^{85}Kr Management Trade-Offs: A Perspective to Total Radiation Dose Commitment

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⁸⁵Kr MANAGEMENT TRADE-OFFS:
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DOSE COMMITMENT

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1.0 SUMMARY

Radiological consequences arising from the trade-offs for ^{85}Kr waste management from possible nuclear fuel resource recovery activities have been investigated. The reference management technique is to release all the waste gas to the atmosphere where it is diluted and dispersed. A potential alternative is to collect, concentrate, package and submit the gas to long-term storage. This study compares the radiation dose commitment to the public and to the occupationally exposed work force from these alternatives.

The results of Table 1.1 indicate that it makes little difference to the magnitude of the world population dose whether ^{85}Kr is captured and stored or chronically released to the environment.

TABLE 1.1. Summary Table of 80-Year Cumulative Radiation Dose Commitments to the World Population

	<u>No Kr Recovery (man-rem)</u>	<u>Kr Recovery (man-rem)</u>
<u>Chronic Routine</u>		
Public Exposure	2.9×10^5	2.6×10^4
Occupational Exposure		3.7×10^5 to 4.2×10^1
SUBTOTAL	2.9×10^5	4.0×10^5 to 2.6×10^4
<u>Accidental Release</u> ^(a)		
Public Exposure		1.3×10^4
Occupational Exposure		2.9×10^2
TOTAL	2.9×10^5	4.1×10^5 to 3.9×10^4

(a) Mathematical expectation of dose from estimates of probability and extent of release of 15 accidents selected to bracket the spectrum of anticipated design-basis events.

Further, comparisons of radiation exposures (for the purpose of estimating health effects) at very low dose rates to very large populations with exposures to a small number of occupationally exposed workers who each receive much higher dose rates may be misleading. Finally, cost studies (EPA 1976 and DOE 1979a) show that inordinate amounts of money will be required to lower this already extremely small 80-year cumulative world population dose of 0.05 mrem/person ($<0.001\%$ of natural background radiation for the same time period).

2.0 INTRODUCTION

The recovery of energy resources from spent nuclear fuel has been indefinitely deferred in the United States while the international safeguards issues associated with plutonium extraction and recycle are reviewed. If it is eventually decided to reprocess spent fuel to recover the residual energy, collection of ^{85}Kr has been required by the Environmental Protection Agency. Collection of about 86% of the ^{85}Kr and long-term storage will be required by Federal Regulations for commercial fuel irradiated after 1982 (EPA 1977). This EPA requirement is in contrast with the historical ^{85}Kr management technique of direct release from tall stacks.

A number of questions have been raised in the technical community about the relative advantages of these two management techniques. Although ^{85}Kr collection would lower public radiation exposure from this radionuclide, it could lead to increased occupational exposures and the potential for accidental releases from storage facilities. This study was undertaken to quantify and compare the radiological consequences of these alternative management techniques. The work is part of the Waste Management Safety Studies Project being performed by PNL for the Office of Nuclear Waste Management in the U.S. Department of Energy.

^{85}Kr is a radioactive noble gas with a half-life of 10.7 years. Important physical properties of Kr are summarized in Section 3.0. Sources of ^{85}Kr in the environment include natural processes, nuclear weapons tests and commercial nuclear power production. These sources are discussed in Section 4.0. This document focuses on management of ^{85}Kr from possible reprocessing of spent nuclear fuel.

A number of ^{85}Kr recovery and storage techniques have been proposed. These were reviewed and a representative system using cryogenic distillation and storage in steel cylinders was selected as the basis for this study. The reference system is described in Section 5.0 and Appendices A, B and E. Because ^{85}Kr recovery has not been demonstrated on a commercial scale, a number of alternative designs for the reference system that have potential to reduce occupational exposure during operation and maintenance of the facility were

also considered in the analysis. The results indicate the range of occupational exposure that could be experienced in a commercial facility, although no attempt was made to determine the economic feasibility of these alternative designs.

Radiological consequences from the total release of ^{85}Kr to the atmosphere are evaluated in Section 6.1. Currently available radiation dose models are used for calculating the dose to the maximum individual and to the general public in the vicinity of the reprocessing facility. Because ^{85}Kr disperses and mixes throughout the global atmosphere, the dose to the world population becomes the most important dose in the total release case. These calculations are also presented in Section 6.1.

For the case of ^{85}Kr recovery, both public and occupational radiation exposures have been evaluated. Radiation doses may occur to workers in the recovery and storage facility during operations and maintenance activities and for accidental releases. Doses to the general public occur from release of the small fraction of the ^{85}Kr not recovered, from release of residual ^{85}Kr from the storage facility at the end of the storage period, and from possible accidents. Occupational exposures during routine operations are evaluated in Section 6.3. Doses to the public and to workers from accident releases are presented in Section 7.0. Overall conclusions and recommendations from the study are presented in Section 8.0.

3.0 PROPERTIES OF ^{85}Kr

This study on radioactive noble gas waste management has focused on ^{85}Kr because it is the most significant of the noble gases from a radiological safety standpoint.

Krypton-85 has a number of unique chemical, radiological and biological properties that are described in this section.

3.1 CHEMICAL PROPERTIES

The noble gases, which include helium, neon, argon, krypton, xenon, and radon, are colorless, tasteless, and in general, chemically nonreactive. In recent years, however, they have been shown to be capable of entering into ionic or covalent bonding with highly reactive elements such as fluorine or oxygen (NCRP 1975). Krypton becomes a liquid at 153°C below zero.

Noble gases have been shown to enter into compounds called clathrates in which the gas atoms are physically entrapped in molecular cages of hydroquinone, or other organic compounds (NCRP 1975). Noble gases have also been shown to be highly soluble in nonpolar solvents with solubility increasing as the temperature decreases (Nichols and Binford 1971).

3.2 RADIOLOGICAL PROPERTIES

Krypton-85 has a half-life of 10.73 years and decays to stable ^{85}Rb . Its decay scheme is rather simple, in that only two beta rays and a single gamma photon are emitted as shown in Figure 3.1. Secondary radiations from ^{85}Kr decay and a tabulation of fission yields are presented in NCRP-44. These data indicate a thermal fission yield from ^{235}U of from 0.29% to 0.34%.

3.3 BIOLOGICAL PROPERTIES

Being chemically inert, krypton and other noble gases are not usually involved in biological processes. They are, however, absorbed into the tissues of the body via inhalation and dissolution in body fluids and tissues. Krypton is characterized by low blood solubility, high lipid solubility, and

FIGURE 3.1. Kr Decay Scheme (NCRP 1975)

rapid diffusion in tissue (Kirk 1972). Several studies of the rate of absorption and desorption in human tissue have been performed (Susskind et al. 1976 and Ellis et al. 1977). The clearance of inhaled krypton from the human body indicates five components ranging in biological half-lives from 14 sec to 9.6 hr (Cohn, Ellis and Susskind 1979). The slowest components, varying between 4.2 and 9.6 hr, correlated highly with percent body fat. From a radiological analyses of these results Cohn, Ellis and Susskind (1979) concluded that when radiation exposure from ^{85}Kr includes an external irradiation of the body, the dose from the inhaled ^{85}Kr gas increases the whole-body dose by about 1%. Kirk (1972) has listed some exceptions to the biologically inert characterization of noble gases obtained from reviewing the literature. Among these are an anesthetic action of xenon in humans, and a radioprotective action by inert gases in animals and bean sprouts.

4.0 PRODUCTION OF ^{85}Kr

Atmospheric ^{85}Kr has its origin from a variety of natural and man-made sources including cosmic activity, weapons detonations, and commercial power generation.

4.1 NATURAL SOURCES

Krypton-85 is present to a small degree (about 12 Ci) in the natural environment due to the mechanisms of spontaneous and neutron-induced fission of natural uranium and neutron capture reactions from cosmic ray neutrons interacting with naturally occurring atmospheric ^{84}Kr .

4.2 NUCLEAR WEAPONS TESTING AND PLUTONIUM PRODUCTION

Since ^{85}Kr is produced during fission, it has been produced and distributed by nuclear weapons tests. The Federal Radiation Council (1963) estimated that the integrated ^{85}Kr generation, between 1945 and 1962, was about 5 MCi, leaving a current inventory from these tests of about 2 MCi, or an average concentration in the atmosphere of 0.5 pCi/m³ (NCRP 1975).

Nuclear reactors and fuel reprocessing plants associated with weapons grade plutonium production have also contributed to the worldwide inventory of ^{85}Kr . This production took place mainly in the Hanford and Savannah River reactors. Although the power levels of these reactors have been classified through most of their histories, there are indications that reprocessing of the fuel from these production reactors accounted for higher ^{85}Kr atmospheric concentrations than did weapons tests (NCRP 1975). The worldwide contribution from plutonium production has been estimated to be 39 MCi as of 1970 (Bernhardt, Moghissi and Cochran 1975). By 1973, the world inventory was reported to be about 53 MCi (Telegadas and Ferber 1975).

4.3 COMMERCIAL NUCLEAR POWER GENERATION

Krypton-85 is formed as a fission product in fuel elements of nuclear power plants. The vast majority of the krypton is retained in the fuel element

and would only be released if the fuel were reprocessed. The quantities of fission products present in spent nuclear fuel have been previously reported, based on calculations using the computer code ORIGEN (ORNL 1970; Exxon Nuclear 1976; and Brown et al. 1978). For ^{85}Kr this value ranges from 8,500 to 11,200 Ci/MTHM (curies/metric ton of heavy metal) in the fuel. For fuel irradiated at a specific power of 30 MW/MTHM with a total burnup of 33,000 MWd/MTHM, the calculated ^{85}Kr content 160 days after discharge of the fuel from the reactor is about 10,500 Ci/MTHM. It has been assumed for this study that a light-water-cooled nuclear power plant operates at 33% thermal efficiency, producing about 33 MTHM of spent fuel annually with this burnup for each 1000 MWe of power produced.

A small percentage of fuel elements develop minute leaks in the fuel cladding during irradiation and handling releasing <1% of the noble gases to the reactor off-gas system. Therefore, the routine releases of ^{85}Kr from commercial LWR's are insignificant when compared to the potential releases from fuel reprocessing operations. The reference fuel reprocessing plant has a throughput capacity of 2000 MTHM/yr. Such a plant would annually generate radioactive waste gases containing about 2.1×10^7 Ci of ^{85}Kr . A reprocessing plant of this size will service about sixty 1000 MWe nuclear power plants.

5.0 RECOVERY, PACKAGING AND STORAGE OF ^{85}Kr

While the past practice of noble gas management has been to disperse ^{85}Kr to the atmosphere, industrial control methods have been proposed for its recovery and storage to avoid long-term global buildup. Of these, collection of ^{85}Kr by cryogenic distillation (see Appendix A) and storage in pressurized cylinders (see Appendix B) is the only technology considered to be currently available. Cryogenic distillation is the reference krypton recovery system in this study. Because the ^{85}Kr is potentially highly mobile in pressurized cylinder storage, ion implantation of ^{85}Kr into amorphous metals has also been chosen for examination because of its immobilization capabilities for ^{85}Kr (see Appendix B).

Other krypton recovery systems, in various stages of development, have been proposed. These alternatives include liquid fluorocarbon absorption (Stephenson, Eby and Huffstetler 1977), cryogenic charcoal adsorbents (Kanazawa 1976), ambient-temperature adsorption, permselective membranes and clathrate precipitation (Nichols and Binford 1971). The application of many of these to the process flow stream anticipated at a commercial FRP is questionable (ERDA 1976).

