere. Nitrogen oxide gases are also recovered from the dissolver off-gas and converted into nitric acid for recycle. The off-gas stream from the head-end operations (shearing and voloxidation) is called the HOG system. The off-gas stream from the dissolver is called the DOG system. The off-gas treatment system handles three different cleanup operations:

- Particulate matter in the micron and submicron regions.
- Volatile and semivolatile elements and compounds.
- Nitrogen oxides.

The first units in both the HOG and DOG off-gas systems remove particulate matter and any ruthenium present in the off-gas. Removal of particulates is normally accomplished with fiberglass filters or by the more compact HEPA (high-efficiency particulate air) filters. Ruthenium is removed with filters containing a wire mesh. The following section discusses the reference system for removing the waste radioactive gases from these systems before they are released to the environment.

A.2 Head End Off-gas System

After the particulates and the volatile ruthenium are removed from the voloxidation off-gas, the iodine is adsorbed on silver-exchanged mordenite. The off-gas stream is enhanced with hydrogen and passed through a catalytic oxidizer to form HTO from the tritium and hydrogen present. The HTO is then adsorbed in molecular sieve material, the permanent storage medium for the HTO. The remainder of the head-end off-gas stream is combined with the dissolver off-gas stream.

FIGURE A-1. Origins of Radioactive Waste Gases

FIGURE A-2. Off-Gas Treatment Facility

A.3 Dissolver Off-gas System

After the particulates and the volatile ruthenium are removed from the dissolver off-gas stream, oxides of nitrogen are absorbed in water to form nitric acid for recycle. The DOG is next passed through a solvent trap to remove organic material present in the recovered acid. Any remaining oxides of nitrogen are then reacted with ammonia to form nitrogen and water. From there, the DOG is passed through an iodine absorber which functions in the same manner as the one which was described above for the HOG system.

A.4 Combined Off-gas Stream

The remaining off-gases in the HOG and DOG are combined and passed in succession through the following units:

- A water absorber using a molecular sieve which is regenerated to the stack.
- A carbon dioxide absorber using 13X or 3A zeolite which is regenerated to release the carbon dioxide for fixation on barium hydroxide, the permanent storage medium for carbon-14.

The remaining off-gas is then transferred to the Krypton Recovery System.

A.5 Krypton Recovery System

The reference krypton removal system is absorption of a liquid fluorocarbon, or the FCRS. In this system, a refrigerant R-12 based solvent dissolution scheme is used to separate krypton from xenon and the nonradioactive gases. The krypton is then stored in gas cylinders in a temporary storage facility. The product gas is greater than 99% krypton containing about 10% krypton-85. Separated xenon gas can be further purified for potential sale.

A.6 Costs

To develop cost estimates of annual operating and maintenance costs and capital costs, it is necessary to generate conceptual system designs and process flow diagrams. In addition, equipment sizing calculations must be performed for major components based on processing flow rates and required equipment capacities. Since the major objective of this study was to identify alternative processes for treating radioactive gases, a detailed cost estimate was not attempted. The cost estimates presented in Table A1 are

for an off-gas treatment facility associated with an integrated Fuel Reprocessing Complex for processing BWR and PWR irradiated fuel. The design capacity of the FRP is 3000 MTHM/year. The facility is described in References 1 and 2. The cost estimates are values summarized from cost data given in References 2 and 3. As noted in Table A1, the major cost item is the removal and fixation of krypton-85 (44%), followed by tritium (29%), carbon-14 (14%) and iodine-129 (13%). The cost estimates include the cost of recovering krypton-85 using the FCRS method. Reference 4 indicates that the cost would be essentially the same if the cryogenic distillation process was used.

TABLE A1

Capital Costs

<u>Basis</u>	<u>Off-gas Recovery Systems Cost</u>	<u>Million</u>
I-129	AgZ with Hydrogen Regeneration	7
C-14	Molecular Sieve and Barium Hydroxide	8
Kr-85	Fluorocarbon Absorption	25
H-3	Molecular Sieve	<u>16</u>
Total Cost (Third quarter 1978 dollars)		56

A.7 Reference Storage Forms

A.7.1 Iodine-129

Reduced AgZ was chosen as the reference collection medium because of its higher loading capacity, and because it can be regenerated for reuse or for silver recovery. The reference process does not include a regeneration step, however. Thus, the reference storage form is AgIZ (iodine loaded, reduced, silver mordenite). This should be considered only as an interim storage form, however. The ultimate storage form will probably be iodine loaded PbX (lead exchanged faujcrite) for the following reasons:

- Permanent removal of large quantities of silver from the world economy is both expensive and undesirable.
- Costorage of the iodine with silver could result in release of the iodine-129 inventory in the future by unknowledgeable "grave robbers."

AgIZ beds from the reference FRP should be placed in a secure, retrievable storage facility until a less hazardous silver recovery technology is developed. The current technology utilizes hot (500°C) hydrogen gas to convert the AgI to HI. The HI is then trapped on a bed of PbX.³ PbX cannot be used directly in the Plant since it does not collect elemental iodine efficiently.

A.7.2 Krypton-85

The reference storage form for krypton-85 in this study is bottled gas in pressurized cylinders housed in an interim storage facility. Storage of krypton-85 in cylinders under pressure is only an interim storage form, however. Ion implantation of krypton in a metal matrix appears, at this time, to be the safest encapsulation technique for permanent krypton-85 storage. The ion implant technique does not require the high temperatures and pressures necessary for zeolite encapsulation or glass encapsulation. Cylinder storage was chosen for this study because the ion implantation technique is not fully developed.

A.7.3 Carbon-14

The reference storage form for carbon-14 is BaCO₃ encased in concrete. Direct casting of the Ba(OH)₂-8H₂O collection beds in a secondary drum is the preferred technique to minimize handling of the radioactive materials. The fixed bed could be placed in a geologic repository or buried in deep ocean sediments.

A.8 Reference Facility References

1. Spent LWR Fuel Recycle Complex, Conceptual Design, Case A-1, Separated Streams. USDOE Report DP-CFP-78-121, Savannah River Laboratory, E. I. du Pont de Nemours and Co., Aiken, SC (November 1978).
2. Spent LWR Fuel Recycle Complex, Conceptual Design, Coprocessing Case (A-2). USDOE Report DP-CFP-79-121, Savannah River Laboratory, E. I. du Pont de Nemours and Co., Aiken, SC (November 1978).
3. D. T. Pence. Technology Assessment and Cost Estimates for Applying Removal Techniques for Tritium, Carbon-14, Krypton-85, and Iodine-129 from the Nuclear Fuel Cycle. USDOE Report SAI78-737-LJ, Science Applications, Inc., La Jolla, CA (October 20, 1978).
4. J. L. Kovach. Technology and Cost Evaluation of Krypton-85 Control for Nuclear Fuel Reprocessing Plants. NUCON-128, Nuclear Consulting Services, Inc. (June 14, 1976).