Alternative packaging techniques to pressurized steel cylinders include sorption by zeolites (Knecht 1977), ion implantation/sputtering into metals (Tingey et al. 1979), and immobilization into solid granules (Benedict 1977). Thamer, Muhlfeith and Macbeth (1979) have conducted an extensive review of ultimate disposal alternatives for ^{85}Kr . In addition to the above, their report considers disposal in underground media, hydrofracture, extraterrestrial, disposal in the oceans, low-pressure storage in caverns and others. Recently, Klett (1979) conducted a conceptual evaluation of disposal alternatives. Five above-ground and two near-surface geologic concepts were evaluated. Fifteen features that are important for ^{85}Kr disposal were selected for evaluation. Of seven alternatives, the dry well concept was determined to be the best choice for ^{85}Kr disposal and the air-cooled vault the least desirable. We submit that the methodology used for this trade-off study can be readily applied to any alternative of gas recovery, packaging or storage.

A ^{85}Kr storage facility has been conceptually designed (Brown, Knecht and Thomas 1978) and modified for use in the Commercial Waste Management Statement (DOE 1979). This conceptual design has been adopted for use in this study as a base case. A description of the reference design and a few alternatives are presented in Appendix E. Alternative designs include relocating offices, adding shielding and removing high frequency maintenance equipment from radiation zones to further reduce occupational radiological exposure.

6.0 THE DOSIMETRY OF THE RELEASE OF ^{85}Kr TO THE ATMOSPHERE

A detailed discussion of the radiation dosimetry of ^{85}Kr is given in Appendices C and D. This section briefly reviews the dosimetry and compares the 80-year cumulative doses expected from different ^{85}Kr management strategies. The 80-year cumulative dose is a combination of 30 years of plant operation followed by 50 years of krypton storage. This scenario is discussed in more detail in Appendix D.

6.1 PUBLIC EXPOSURE TO ^{85}Kr FROM FUEL REPROCESSING

Three segments of the public are evaluated for ^{85}Kr exposure. The maximum individual is hypothetically located at the point where the gaseous plume first touches the ground. The regional population dose is evaluated to a distance of 50 miles in all directions from the FRP and finally the world population dose is calculated.

If it is decided to recover uranium and plutonium from spent nuclear fuel, the major source of ^{85}Kr from the nuclear fuel cycle will be the dissolver off-gas (DOG) system of a spent fuel reprocessing facility. The ^{85}Kr management technology adopted in previously built reprocessing plants was dilution and dispersion from tall stacks. The total release of all the ^{85}Kr entering the DOG system of the reference reprocessing plant would be about 2.1×10^7 Ci/yr or 0.67 Ci/sec. This value assumes 365 days of plant operation annually, even though the plant is expected to be operating for only about 300 days each year. This is the amount of ^{85}Kr which would make up the total release case.

Cryogenic distillation and fluorocarbon absorption appear to be the most promising alternatives for rare gas recovery. A discussion of alternative recovery systems is presented in Appendix A. Cryogenic distillation is chosen as the reference process to isolate a relatively pure krypton stream. Isotopes of xenon present in amounts about 6 to 12 times greater than krypton, are either stable or have half-lives so short (5 days maximum) that xenon does not present a radiation hazard at the time of reprocessing (Brown, Knecht and Thomas 1978). Krypton-85 will comprise about 6% of the total krypton which yields an activity in the range of 11 to 14 Ci/m³ in the collected gas (Brown,

Knecht and Thomas 1978). Of the 21 million curies of ^{85}Kr which enter the DOG of a 2000 MT/yr reprocessing plant, almost 20 million curies can be recovered by a properly designed and operated krypton collection system based on known cryogenic technology. The release fraction of 0.05 yields an annual release of 1.05×10^6 curies, or 0.033 Ci/sec to the environment. This discharge is about 1.8×10^4 Ci/GWyr of electricity generated by the 60 reactors served by the facility. This is less than the proposed EPA guidelines of 5×10^4 Ci/GWe. Radiation doses are calculated in accordance with the models presented in Appendix D.

6.1.1 Maximum Individual Doses

The radiation doses to an individual located at the point of maximum ground level concentration of ^{85}Kr were calculated utilizing a release height of 110 m, and the meteorology and population distribution from Appendix D. The results are shown in Table 6.1.

TABLE 6.1. The 80-Year Cumulative Radiation Doses to the Maximum Individual

<u>Organ</u>	<u>Total Release Dose (rem)</u>	<u>95% Recovery Case Dose (rem)</u>
Skin	4.2×10^{-1}	2.1×10^{-2}
Total Lung ^(a)	1.1×10^{-2}	5.5×10^{-4}
Total Body	5.7×10^{-3}	2.8×10^{-4}

(a) Inhalation dose plus total-body dose.

6.1.2 50-Mile Population Doses

The 80-year cumulative total-body radiation dose to the 1.96×10^6 people (assumed constant for the duration of plant operations) residing within 50 miles of the facilities is about 1.3×10^3 man-rem for total release and 6.4×10^1 man-rem for 95% recovery of ^{85}Kr .

6.1.3 Worldwide Population Doses

Krypton-85 discharged from a tall stack will be dispersed from the plant site and become fairly well mixed throughout the global atmosphere in about 2 years.

It has been estimated by the NCRP (1975) that the world ocean is a trivial sink for the ^{85}Kr discharged into the atmosphere, accounting for about 3% of atmospheric krypton. Atmospheric depletion of ^{85}Kr by means of washout, dry deposition and deposition through adsorption on particulate matter can act as a sink for only small amounts of ^{85}Kr because of competition from the much larger quantities of natural krypton in the atmosphere (NCRP 1975). Therefore, the majority of the ^{85}Kr released to the atmosphere will stay there where its concentration will be reduced only by radioactive decay. Krypton-85 disperses fairly evenly over the entire globe because of its 10.7-year half-life and the lack of significant sinks. Most previous global dispersal schemes have assumed instantaneous dispersion into the world atmosphere. However, more sophisticated analyses are available in the literature (Machta 1974; Heffter and Taylor 1975; and Machta, Ferber and Heffter 1974). The more detailed multi-compartment models differ from the simpler models in their treatment of regional distribution of doses delivered immediately following release. The total collective doses, however, agree within a few percent of those calculated by the simpler models.

For the purposes of this study, the important dose is that from many years of continuous exposure. Hence, the less sophisticated uniform global mixing models have been used.

The simple model ignores the higher concentrations of ^{85}Kr both near the source and on the first few passes around the world. As a result, this model underestimates the local and regional dose at short times after release, (hence, the independent calculations in Sections 6.1 and 6.2). However, the net effect on the worldwide dose from long-term accumulated dose commitment is small (<10% for a 68-year exposure period) depending on whether the nuclear facility is sited in the midwest or east coast (NCRP 1975). The rapid mixing across

the equator (in relation to the 80 years of interest) even makes separate accounting of the northern and southern hemisphere population doses unnecessary.

The volume of the world's atmosphere is $3.96 \times 10^{18} \text{ m}^3$ (NCRP 1975). The average concentration of ^{85}Kr in the atmosphere at any point in time is simply the cumulative amount released corrected for radioactive decay divided by the volume of the atmosphere.

Using a constant world population of 6.4 billion persons and the radiation dose models from Appendix D, the 80-year cumulative world population dose to the total-body from the operation of one 2,000 MT reprocessing plant is about 2.9×10^5 man-rem for the total release case and 1.5×10^4 man-rem for 95% recovery of ^{85}Kr .

In addition to the public exposure from the ^{85}Kr released from the reprocessing stack, the public will be exposed to radiation during transportation of the ^{85}Kr to its storage facility if the storage facility was not colocated with the FRP. In this study the storage facility shares the same site with the FRP. The public will, however, be exposed to the ^{85}Kr which is expected to be discharged from the facility during storage and finally to ^{85}Kr released to the atmosphere intentionally after 50 years of storage. These doses must be added to the 1.5×10^4 man-rem world population dose from ^{85}Kr recovery.

The world population dose resulting from the accidental release of ^{85}Kr from the reprocessing facility and the storage facility must also be added to this analysis.

6.2 RADIATION DOSE TO THE PUBLIC FROM THE STORAGE OF ^{85}Kr

After the ^{85}Kr pressurized gas cylinders are filled within the FRP, they are transferred to the colocated storage facility. Each year 150 cylinders will enter storage. Upon entry into storage, each cylinder contains 130,000 Ci of ^{85}Kr . After 50 years of storage, about 4% of the original activity (about 5100 Ci) remains and will be released. Therefore, ^{85}Kr will be released to the atmosphere from 150 fifty-year old cylinders each year for

30 years. Leakage during the 50 years of storage is negligible. Therefore about 7.65×10^5 Ci of ^{85}Kr will be intentionally released each year for 30 years.

6.2.1 Maximum Individual Dose

The methodology for this dose calculation is found in Section D.1.3 of Appendix D. The 80-year cumulative dose to the total body of the maximum individual from the release of almost one-half a cylinder per day for 30 years is 2.1×10^{-4} rem.

6.2.2 50-Mile Population Dose

The 50-mile population 80-year cumulative dose to the total body is about 46 man-rem.

6.2.3 Worldwide Population Dose

The 80-year cumulative radiation dose to the world population is calculated as shown in Section D.1 of Appendix D, and is about 1.07×10^4 man-rem.

6.3 OCCUPATIONAL DOSE RATES

When all the produced ^{85}Kr is released chronically from tall stacks, there is essentially no occupational exposure to operational or maintenance personnel which can be specifically attributed to ^{85}Kr . There are no transportation or storage activities to be concerned with, so the only radiation exposures identified with the chronic release of all the ^{85}Kr from the plant are the nonoccupational ones already discussed.

Occupational doses are received when ^{85}Kr is recovered during the operation of the ^{85}Kr recovery and storage facilities. Radiation doses could include contributions from external and internal exposures. External exposure may arise from discrete shielded sources of ^{85}Kr , such as gas cylinders or cryogenic distillation columns, as well as from airborne ^{85}Kr (air submersion). In the latter case, inhalation of ^{85}Kr leads to internal exposure to the lungs.

It is expected that most of the occupational exposure will arise from external radiation exposure. Since almost all of the ^{85}Kr operations will be remotely handled, most of the occupational doses will arise from equipment maintenance.

The following sections give the details of the occupational doses received under various operating regimes for ^{85}Kr recovery, transportation and storage, respectively.

6.3.1 Occupational Doses for Krypton Recovery Facility

Cryogenic distillation has been chosen as the reference process for recovery of the ^{85}Kr . The details of this process are given in Appendix A. Briefly, this method involves removing small amounts of various gases, such as NO_x , O_2 and H_2O , from the DOG system which may interfere with the cryogenic process. The purified gas is then cooled and enters a recovery column. Krypton and xenon are stripped from the gas stream by liquefaction. The krypton and xenon are separated by distillation at cryogenic temperatures. The xenon is released up the stack. The liquid krypton is held in a cryogenic receiver until enough is collected to fill a gas bottle, then the liquid krypton is heated to form a gas which is bottled in stainless steel gas cylinders. The process flow sheet for the operation is given in Figure A.3 of Appendix A.

It is assumed that the cryogenic recovery facility operates as part of the fuel reprocessing plant. The cryogenic ^{85}Kr recovery facility is assumed to operate 24 hours per day, 7 days a week, for 300 days per year. Normal work activities include monitoring the NO_x , H_2O and CO_2 and Kr removal systems and performing normal maintenance and replacement. A floor plan of the facility is given in Figure A.1 and an elevation view is given in Figure A.2 (DOE 1979). All normal process operations are handled in the hot cell, and workers do not normally come in contact with the ^{85}Kr .

There are several potential mechanisms by which the workers in this facility can receive occupational doses. The first is internal dose from ^{85}Kr in air the workers breathe. It is assumed that a radiation zone concept is employed and that negative air balances with high enough flow rates are

maintained so that the workers are never exposed to ^{85}Kr from small leaks under normal operations. Barring accidents, the workers will not enter areas where ^{85}Kr may be present in the air except during equipment maintenance in the hot cell. Under these latter conditions, the air is carefully monitored for radioactivity before the workers enter the cell. Unless some major accident occurs, there is essentially no internal occupational dose in the ^{85}Kr recovery operation. All ^{85}Kr contaminated air is vented to a 110-m stack.

The second way workers can receive occupational doses is by external exposure during routine operations. All operations are remote behind heavily shielded walls. The highest dose rate outside the hot cell is only 0.015 mrem/hr behind the 3-ft thick concrete walls where an operator stands to use the manipulators. Assuming that 150 gas bottles are filled per year and that it takes approximately 4 hours to fill a bottle, the annual dose is 9×10^{-3} man-rem. Preparing the filled gas bottles for shipment would take one hour each in an area with a dose rate of 0.013 mrem/hr. The annual dose from this operation is 2×10^{-3} man-rem. All other operations are carried out in areas where the exposure rates are even lower than those listed above, and therefore, no additional occupational dose is accrued.

Since these operations will continue for 30 years at approximately the same exposure rates, the total integrated occupational dose for routine operation of the krypton recovery system is 0.33 man-rem.

The final way workers can receive an occupational dose is by performing maintenance on the large amount of equipment in the hot cells. This is the most significant source of exposure. There are two types of maintenance; preventive maintenance and breakdown maintenance.

Preventive maintenance can be performed on a scheduled basis. For this study, it is assumed that the plant works on an established shutdown and maintenance schedule and follows ALARA practices. It is assumed that the plant is operated 300 days per year; the remaining 65 days will be used for maintenance.

Breakdown maintenance can be handled two ways. The first is to purge the equipment of all ^{85}Kr and repair it in a low radiation field; the second is

to enter the hot cell and work very rapidly to remove modularized equipment so it can be easily removed to a shop outside the radiation zone for repair. If the ^{85}Kr is not removed from the cold box, the second method can lead to very high exposures. To minimize the radiation doses during breakdown maintenance, it is assumed that critical systems, such as compressors, some transducers and liquid level indicators, will have a redundant back-up unit so that the plant will not have to be shut down frequently due to failure of these components. It was also assumed that much of the equipment is modularized, so that it can be easily removed and repaired during scheduled shutdowns for preventive maintenance. It may be a good idea to have a second cold box for a replacement if equipment in the first cold box malfunctions.

The type of equipment used in the krypton recovery facility and the maintenance requirements are listed in Table 6.2. The maintenance needs for equipment in the recovery system is based on data from and experience of PNL Maintenance and Craft Services. A factor of two is used to account for the increased difficulty of doing work in a radiation zone. The ^{85}Kr inventories in each piece of equipment are determined based on data taken from Brown, Knecht and Thomas (1978) which is presented in Figure A.3 for the composition of the gas and for the volumes of the pieces of equipment. Dose rates are calculated at various points near the pieces of equipment. The results of some of these calculations are presented in Figure 6.1. Note that the hot cell for filling the gas cylinders and the cells containing the cold box are assumed to have 3-ft thick concrete walls, and the doses outside the cells are very low.

Occupational doses for maintenance of the krypton recovery system are calculated for the lifetime of the reprocessing facility (30 years). This is done by multiplying the man-hours required for maintenance per year by the dose rate (rem/hr) and then by 30 years. Since there is a constant supply of krypton to this system, an accurate estimate of the integrated dose is given without taking radioactive decay into consideration.

Occupational dose calculations have been made for several system design and operating alternatives. The first alternative considered maintenance of

TABLE 6.2. Maintenance Requirements for Recovery Facility

Equipment	Number	Man-hr/yr per Piece of Equipment (a)
Valves	91	1
Crane	1	100
Primary process condenser	1	70
Secondary process condenser	1	70
Demister	1	15
Process gas dryer	3	150
Cryogenic cold box		
columns	5	40
receivers	2	15
valves	20	1
electric heaters	9	4
Oxygen recombiner	1	0.5
Hydrogen generator and gas holder	1	52 ^(b)
Recycle gas cooler	2	70
Process gas compressor	2	70
Refrigeration unit	1	100
Deisel engine and electric generator	1	170 ^(b)
Manipulators	6	280

(a) This maintenance time must be multiplied by two (unless otherwise noted) since the work will be done in a radiation zone.

(b) This equipment is outside a radiation zone.

the system with krypton remaining in the equipment and with one inch of lead shielding on the cold box. The occupational dose for this case is 470 man-rem/yr most of which is received from work in or near the cold box. This dose level is not consistent with good ALARA practices. This design should not be considered as a viable alternative, but is used for comparison with other alternatives. It does, however, point out the component which is of most concern for radiation protection of workers.

EQUIPMENT

1. OXYGEN RECOMBINER	7. PROCESS GAS COMPRESSOR (2)
2. HYDROGEN GENERATOR	8. SEPARATOR - DEMISTER
3. HYDROGEN STORAGE GAS	9. PROCESS GAS DRYER (3)
4. PRIMARY GAS COOLER (2)	10. CRYOGENIC COLD BOX
5. PRIMARY PROCESS GAS CONDENSER	11. MANIPULATORS
6. SECONDARY REFRIGERATED GAS COOLER	

<u>POINT</u>	<u>DOSE RATE (mrem/hr)</u>
A	0.015
C	0.009
D	0.001
H	0.67
I	0.61
J	0.002 *
L	0.004 *
P	0.003 *

* NO ⁸⁵Kr IN COLD BOX

FIGURE 6.1. DOG Krypton Recovery System

Other design options for which occupational dose calculations were made consider adding another inch or two of lead shielding on the cold box and consider purging the cold box prior to maintenance. Table 6.3 lists occupational doses for both operational and maintenance activities for the various design alternatives.

TABLE 6.3. Occupational Exposure for Kr Recovery Facility Operated for 30 Years (man-rem)

<u>Case</u>	<u>Operational</u>	<u>Maintenance</u>	<u>Total</u>
One-inch Pb on cold box	0.33	14,000	14,000
Two-inch Pb on cold box	0.33	67	67
Three-inch Pb on cold box	0.33	8.6	8.9
Cold box system purge for maintenance	0.33	8.0	8.3

It will probably require one to two days to shut down the cold box, warm it to room temperature, purge it of ^{85}Kr with dry nitrogen gas, repair or replace any malfunctioning equipment, and cool down the box to liquid nitrogen temperatures to return it to service. For this reason, it is assumed that there are redundant liquid level indicators and pressure and temperature transducers so that downtime may be held to a minimum.

As seen in the Table 6.3, significant savings of occupational dose can be made by adding some shielding or by purging the cold box. The decision as to which alternative should be chosen must be based on an analysis that includes both savings in occupational exposures and economic factors such as capital cost.

6.3.2 Occupational Doses from Transportation Between Facilities

Occupational external exposures will result from the transport of ^{85}Kr filled gas cylinders between the recovery facility and the storage facility. The personnel who are exposed to radiation from transportation include fork-lift operators, radiation monitors, and security guards.

Radiation exposure rates calculated for a gas bottle filled with 130,000 Ci of ^{85}Kr are listed in Table 6.4. These calculations clearly show that these gas bottles must be well shielded if any individual could be in the area. This being the case, the gas bottles will be placed inside a shielded cask for transport. The cask is assumed to have 7-cm thick lead walls.

TABLE 6.4. Radiation Exposure Rates from a Gas Bottle Filled with 130,000 Ci of ^{85}Kr (R/h)

<u>Distance from Side of Container</u>	<u>Unshielded Cylinder</u>	<u>Cylinder Inside Shielded Cask(a)</u>
Surface	3.1×10^3	7.4×10^{-5}
1 meter	1.2×10^2	1.1×10^{-5}
15 meters	6.2×10^{-1}	5.7×10^{-8}

(a) Seven centimeters of lead shielding is assumed.

Each of the 150 bottles filled during a year must be transported from the recovery facility to the storage facility. These facilities are assumed to be about 0.25 miles apart. The special transport cask is carried by a forklift designed to accommodate it. The forklift travels at 250 ft/min, taking about 5.5 minutes to complete the trip.

It is assumed that the forklift operator sits about 3 ft from the cask during transit. At 3 ft from the surface of the cask the dose rate is 1.2×10^{-5} R/h. The dose to the forklift operator received as a result of 150 shipments is 1.7×10^{-4} man-rem/yr.

The transfer casks will be surveyed routinely, both upon leaving the recovery facility and arriving at the storage facility. This will require that a monitor be present, 3 ft from the cask, for a total of 20 minutes per shipment (10 minutes at the recovery facility and 10 minutes at the storage facility). The dose to monitors as a result of 150 shipments per year is 6.0×10^{-4} man-rem/yr.

Security measures are expected to require that two guards be present at both the ^{85}Kr recovery and storage facilities. Every shipment will be inspected at each facility. Assuming that each two-guard team spends 10 minutes per shipment for vehicle inspection, etc., at 3 ft from the cask, the dose is 6.0×10^{-4} man-rem/yr. Based on these calculations, occupational doses from the routine transportation of ^{85}Kr gas cylinders from the recovery facility to the storage facility are about 1.4×10^{-3} man-rem/yr. This results in an integrated occupational dose for the 30 year recovery facility operating lifetime of only 4.1×10^{-2} man-rem. This dose does not contribute significantly to the total occupational doses received for any of the recovery/storage alternatives.

6.3.3. Occupational Doses in the Krypton Storage Facility

Three possible krypton storage facility designs are considered. The base case is the horizontal gas cylinder storage with gas cooling (Brown, Knecht and Thomas 1978) as described in Section E.1.2. This is probably the most widely accepted storage design. Two alternative designs are also considered only from the point of view of reducing the occupational dose. These may be unrealistic design alternatives because no consideration is given to construction and operation economics, heat loading limitations, seismic or tornado design limitations, etc. These designs are offered only as possibilities that may lead to lower radiation doses. The first alternative is for vertical gas cylinder storage using bottom loading casks to minimize operator exposure during transfer of cylinders into the cask. Bottom loading casks are commonly used by the nuclear industry and are designed to prevent excessive dose rates during transfers. The second alternative is pool storage. This is also used quite commonly in the nuclear industry for storage of spent fuel elements.

A primary advantage of pool storage is the use of an inexpensive shield material, water. Higher storage densities and lower building costs may be possible. Pool storage may be applicable to the sputtering technique discussed in Section B.2.2. Pool storage may have the disadvantage of potential hazards from leaking gas cylinders. Each of these options is discussed in detail in the following text.

Horizontal Storage with Gas Cooling

The horizontal storage facility is discussed in detail by Brown, Knecht and Thomas (1978) and the DOE (1979 and 1979a). This design assumes that gas cylinders are brought into the facility inside a shielded transport cask. The cylinders are transferred into a hot cell and undergo inspection and are then transferred into a horizontal cask on a specially designed forklift device for movement to storage. The cylinders are transported into a long corridor with horizontal, shielded storage racks on each side. The cask is attached to a ball valve, and the gas cylinder inserted into a horizontal storage rack behind a concrete shield. The gas cylinder may also be removed and taken back to the hot cell. These cask transfer operations may be performed manually by an operator on the forklift or remotely by computer control. There are currently several computer controlled remote storage facilities in the nuclear industry. (For example, a remotely operated stack/retriever is used at Rocky Flats for handling plutonium.) There are external and internal radiation exposures associated with this storage facility.

Air submersion and inhalation doses to storage facility employees may result from the radioactive material released routinely by the reprocessing plant. Table 6.5 lists the source term used to calculate doses to workers at the storage facility. An average of the atmospheric dilution factors for all directions at 0.25 miles was calculated to be 5×10^{-9} sec/m³ from an AEC data base (1972a) and modified by an NRC computer code (Sagendorf and Gall 1977). Using the atmospheric dilution factor a total body dose of 3.1×10^{-4} rem/yr is calculated from inhalation and 3.1×10^{-6} rem/yr from air submersion. This yields an integrated dose of about 0.11 man-rem for the life of the reprocessing facility to the dozen men at the storage facility. This dose does not contribute significantly to total occupational doses received for any of the recovery/storage alternatives.

Radiation doses to workers result from the operation and maintenance of the storage facility. Several cases are considered for the horizontal storage alternative with different amounts of shielding. In this section, all standard operating activities are assumed to be manual (hands-on) rather than remote. Maintenance activities are also handled manually. The type of

TABLE 6.5. Radionuclides Released from
the Reprocessing Facility Stack

<u>Nuclide</u>	<u>Ci Released per Second</u>
^3H	4.2E-2
^{14}C	4.0E-5
^{85}Kr	3.3E-2
^{89}Sr	5.4E-11
^{90}Sr	5.0E-11
^{90}Y	5.0E-11
^{91}Y	9.5E-11
^{95}Zr	1.8E-10
^{95}Nb	3.2E-10
^{103}Ru	1.1E-9
$^{103\text{m}}\text{Rh}$	5.1E-11
^{106}Ru	7.2E-9
^{106}Rh	3.4E-10
^{129}I	4.0E-10
^{131}I	6.1E-9
$^{131\text{m}}\text{Xe}$	8.7E-5
^{133}Xe	4.9E-8
^{134}Cs	1.4E-10

equipment used in the horizontal storage facility and the maintenance requirements are listed in Table 6.6. The maintenance needs for equipment in the storage facility is based on data from an experience of PNL Maintenance and Craft Services. A factor of two is used to account for increased difficulty of doing work in a radiation zone. A computer code is used to calculate dose rates of positions where workers might be when maintaining equipment.

External doses from routine operations are highly dependent upon the amount of shielding used and the mode of plant operation. After a gas bottle is received and inspected, it is transported to a storage cell. If this transfer is done using a forklift with an operator, not remotely, virtually all of

TABLE 6.6. Maintenance Requirements for
Horizontal Storage Facility

<u>Equipment</u>	<u>Number</u>	<u>Man-hr/yr per Piece of Equipment</u> ^(a)
Valves	100	1
Casks		
electric motors	6	12
hydraulic equipment	3	110
Carriage/Forklift	2	100
Cranes	3	100
Blowers	42	60
Chillers	42	100
Diesel engine and electric generator	1	170 ^(b)
Filters	40	140 ^(b)
Manipulators on examination cell	6	280

(a) This maintenance time must be multiplied by two (unless otherwise noted) since the work will be done in a radiation zone.

(b) This equipment is outside a radiation zone.

the occupational dose results from the operator being inside of the storage corridor near the storage cells. Since the hot cell has at least 3-ft thick concrete walls and the transfer cask is designed so that there is no massive radiation doses, those persons engaged in activities related only to these entities receive an insignificant dose.

The forklift operator, however, may receive a significant dose. If the cask transport travels at 100 ft/min, the average time in the storage area is about 15 minutes per transfer. Table 6.7 lists the occupational doses associated with the horizontal storage facility. The second column lists the occupational dose from routine operational activities for the shielding options shown in column one. This table also reveals that the occupational doses for preventive and breakdown maintenance are much larger than those associated with plant operations.

TABLE 6.7. Occupational Doses for Kr Storage Facility-
Manual Operation Using Horizontal Storage
Operated for 80 Years (man-rem)

<u>Case</u>	<u>Operational</u>	<u>Maintenance</u>	<u>Total</u>
24 inch side and 18 inch end walls on cells	1500	3.6×10^5	3.6×10^5
30 inch side and 30 inch end walls on cells			
blowers and chillers in cell	33	2100	2130
blowers and chillers in gallery	33	49	82
Shielded to maximum of 1 mrem/hr	21	600	620

Occupational doses for maintenance of the krypton storage facility are calculated for the expected plant life of 80 years. This is done by multiplying the man-hours required for maintenance per year by the dose rate and then by a factor which accounts for the loading rate of the facility, the radioactive decay of ^{85}Kr and the plant life.

Several different options are considered for this design. In one design (see Appendix E, Figure E.2), the fans and chillers are contained in cells with 18-inch thick walls in line with the ^{85}Kr storage cells. This gives very high dose rates (up to almost 500 mrem/hr) in the area where maintenance would be performed. This design option is recognized to be unrealistic and would not be utilized in an operating facility. It is, however, presented in the open literature (Brown, Knecht and Thomas 1978) and is therefore included here for comparison. A second set of dose calculations with 30 inches of concrete shielding results in a significant savings in occupational exposure. Using this design and moving the blowers and chillers to a separate equipment gallery saves even more occupational dose as illustrated in column 3 of Table 6.7.

As seen in this table, significant savings of occupational dose can be made by adding shielding, moving equipment that requires maintenance to a low exposure area or both.

Alternate Storage Designs

In this section, several alternatives to the horizontal storage facility with manual operation are discussed. The first alternative is a remotely operated horizontal storage facility. The other designs are a vertical storage facility and a pool storage facility.

The remote horizontal storage facility is operated in the same manner as the manual facility with one basic difference. In this design, no operator is needed on the forklift which transfers the krypton cylinders between the storage cells and the hot cell. Occupational doses for this design are about an order of magnitude lower than for the manually operated horizontal facility.

The first of the other two alternative designs is a vertical storage facility with gas cooling as shown in Appendix E, Figures E.4 and E.5. The design is very similar to that used for horizontal storage. The blowers and chillers and other items requiring maintenance are located away from a high radiation zone and 36 inches of concrete is used as shielding. The operation of the vertical storage facility is presented in Appendix E, Section E.1.2.2.

The type of equipment used in this krypton storage facility and the maintenance requirements are listed in Table 6.8. A factor of two is used, when calculating occupational doses, to account for the increased difficulty of doing work in a radiation zone. Radiation exposure rates are calculated near the pieces of equipment that require maintenance. Occupational doses for this design are listed in the next section.

The maintenance doses are significantly reduced by placing the chillers and blowers behind 3 ft of concrete shielding. Narrow ducts with several bends in them are used in the design to minimize radiation scattering into the equipment bays. Note that every two units are paired so that, if a blower or chiller fails on one unit, cooling can still be provided by the other. The equipment is placed in equipment bays under hatches so that defective equipment can be easily replaced if necessary. Forty-two units containing 35 gas cylinders each are required to match the storage capacity of the horizontal design.

TABLE 6.8. Maintenance Requirements for Vertical Storage Facility

<u>Equipment</u>	<u>Number</u>	<u>Man-hr/yr per Piece of Equipment (a)</u>
Valves	100	1
Casks		
electric motors	4	6
hydraulic equipment	3	110
gaskets	2	12
Cranes	2	100
Blowers	43	60
Chillers	43	100
Diesel engine and electric generator	1	170 ^(b)
Filters	43	140
Manipulators on examination cell	6	280
Radiation sensors	42	52 ^(b)

(a) This maintenance time must be multiplied by two (unless otherwise noted) since the work will be done in a radiation zone.

(b) This equipment is outside a radiation zone.

The last alternative design is pool storage using water rather than air as a coolant. Water has a much higher specific heat and better heat transfer characteristics, so that much greater storage densities may be used and smaller amounts of water coolant need to be pumped. A plan and elevation view of a pool storage facility is shown in Figures E.6 and E.7, respectively. The operation of the pool storage facility is presented in Section E.1.2.3.

The type of equipment used in the pool storage design and the maintenance requirements are listed in Table 6.9. A factor of two is used when calculating occupational doses to account for the increased difficulty of working in a

TABLE 6.9. Maintenance Requirements for Pool Storage Facility

<u>Equipment</u>	<u>Number</u>	<u>Man-hr/yr per Piece of Equipment^(a)</u>
Valves	184	1
Electric motors	25	12
Cranes	2	100
Pumps	23	80
Heat exchangers	23	70
Diesel engine and electric generator	1	90 ^(b)
Filters	24	148
Clean ion exchange columns	23	12
Clean pool area	23	70
Radiation sensors	23	52 ^(b)

(a) This maintenance time must be multiplied by two (unless otherwise noted) since the work will be done in a radiation zone.

(b) This equipment is outside a radiation zone.

radiation zone. Radiation exposure rates are calculated near the pieces of equipment that require maintenance. Occupational doses for this design are listed in the next section (see Table 6.10).

TABLE 6.10. Occupational Doses for Kr Storage Facilities - Alternative Designs Operated for 80 Years (man-rem)

<u>Alternative</u>	<u>Operational</u>	<u>Maintenance</u>	<u>Total</u>
Horizontal storage with remote operation and shielded to maximum of 1 mrem/hr	2.6	600	603
Horizontal storage with remote operation and blowers and chillers in gallery	2.6	49	52
Vertical storage with remote operation	0.70	66	67
Pool storage with remote operation	1.7	32	34

Occupational Doses for Alternative Storage Facility Designs

In order to evaluate the occupational dose from design improvements to the krypton storage facility, dose rate calculations have been done for each of the designs. Table 6.10 compares operational and maintenance doses for the three basic alternative designs. As will be noticed from the table, significant reductions in dose rates occur as the design is improved.

Occupational doses for maintenance operations are calculated for the expected plant life of 80 years. This is done by multiplying the man-hours required for maintenance (as listed in Tables 6.7, 6.8 or 6.9) by the calculated dose rate near the equipment and then by a factor which accounts for the loading rate of the facility, the radioactive decay of ^{85}Kr and the plant life.

6.3.4 Comparison of Occupational Doses for Various Options of Recovery and Storage

In order to evaluate the possible savings in occupational dose, the calculated doses for the various options for recovery and storage facility designs have been tabulated. Table 6.11 contains occupational doses for the manually operated horizontal storage facility (base case) plus those for the recovery facility options. Table 6.12 contains occupational doses for the alternative designs of the storage facility plus those for the recovery facility options. The two tables consider all of the options discussed in the sections on occupational dose. These tables show the total occupational dose commitment from routine operation and maintenance for the various recovery/storage options. The commitment period for these doses is 80 years; they include all doses from the 30 years of recovery facility operation plus the doses from 80 years of the storage facility operation. No occupational doses due to accidents have been included in this tabulation.

TABLE 6.11. Comparison of 80-year Cumulative Occupational Doses for the Manually Operated Horizontal Storage (Base Case) Plus Krypton Recovery

Recovery Facility Options	Storage Facility Shielding Options			
	24-in. Side 18-in. End	30-in. Side and End Blower and Chillers in Storage Cell	30-in. Side and End Blower and Chillers Outside Gallery	Shielding to Max. of 1 mrem/hr
1-in. Pb shielded cold box	3.7×10^5	1.6×10^4	1.4×10^4	1.5×10^4
2-in. Pb shielded cold box	3.6×10^5	2.2×10^3	1.5×10^2	6.9×10^2
3-in. Pb shielded cold box	3.6×10^5	2.1×10^3	9.1×10^1	6.3×10^2
Cold box system purged for maintenance	3.6×10^5	2.1×10^3	9.1×10^1	6.3×10^2

TABLE 6.12. Comparison of 80-year Cumulative Occupational Doses for Alternative Storage Designs Plus Krypton Recovery

Recovery Facility Options	Options for Storage			
	Horizontal W/Remote Op. Shielded to 1 mrem/hr Max.	Horizontal W/Remote Op. Blowers and Chillers in Gallery	Vertical Storage	Pool Storage
1-in. Pb shielded cold box	1.5×10^4	1.4×10^4	1.4×10^4	1.4×10^4
2-in. Pb shielded cold box	6.7×10^2	1.2×10^2	1.3×10^2	1.0×10^2
3-in. Pb shielded cold box	6.1×10^2	6.2×10^1	7.6×10^1	4.3×10^1
Cold box system purged for maintenance	6.1×10^2	6.1×10^1	7.5×10^1	4.2×10^1

7.0 ⁸⁵Kr RECOVERY AND STORAGE FACILITY ACCIDENT ANALYSIS

Releases of radioactive material to the environment result from two types of events: 1) normal operational releases, and 2) releases resulting from accidents. Operational releases result from limited capability of the system to recover ⁸⁵Kr. For purposes of this study, a ⁸⁵Kr recovery efficiency of 95% was assumed. Accidental releases occur intermittently because of operational errors or system component and containment failures. Most accidental releases are small since the system is designed to capture these releases. Radiation doses may impact both the general public and the occupational work force.

7.1 POSTULATED ACCIDENT DESCRIPTIONS, FREQUENCIES AND SOURCE TERMS

The cryogenic krypton recovery system and the storage concepts considered in this report were examined for potential accidents. Table 7.1 summarizes the accidents (and source terms) which were postulated for these systems. The accident analysis methodology is discussed in detail in Appendix F. Included are the accident descriptions and a discussion of the supporting data which were available for this analysis.

7.2 PUBLIC EXPOSURE FROM ACCIDENTS

The equations used for calculating the 80-year cumulative world population dose from the accidental release of radioactivity to the environment are the same as those presented in Section D.1. However, the treatment of atmospheric dispersion factors for the maximum individual and 50-mile population doses differ. These values take into consideration the probability of occurrence yielding a mathematically expected dose.

TABLE 7.1. Postulated Accident Descriptions, Frequencies and Source Terms

Postulated Accident	Estimated Frequency of Occurrence Events/yr	Source Term		
		Release Point	Release Rate Ci/hr(a)	Release Period
1. Cryogenic Kr Recovery				
Minor				
1.1 Freezing	6	FRP Stack	2,400	8 hr
1.2 Plugged lines or system malfunction resulting in process gas routed to FRP stack	1	FRP Stack	2,400	48 hr
1.3 System leak to process cell atmosphere	0.5	FRP Stack	8,600, then 2,400	12 hr 36 hr
	0.5	FRP Stack	2,400	48 hr
	2	FRP Stack	2,400	12 hr
1.4 Loss of cooling supply to cryogenic separator	0.2	FRP Stack	2,400	24 hr
Moderate				
1.5 Extended process shutdown, with Kr venting	0.2	FRP Stack	2,400	30 days
1.6 Oxygen recombiner explosion; with or w/o building breach	0.1	FRP Stack	56 Ci	1.4 min
	0.02	Ground (restricted zone)	56 Ci	1.4 min
1.7 Cryogenic system or cell explosion with or w/o building breach	0.05	FRP Stack	245,000 then 2,400	1 hr 30 days
	0.005	Ground (restricted zone) FRP	245,000, then 2,400	4.6 min 30 days
2. Kr Gas Cylinder Filling, Transportation Interim Storage and Long Term Storage				
Minor				
2.1 Pinhole cylinder leak - indoors or outdoors	3	Storage Facility Stack	0.2	24 hr
	0.02	Ground (FRP Site)	2.0	24 hr
2.2 Direct exposure from Kr cylinder	2	(see Table 7.4)		
2.3 Loss of storage cell cooling	1	(no release - see accident description in Appendix F)		
Moderate				
2.4 Krypton cylinder ruptures in filling cell, hot cell or storage vault	0.05	FRP or Storage Facility Stack	2,400	50 hr
2.5 Krypton cylinder corrodes	0.05	FRP or Storage Facility Stack	2,400	25 hr
2.6 Moderate direct exposure from Kr cylinder due to mishandling	0.1	(see Table 7.4)		

(a) Or as stated.

TABLE 7.1. (cont'd)

Postulated Accident	Estimated Frequency of Occurrence Events/yr	Source Term		
		Release Point	Release Rate Ci/hr(a)	Release Period
<u>Severe</u>				
2.7 Cylinder ruptures indoors or in transit outdoors (operations personnel present).	0.02	Storage Facility Stack (1 worker within 10 ft of rupture-Table 7.4)	130,000 Ci	1/2 hr
	0.001	FRP site (1 worker within 50 ft-Table 7.4)	130,000 Ci	1.4 min
3. Immobilized Kr Loading, Transportation Interim Storage and Long Term Storage				
<u>Minor</u>				
3.1 Pinhole leak in Kr canister - indoors or in transit outdoors	1	Storage Facility Stack	0.2	24 hr
	0.01	Ground (FRP site)	2.0	2.5 hr
3.2 Direct exposure from Kr canister	2	(see Table 7.4)		
<u>Moderate</u>				
3.3 Kr canister rupture in loading cell or storage cell	0.05	Storage Facility Stack	5 Ci	2 min
3.4 Corrosion	0.05	Storage Facility Stack	5 Ci	2 min
3.5 Moderate direct exposure	0.5	(see Table 7.4)		
3.6 Canister rupture indoors or outdoors	0.1	Indoors (1 worker within 10 ft- Table 7.4)	5 Ci	2 min
	0.1	Outdoors (1 worker within 10 ft- Table 7.4)	5 Ci	2 min
<u>Severe</u>				
3.7 Kr/zeolite autoclave rupture	0.1	FRP Stack	2,400	5 days
	0.001	Ground (restricted zone)	288,000 Ci	4.7 min

(a) Or as stated.

7.2.1 Maximum Individual Doses

The atmospheric dispersion factors for the 80-year cumulative maximum individual dose calculations were taken from the Atomic Energy Commission Regulatory Guide 1.3 (AEC 1974). This methodology allows for up to five releases lasting more than one hour, the concentrations are integrated to yield total exposure (Streng, Soldat and Watson 1978). Regulatory Guide 1.3 employs a bivariate straight-line Gaussian expression for release of eight hours or less and a crosswind averaged expression for longer releases. This model assumes the plume travels in a straight line. Table 7.2 presents the atmospheric dispersion factors used in the accident analysis as a function of stack height and time of release for the maximum individual. Table 7.3 presents the 80-year cumulative radiation doses which are expected to result from these postulated accidents.

TABLE 7.2. Atmospheric Dispersion Factors for Maximum Individual Dose Calculations (AEC 1974)

<u>Stack Height</u>	<u>0 to 8 hr (sec/m³)</u>	<u>8 to 24 hr (sec/m³)</u>	<u>1 to 4 days (sec/m³)</u>	<u>4 to 30 days (sec/m³)</u>
110 m	7×10^{-6}	2.5×10^{-6}	8×10^{-7}	2.4×10^{-7}
50 m	4×10^{-5}	5×10^{-6}	2×10^{-6}	6.4×10^{-7}
G.L.	3.2×10^{-4}	6.6×10^{-5}	---	---

7.2.2 50-Mile Population Doses

The annual average atmospheric dispersion factors for all 160 sectors for each release height (ground-level, 50-m and 110-m) were multiplied by the corresponding population in each sector and then weighted for the annual wind frequency distribution. The atmospheric dispersion factor/population distribution values used in this study are 3.5×10^{-3} sec · man/m³ for the 110-m release height and 7.1×10^{-3} and 1.6×10^{-2} sec · man/m³ for the 50-m and ground-level releases, respectively. These factors were then used in Equation 4, Appendix D, to calculate the long-term cumulative 50-mile population dose.

TABLE 7.3. The 80-Year Cumulative Radiation Doses to the General Public from Accident Conditions

Accident Number	Maximum Individual (rem)			50-Mile Population Dose (man-rem)	World Population Dose (man-rem)
	GL	50 m	110 m		
1.1			1.5×10^{-2}	7.8	1.6×10^3
1.2			5×10^{-3}	7.8	1.6×10^3
1.3			1.6×10^{-2}	14.2	2.9×10^3
1.4			8.4×10^{-4}	0.77	1.6×10^2
1.5			2.6×10^{-3}	23.4	4.8×10^3
1.6	6.5×10^{-3}		7.1×10^{-7}	7.3×10^{-4}	9.4×10^{-2}
1.7	7.1×10^{-3}		2.2×10^{-3}	7.6	1.5×10^3
2.1	2.6×10^{-6}	4.3×10^{-6}		2.2×10^{-3}	2.1×10^{-1}
2.4		9.8×10^{-4}		0.80	83.5
2.5		8.8×10^{-4}		0.39	41.2
2.7	7.5×10^{-4}	1.9×10^{-3}		0.39	38.0
3.1	2.9×10^{-7}	1.4×10^{-6}		6.5×10^{-4}	6.7×10^{-2}
3.3		1.8×10^{-7}		3.3×10^{-5}	3.5×10^{-3}
3.4		1.8×10^{-7}		3.3×10^{-5}	3.5×10^{-3}
3.7	1.7×10^{-3}		6.9×10^{-4}	2.0	4.0×10^2
Total	1.4×10^{-2}	3.8×10^{-3}	4.2×10^{-2}	63.2	1.3×10^4
(85Kr Recovery + gas storage option)					
Total	1.5×10^{-2}	1.8×10^{-6}	4.2×10^{-2}	63.6	1.3×10^4
(85Kr Recovery + immobilized storage option)					

7.3 OCCUPATIONAL EXPOSURE FROM ACCIDENTS

Of the accidents considered in Table 7.1, only a few were recognized as significant possible contributors to occupational dose. These accidents have been evaluated and an estimate of the resulting doses are given in Table 7.4. It is obvious that even most of these doses are minimal.

TABLE 7.4. The 80-Year Cumulative Radiation
Doses to the Occupational Work
Force from Accident Conditions

<u>Accident Number</u>	<u>Occupational Dose (man-rem)</u>
2.2	2×10^2
2.6	8×10^1
2.7 (indoors)	8×10^{-2}
(outdoors)	9.3
3.2	1.6×10^{-5}
3.5	3.2×10^{-5}
3.6 (indoors)	3.4×10^{-3}
(outdoors)	<u>4.6×10^{-3}</u>
TOTAL	2.9×10^2

8.0 CONCLUSIONS AND RECOMMENDATIONS

Results of the comparison of the radiation dose commitments to the public and to the occupationally exposed work force from the two waste management alternatives are summarized in Table 8.1. The recovery case includes values for occupational exposure and the mathematical expectation of dose from the probabilistic treatment of accidents.

TABLE 8.1. Summary Table of 80-Year Cumulative Radiation Dose Commitments to the World Population

	<u>No Kr Recovery (man-rem)</u>	<u>Kr Recovery (man-rem)</u>
<u>Chronic Routine</u>		
Public Exposure	2.9×10^5	2.6×10^4
Occupational Exposure		3.7×10^5 to 4.2×10^1
SUBTOTAL	2.9×10^5	4.0×10^5 to 2.6×10^4
<u>Accidental Release</u> ^(a)		
Public Exposure		1.3×10^4
Occupational Exposure		2.9×10^2
TOTAL	2.9×10^5	4.1×10^5 to 3.9×10^4

(a) Mathematical expectation of dose from estimates of probability and extent of release of 15 accidents selected to bracket the spectrum of anticipated design-basis events.

The wide range of occupational doses from recovery operations result from the investigation of several design/operating options. The results indicate that some currently proposed options (e.g., maintenance of recovery system without purging ^{85}Kr and one inch of lead shielding on the cold box) may not be viable under the existing philosophy of maintaining radiation exposures as low as reasonably achievable. The results have also identified the equipment components and design features (e.g., cold box shielding and placement of chillers and blowers in storage facilities) that are of most concern for

radiation protection of workers. Further attempts to reduce occupational exposure would be of little value, for the recovery case. Almost 70% of even the lowest value in the range of dose is still estimated to be the result of continuous release of the nonrecoverable ^{85}Kr .

Results of Table 8.1 indicate that it makes little difference to the world population dose whether ^{85}Kr is captured and stored, or chronically released to the environment. These results clearly blur any mandate for ^{85}Kr recovery, i.e., the low dose calculated for the nonrecovery case coupled with only an order of magnitude difference between the two waste management techniques. The case for recovery is further diminished when it is remembered Table 8.1 includes comparisons of radiation exposure at very low dose rates to very large populations with those to a small number of occupationally exposed workers who each receive much higher dose rates.

Finally, the EPA (1976) reports that a cryogenic distillation facility for a 2100 MTHM/yr reprocessing plant could cost between \$16 million and \$20 million (1976 dollars) with annual operating and maintenance costs of \$300,000 to \$425,000. In addition, the capital cost for constructing a krypton storage facility has been estimated to be in excess of \$150 million with annual operating and maintenance costs of about \$900,000 (DOE 1979a). In 1980 dollars then, the construction of a krypton recovery and storage facility may cost \$200 million and cost well over \$1 million/yr to operate. These cost studies show that inordinate amounts of money may be required to lower the already insignificant 80-year cumulative dose to members of the world population of 0.05 mrem/person.

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APPENDIX A
ALTERNATIVE RECOVERY SYSTEMS

APPENDIX A

ALTERNATIVE RECOVERY SYSTEMS

While the technology exists for dispersal of ^{85}Kr to the atmosphere, methods have been proposed for its collection to avoid long-term global buildup. Of these, collection of ^{85}Kr by cryogenic distillation is considered to be currently available technology. Another method which could be used at a commercial reprocessing facility, but which requires further development, liquid fluorocarbon absorption (ERDA 1976). Additional collection techniques have been considered (Pence et al. 1978; Barton 1974; Keilholtz 1971; Slansky 1971; Nichols and Binford 1971, Rainey, Carter and Blumkin n.d.) but the application of many of these to the process flow stream anticipated at a commercial facility is questionable (ERDA 1976). A brief description of the two more promising methods is presented.

A.1 CRYOGENIC DISTILLATION - REFERENCE METHOD

Cryogenic separation provides an effective, continuous small-size system for the separation of krypton and xenon from the dissolver off-gas (DOG) system by utilizing the different boiling points of the main components. After various pretreatment steps to remove potentially hazardous or troublesome components, nitrogen oxides (to prevent solidification of NO_2), oxygen (forms radiolytic ozone), and nitrous oxide (prevents excessive freezing), the purified process gas is cooled and then enters the recovery column. Krypton and xenon are stripped from the gas stream by a counter current flow of liquid nitrogen. The concentrated Kr and Xe proceed to the separation column, which operates in two stages. Krypton and xenon are collected continuously in the first stage while light gases, mainly nitrogen and argon, are recycled in the holding tank. The second stage is operated periodically by shutting the feed stream to the separation column, and by fractionating the contents to yield krypton at the top and xenon at the bottom.

Cryogenic distillation has been intermittently operated at the Idaho Chemical Processing Plant (ICPP) since 1958 for research and industrial applications (Bendixsen and Offutt 1969). Although the ICPP unit was not designed to achieve the high ^{85}Kr and continuous recovery efficiencies necessary for environmental control, its operation has indicated such potential. The cryogenic system itself is expected to exhibit a decontamination factor (dF) of at least 1000 (EPA 1976). However, the overall efficiency for removal of krypton from the plant is expected to be lower because of potential leakage through the system during maintenance, startup and shutdown operations. Therefore, an effective plant dF of between 10 and 100 (release fraction of 0.1 to 0.01) has been projected for routine operation of such a system (EPA 1977). One vendor warrants its product for a krypton dF of 10,000 (Airco 1974).

Ademonstration system on the scale of a commercial facility is required to establish the practical limits for ^{85}Kr recovery from commercial fuel reprocessing facilities. A serious concern in this process, particularly when applied to an FRP, is the explosion hazard that results from the presence of ozone or mixtures of liquid oxygen with hydrocarbons and nitrogen oxides.

Exxon Nuclear Company submitted an application to the NRC for a construction permit to build the Nuclear Fuel Recovery and Recycling Center (Exxon Nuclear 1976). This plant is designed to include a cryogenic distillation system for the recovery of ^{85}Kr .

A.1.1 Process and Facilities

The cryogenic distillation rare gas recovery system is designed to be an integral part of the 2,000 MT/yr fuel reprocessing plant. The two facilities share utilities, laboratories, health physics support, operating personnel, change areas, maintenance areas, warehousing, shops and offices. The off-gas treatment system has sufficient capacity to handle the total dissolver off-gas (DOG) stream from the reprocessing plant. A floor plan of the facility is given in Figure A.1 and an elevation view is given in Figure A.2 (DOE 1979a). All normal process operations are handled in the hot cell, and workers do not normally come in contact with the ^{85}Kr .

FIGURE A.1. Dissolver Off-Gas Krypton Recovery System, General Plan

FIGURE A.2. Dissolver Off-Gas Krypton Recovery System,
(See Figure A.1 for key to circled numbers.)

The reference design of the treatment facility is a modification of that described in detail by Brown, Knecht and Thomas (1978) and the Draft Commercial Waste Management Statement (1979), and briefly summarized here. The cryogenic unit includes a precooler, and absorption-stripping column and a fractionation still. Krypton and xenon are condensed in liquid nitrogen in the adsorption column. The liquid is transferred to the fractionation still and the nitrogen, krypton and xenon are separated by fractional distillation. The product which boils off is a mixture of 75% Kr and 25% Xe. The design is for 90% recovery of the ^{85}Kr inventory. We have assumed 95% recovery for this analysis. The overall krypton recovery system is shown in Figure A.3 (Brown, Knecht and Thomas 1978).

All volatile fission products other than ^{85}Kr have been removed from the gas before it reaches the rare gas plant. However, ^{85}Kr is present in large enough quantities to present a radioactive hazard throughout the entire plant. Therefore the design of the facilities must take this into account.

Radioactivity of some process streams rises rapidly in the cryogenic section of the process and the final krypton product available for disposal will be in excess of 230 Ci/l at 2.7 atm.

The oxygen recombiner vessel consists of a duplicated vertical stainless steel vessel (approximately 1 m diameter and 3 m high). Each recombiner vessel is provided with an explosion disc and relief piping. Each vessel will be enclosed in a three-sided concrete barrier, with the open fourth side directed toward a restricted area outside breakway panels in the building. The safety of this system is explored in greater detail in Appendix F.

The recycle gas cooler which controls the temperature in the recombiner will be a conventional shell-and-tube heat exchanger of 347 stainless steel construction. Oxygen-free gas leaves the recombiner circuit, is cooled and part of the water of reaction removed by condensation. The process gas leaving the primary condenser is saturated. To reduce the water load on the dryers, a secondary refrigerated condenser is provided. The process gas to the compressors is ready for final drying before entering the cryogenic section of the process. All cryogenic equipment, piping, and accessories are

PROC GAS FEED	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16
H ₂ Kg/HR	171	--	--	172	--	--	172	172	162	55	53	2.3	9.1	--	--	--
O ₂	50.8	--	52.5	*TR	--	--	TR	TR								
NO _x	TR	--	--	TR	--	--	TR	TR								
N ₂ O	0.28	--	--	TR	--	--	TR	TR								
H ₂	TR	6.6	--	0.12	--	--	0.12	0.12								
H ₂ O	TR	--	--	57.7	50.3	6.7	0.7	0.009	--	--	--					
Kr	0.09	--	--	0.09	--	--	0.09	0.09	--	--	--	--	0.09	0.09	0.089	TR
Xe	1.3	--	--	1.3	--	--	1.3	1.3	--	--	--	--	1.3	1.3	TR	1.3
Ar	2.8	--	--	2.8	--	--	2.8	2.8	2.8 [‡]							

* TR = TRACE
[‡] FINAL DISTRIBUTION OF AR IN COLUMN MAY NOT BE AS SHOWN. DATA NEEDED.

FIGURE A.3 Krypton Recovery System

located in a heavily insulated and shielded, self-contained structure, commonly called a cold box. The cold box has inside dimensions 1.2 x 1.2 x 3.7 m, with a volume of 5.33 m³. Internal temperatures will be about -170°C during operations.

In this concept, krypton product will be collected as a liquid.

A.1.2 Shielding and Special Handling

There are several operations within the facility which require special treatment to protect workers from potential hazards.

The process gas being treated has an activity of about 13 Ci/m³^(a) before ⁸⁵Kr is concentrated in the cryogenic section of the plant. The cryogenic distillation hardware can be adequately shielded by 15-cm concrete walls, except for the cold box and the hot cell which would require about 3 ft of concrete and limited personnel access to such equipment is possible.

Radioactivity increases as process streams are enriched in krypton, leading to an activity of the liquid product of about 27 Ci/g. One day's production, if stored in a receiver, represents a source of about 47 kCi in a volume of 0.78 ℓ of liquid.

If pressurized cylinder packaging is used (Appendix B) almost four day's production are required to fill one 50-ℓ cylinder. This liquid will be stored until heated for cylinder loading. This source requires shielding inside the cold box shield; approximately 5 cm of lead. The overall cold box krypton inventory (exclusive of the above-mentioned) is estimated to be 50-75 kCi, located almost entirely in the column reboilers and final product column. Shielding of these sources requires about 10 cm lead or its equivalent.

Pressurized cylinders will be filled outside of the process cold box, but within a hot cell, through shielded lines by warming the krypton receiver to approximately -84°C. The product cylinder, in its transport shield will be positioned at the hot cell filling station and connected to the filling line.

(a) This value is a ratio of fuel burnup from ORNL-4451 and ICP-1126.

Line and cylinder will be evacuated before filling; the filled cylinder will then be isolated and the fill line evacuated before removing the cylinder.

Concrete wall thicknesses for the facility are shown in Figure A.4 (Brown, Knecht and Thomas 1978).

A.1.3 Operation and Maintenance

This cryogenic distillation facility is expected to operate 24 hours a day, 7 days a week for 300 days each year. Normal activities include monitoring of the NO_x , H_2O , CO_2 , and krypton, maintenance and replacement.

All normal process operations are handled remotely in the hot cell. Compressors and pumps are located in normally unoccupied galleries. Personnel enter the gallery only to install or remove equipment. Where infrequent operations, instrument readings and some maintenance activities are needed, personnel may be exposed to higher radiation dose rates. In such cases the dose is controlled and limited to less than 1 mrem/hr.

A.1.4 Personnel

Estimates for staffing the krypton recovery system are taken from the DOE (1979a) and presented in Table A.1.

TABLE A.1. Cryogenic Distillation System Staffing Requirements

<u>Job Classification</u>	<u>Personnel Required, man-yr/yr</u>
Operators	5
Radiation Monitors	1
Maintenance Craftsmen	2

A.2 LIQUID FLUOROCARBON ABSORPTION

While selective absorption by liquid fluorocarbons has been offered commercially for removing krypton from the off gas of LWRs (ERDA 1976), the technology has not yet been demonstrated for removing krypton from a commercial reprocessing plant.

FLOOR PLAN - EL.0

FIGURE A.4. Dissolver Off-Gas Process Building

Typical off-gas impurities such as nitrogen oxides, carbon dioxide, water, iodine, and methyl iodide have caused problems with krypton removal utilizing this process (ERDA 1976). On the other hand, Stephenson et al. (1976) report that the fluorocarbon-based process has a remarkable tolerance for typical feed gas impurities. Many process solvents have been considered and listed by Stephenson et al. (1977) such as Freon-11, carbon tetrachloride, kerosene-base liquids, liquid carbon dioxide, dichlorodifluoromethane, nitrous oxide, and other organic solvents. Based on its properties for capacity of krypton, separation factors, impurity tolerances, thermal radiation stability and overall process safety and economic features, dichlorodifluoromethane (Freon-12) demonstrated the most overall promise. This process may also be equally effective in removing iodine, methyl iodide, and ^{14}C as carbon dioxide (Stephenson 1976).

Figure A.5 is a schematic of the proposed fluorocarbon-based selective absorption process for removing krypton from the offgas of an LWR fuel reprocessing plant. ERDA (1976) describes the process as follows: Three packed columns comprise the main working sections of the process. Each column is designed to exploit certain gas-liquid solubility differences that exist between the solvent and the various gas constituents that might be present. The main separation of the noble gases is accomplished in the absorber. The other two columns, each fitted with a reboiler and an overhead condenser, comprise the intermediate and final stripper sections of the plant. The final stripper produces a highly concentrated krypton gas product suitable for long-term storage.

Pilot plant-scale fluorocarbon processes have been in developmental stages at the ORGDP since 1968 (ERDA 1976). Early efforts were directed toward LWR applications demonstrating krypton recovery efficiencies as high as 99.9% (ERDA 1976), and xenon recovery efficiencies of 99.99% (Bray et al. 1977). Total vented gas in this case contained about 0.1% of its input activity, or a dF of 1,000 (Bray et al. 1977). An LMFBR fuel reprocessing pilot plant process was built at Oak Ridge and put into operation in 1975.

FIGURE A.5. Process for the Collection of ^{85}Kr by Liquid Fluorocarbon Absorption (ERDA 1976)

Tests have shown that a fluorocarbon process designed to remove 99% of the off-gas krypton can also be relied upon to remove at least that much of the off-gas ^{14}C if present as carbon dioxide (ERDA 1976).

Stephenson and Eby (1976) reported on the application of the refrigerant-12 off-gas removal technique to a reprocessing plant indicates that CO_2 removals were higher, in tests, than those for xenon. The system is versatile, continuous, and adaptable to scaleup. It also appears to be much less susceptible to fire and explosion than cryogenic distillation.

Primary questions concerning fluorocarbon absorption relate to the tolerance of the system to contaminants in the off-gas streams, the effects of

radiation on the solvent, and corrosion problems that may result from the evolution of fluorine and chlorine. Presently krypton and xenon are not separated, but the separation process is under development.

Off-gas flow rates at a current LWR would probably be compatible with the size of the system currently being tested, while both BWR's and fuel reprocessing plants would require additional analysis due to much greater off-gas flows (Bray et al. 1977).

Allied-General Nuclear Services' Barnwell reprocessing plant employed the fluorocarbon-absorption process in its licensing application. The process selection came from a study they funded at ORNL (Murbach, Carr and Gray n.d.).

APPENDIX B
ALTERNATIVE PACKAGING DESIGNS

APPENDIX B

ALTERNATIVE PACKAGING DESIGNS

After ^{85}Kr has been recovered from the reprocessing facility off-gas system, it can be stored as a gas in pressurized cylinders or immobilized prior to storage. To prevent accidental dispersal of the ^{85}Kr during handling, shipping, and storage, immobilization would provide an additional barrier for environmental release. The immobilization material must be compatible with long-term storage containers. Many of the techniques for capture of ^{85}Kr provide an adequate immobilization method. Only minor development of physical methods for containment and transportation of mixed wastes would be necessary (ERDA 1976).

Storage of ^{85}Kr in pressurized cylinders is the only commercially available technology with its costs high, and its contents mobile. Studies are underway to investigate the suitability of cylinders for this 50-year concept. Several alternative methods of immobilizing ^{85}Kr have been reported (Knecht 1977; Benedict 1977; and Tingey et al. 1979) with the expected result of a less expensive long-term storage with improved safety.

B.1 HIGH PRESSURE STEEL CYLINDERS

The use of high pressure stainless steel cylinders is the only method immediately available for the commercial storage of fission gases. High pressure steel cylinders have been used for the storage and handling of industrial gases for over 50 years. The Idaho Chemical Processing Plant at the Idaho National Engineering Laboratory has used cylinders for temporary storage and transportation of gas mixtures containing ^{85}Kr for over 20 years (Foster and Pence 1975).

Several million industrial gas cylinders are filled and transported each year. Highly toxic gases shipped in this manner include phosgene, arsine, and diborane (Foster and Pence 1975). Hazardous gases require certain shipping and storage precautions such as use of nonventing cylinders; i.e.,

having no pressure relief mechanism, packless valves with sealed valve caps, and limits on filling to only 25 to 50% of the normal working pressure of the cylinder. In addition, ^{85}Kr cylinders would require additional shielding for personnel protection and adequate cooling methods. A shipping cask design for ^{85}Kr cylinders has been described by Blomeke and Perona (1969).

B.1.1 Advantages of Using Cylinders for Storage

The advantages of using cylinders for storage are:

- well-established history of development and use in industry
- long life with good thermal and radiation resistance
- high capacity
- low cost
- ease of future recovery.

Longevity of Steel Cylinders

A study made by Union Carbide of several thousand cylinders suggested that a useful life of at least 500 years could be expected, (Foster and Pence 1975) assuming no continued abuse, excessive pressure or contact with corrosives.

Cylinder Capacity

Greater than 99% of the decay energy of ^{85}Kr is dissipated in the form of a 0.69 MeV beta particle (0.246 MeV average energy). Most of this energy will appear as heat within the cylinder walls and interior resulting in an external wall temperature of about 60°C. Heat transfer calculations by Foster and Pence (1975) based on natural convection cooling resulted in the estimated capacities of the 50-l cylinders shown in Table B.1. The reprocessing of one tonne of LWR fuel (33,000 MWd/MT) results in the release of 11,200 Ci of ^{85}Kr (ORNL 1970a). Several ^{85}Kr production numbers appear in the literature as documented in Section 4.3.

TABLE B.1. Storage Conditions for High Pressure Steel Cylinders
Containing 6% ^{85}Kr in Kr (Foster and Pence 1975)

<u>Cylinder Pressure (psi)</u>	<u>Wall Temperature ($^{\circ}\text{C}$)</u>	<u>Heat Generation (cal/sec)</u>	<u>Radioactivity Content (Ci)</u>
500	60	44.5	128,000
2,000	127	146.0	419,000

A 2000 MT/yr plant would recover 21 MCi/yr, at 100% recovery. The annual cylinder requirements for this reprocessing plant are given in Table B.2.

TABLE B.2. Annual Cylinder Requirements for a 2,000 MT/yr
Reprocessing Plant

<u>Cylinder Pressure (psi)</u>	<u>Radioactivity Content (Ci)</u>	<u>Number of Cylinders/year</u>
500	128,000	164
2,000	419,000	50

Cylinder filling equipment will have to be remotely operated, leakproof and monitored continuously. This equipment will be located in a rare gas recovery facility.

Costs

Labor costs for the operation of the cylinder filling equipment will be low, since no full-time personnel would be needed. The major monetary costs associated with the process would be for cylinders and cylinder storage facilities.

Future Recovery

The pressurized gas cylinder storage scenario relies upon the ability to recover 50-year old cylinders for the intentional release of the remaining 5,100 curies of ^{85}Kr , about 4% of the original activity.

B.1.2 Disadvantages of Using Cylinders for Storage

The disadvantages of using pressurized cylinders for storage are:

- potential for sudden failure of a cylinder resulting in the release of its contents
- requirements on the purity of the gas
- potential problems arising from the presence of the decay product rubidium.
- temperature increase from decay heat not only increases gas pressure, but may reduce the cylinder wall strength, resulting in possible cylinder failure (Christensen 1977). This disadvantage becomes academic at cylinder loadings of 500 psi.

Cylinder Failure

There is no reason to expect the spontaneous failure of a cylinder except in the case of operator error from overfilling or from abuse such as overheating, impact, or contact with corrosives. Contaminants such as O_2 , NO_x , and water will be removed from the gas before storage to preclude the possibility of internal corrosion, from these sources, which could result in cylinder failure. Proper handling and storage would minimize the possibility of such occurrences, but the possibility of a release must still be considered. This concept is discussed in greater detail in the accident analysis (Appendix F).

The possibility of a release must be considered at the cylinder filling facility of the reprocessing facility, at the storage facility, and during transportation between the two.

The cylinder filling facility at the reprocessing plant would be contained and remotely operated and monitored so that minimal environmental release or personnel exposure would result from the release of a cylinder inside the facility. The ventilation air from the filling facility could be run back to the rare gas recovery system along with the regular process off-gas streams to insure a negligible release to the environment. A release of the cylinder contents at the storage facility could be a potential problem because air

flow must be maintained for cooling of the cylinders. If storage facilities are not co-located with rare gas recovery facilities, they may be required to install their own. Releases of ^{85}Kr during transportation would have to be contained by the shipping cask. While the probability of such a release is very small, it must be considered and planned for.

Purity Requirements

External as well as internal corrosion could occur if certain contaminants are stored with the krypton. Ozone, NO_x and water are of principal concern and are therefore removed from the krypton before storage. It is known that a radiation field will catalyze the formation of ozone and NO_x if there is any oxygen and nitrogen present. (Foster and Pence 1975). Water and NO_2 present clogging problems at cryogenic temperatures. Thus low temperature separation systems might require the removal of the same impurities that are a problem in long-term cylinder storage.

Foster and Pence (1975) reported that cylinder valve seats made of a resin bonded cloth laminate (micarta) have undergone severe degradation from the radiation, possibly in conjunction with corrosives, that resulted in a hole being formed in the seat. The displaced material collected in the orifice near the seat and sealed it off. This problem has been eliminated by the use of stainless steel valve seats.

Potential Problems Arising from the Presence of the Decay Product - Rubidium

Rubidium is the daughter of ^{85}Kr and therefore constantly builds up in the pressurized cylinder as the ^{85}Kr decays. After a 50-year storage period, the ^{85}Kr activity in a 500-psi cylinder is about 5,100 curies and the Rb about 314 g (Foster and Pence 1975).

Rubidium is an alkali metal and has chemical properties similar to sodium, potassium and cesium. It will react vigorously with water to produce hydrogen. When exposed to air, it will ignite spontaneously. As long as the cylinder remains intact, rubidium should remain a liquid for many years. It is difficult to predict the long-term storage effect that rubidium would have

on the stainless steel cylinder. Eleven candidate materials have been screened for potential application in the ^{85}Kr storage development program at INEL (Pinchback 1979). While 316 stainless steel and other materials were chosen for further evaluation, 304 stainless showed corrosion damage in the forms of external corrosion scale and intergranular penetration.

B.2 IMMOBILIZATION--HIGH TEMPERATURE/PRESSURE SORPTION AND ION IMPLANTATION/SPUTTERING

High temperature and high pressure sorption by zeolites and ion implantation/sputter deposition of crystalline or amorphous solids appear to be the most promising methods of immobilizing ^{85}Kr prior to long-term storage, based on existing knowledge of loading, leakage, and solid product properties.

Allied Chemical and Battelle, Pacific Northwest Laboratories (PNL) have done a considerable amount of work in this area. Knecht (1977) reviewed the literature and reports on Allied's work in presenting three methods of immobilizing ^{85}Kr in solids. The methods of immobilization include: 1) high temperature/pressure sorption in zeolites, glass, metal and other solids; 2) ion implantation/sputtering in solids; and 3) low temperature metal vapor deposition. Knecht concludes from his evaluation that sorption by zeolites and ion implantation/sputtering by crystalline or amorphous metals appear to be the most promising methods of immobilizing ^{85}Kr prior to long-term storage.

B.2.1 High Temperature/Pressure Sorption

In high temperature/high pressure sorption, krypton is imbedded by diffusing into a solid under a concentration gradient. These solids may be zeolites, metals, charcoal or glass.

Zeolite materials are crystalline aluminosilicates which contain a regular array of pores of uniform size (Breck 1974). For a given zeolite type, the pore dimensions are uniquely determined by the structure of the crystal. Figure B.1 shows the structure of a group of sodalite zeolite cages, each containing one atom of krypton.

FIGURE B.1. Representation of Sodalite Crystals Containing Krypton Atoms (ERDA 1976)

Each sodalite cage consists of a truncated octahedron ($\sim 6.6 \text{ \AA}$ free diameter). Each six-ring face is shared with adjoining sodalite cages to make up the overall crystal structure. The krypton atom shown occupying each cage has a room-temperature gas-kinetic diameter of $\sim 3.5 \text{ \AA}$.

The process by which krypton diffuses through such a pore structure is known as activated diffusion, since an activation energy is required for krypton atoms to pass through the $2.3 \text{ \AA}$ apertures (Breck 1974). At high temperature and pressure (450°C ; $1,000 \text{ atm}$), there is sufficient energy for krypton to diffuse into the crystalline cages, while at low temperatures ($<150^\circ\text{C}$) the encapsulated krypton does not have enough energy to diffuse out at appreciable rates (Barrer and Vaughan 1971).

Figure B.2 shows the calculated release of encapsulated krypton from sodalite (Knecht 1977). The " ^{85}Kr decay" line shows the fractional amount of ^{85}Kr remaining at any time in a storage cylinder containing either pressurized or encapsulated ^{85}Kr . The " ^{85}Kr " curve gives the fraction of ^{85}Kr which has leaked out of sodalite at any time, but is still contained in the gas phase of the storage cylinder.

For sodalite, the fraction of ^{85}Kr originally stored which could be released at any time due to rupture of a storage container can be determined

FIGURE B.2. Calculated Release of Original Krypton Inventory from Sodalite at 150°C as a Function of Time (Knecht 1977)

from the ^{85}Kr curve of Figure B.2. For example, at about eight years, the maximum net leakage is about 0.3% and the predicted leakage potential of a damaged storage container represents a safety factor (based on potential losses from a damaged storage vessel) of about 200 (Knecht 1977) for sodalite encapsulation relative to pressurized tank storage (Barrer and Vaughan (1971)).

The predicted net leakage of ^{85}Kr from zeolite 3A was about 35% at eight years for a storage temperature of 50°C (Brown, Hoza and Knecht 1977). Although K-, Cs-, and Rb-exchanged zeolite A samples have larger sorption capacity for krypton than does sodalite, the leakage from K-exchanged zeolite A (zeolite 3A) is also greater.

The Cs- and Rb-exchanged forms may have lower leakage rates due to the smaller cage openings. Experimental studies of these samples are in progress.

A typical process flowsheet for zeolite encapsulation is shown in Figure B.3. Activated zeolite (with interstitial water removed) is loaded into the pressure vessel and heated to the encapsulation temperature. Krypton containing ^{85}Kr is introduced and pressurized to the encapsulation pressure until the pores of the zeolite are filled. The temperature is then lowered, and the pressure and excess krypton is released back to the storage cylinder. The zeolite--encapsulated-- ^{85}Kr is then removed and placed in the storage container for long-term storage.

FIGURE B.3. Process for High Pressure Encapsulation of ^{85}Kr in Zeolite (ERDA 1976)

Long-term storage criteria need to be developed. Stored ^{85}Kr in zeolite will generate heat, which in turn must be dissipated so that the storage temperature does not exceed a value at which appreciable diffusion of ^{85}Kr out of the zeolite occurs. Long-term radiation damage to the zeolite structure must be determined and minimized. The effect of the ^{85}Kr decay product, rubidium, must also be determined.

Another alternative technique for storing radioactive krypton in a solid matrix is the dissolution of krypton in low-density glass followed by densification at high temperatures. Research along these lines is underway at PNL.

Early attempts to dissolve krypton in a glass matrix confirmed the reported low solubility of inert gases in glass. Therefore, to circumvent this problem, krypton was loaded into porous (low-density) glass rods which were then densified by heating to sintering temperatures while retaining the krypton atmosphere. It is expected that low-density glass may retain well over 99% of the krypton charged even at ambient temperatures. Release rates of krypton gas as a function of temperature elevation is also under investigation at PNL.

B.2.2 Ion Implantation/Sputtering

An alternative method of imbedding krypton involves the penetration and retention of krypton ions accelerated into a growing sputter deposited film. Several matrix materials have been studied and summarized by Tingey et al. (1979). One major advantage of this method over encapsulation into sodalites is that it can be achieved at low temperature and low pressure (Bayne et al. 1978).

The brief description of this method presented here is taken from Tingey et al. (1979) and shown in Figure B.4. Gases can be implanted into solids by bombarding their surfaces with energetic ions. To produce large quantities of a solid containing high concentrations of Kr, it is necessary to perform the implantation during film growth by a vapor deposition process such as sputtering. Tingey et al. (1979) have been conducting sputtering experiments at PNL for some time and report that sputtering of solids appears advantageous to other methods for gas incorporation where the sputtering ion is generated from the gas which is to be implanted into the solid.

FIGURE B.4. Thermionically Supported Plasma Sputtering/Ion Implantation Components (Tingey et al. 1979)

Leakage rates of krypton implanted in solids depend on four diffusion mechanisms discussed by Knecht (1977). Low temperature leakage can be minimized by avoiding structural damage during ion implantation.

Krypton has been loaded into several candidate metals by sputtering with the krypton ions. From this work, several alloys appear to be feasible. Excellent gas retention has been observed. Higher loadings are achieved in metals with a glassy structure than those with crystalline structures (McClanahan and Bayne 1977). The $\text{Fe}_{0.85} \text{Y}_{0.13} \text{Me}_{0.02}^{(a)}$ alloy appears to be

(a) Me = minor impurity.

the best candidate based on gas uptake which results in a final matrix of $\text{Fe}_{0.79} \text{Y}_{0.12} \text{Kr}_{0.09}$, although $\text{Fe}_{0.75} \text{Zr}_{0.19} \text{Kr}_{0.06}$ appears to be a promising candidate because of its continued retention of krypton at high temperatures. Other amorphous metals have been studied.

This research effort covering many different potential trapping materials has confirmed the observations of others that amorphous metal substrates are desirable for their ability to effectively trap inert gas ions. Krypton concentrations of 5 to 12 atom percent Kr have been trapped in sputtered amorphous metals.

The long-term release of ^{85}Kr from two amorphous metal deposits was also investigated as a function of temperature by Tingey et al. (1979) as shown in Table B.3. These values were extrapolated from short-term, high-temperature measurements.

TABLE B.3. Fraction of ^{85}Kr Released in 100 Years

	<u>200°C</u>	<u>300°C</u>	<u>400°C</u>
$\text{Fe}_{0.79} \text{Y}_{0.12} \text{Kr}_{0.09}$	1.1×10^{-5}	2.4×10^{-2}	8.5×10^{-1}
$\text{Fe}_{0.75} \text{Zr}_{0.19} \text{Kr}_{0.06}$	7.5×10^{-7}	6.6×10^{-4}	7.1×10^{-2}

Although long-term tests are needed to confirm these results, it appears loss of ^{85}Kr will be negligible at temperatures below 300°C. This data also suggests that packaging this material in additional metal containers may not be necessary.

These release rates are so low that storage of ^{85}Kr immobilized in sputtered metals can be accomplished without any significant environmental contamination.

B.3 IMMOBILIZATION OF ^{85}Kr -LOADED SOLID GRANULES

Benedict (1977) reviewed seven methods of immobilizing solid granules using: 1) glass matrices, 2) metal matrices, 3) polymer matrices, 4) pressed (isostatic) matrices, 5) cement blocks, 6) bitumen blocks, and 7) porous metal

containers. He concludes that of the seven, a metal sintering process to form a calcine and metal matrix best fulfills storage requirements. Although mixtures of solids containing krypton and metal powders have not been tested, present information indicates that this process will work.

Another feasible process, the formation of matrices by isostatic pressing, is compatible with many different materials, and a satisfactory product can be formed. If the complex processing equipment can be scaled up to handle large amounts of radioactive wastes, this process could produce a matrix which meets potential storage requirements (Benedict 1977).

APPENDIX C

^{85}Kr DOSIMETRY

APPENDIX C

⁸⁵Kr DOSIMETRY

The dosimetry of ⁸⁵Kr was discussed at length during the Nobel Gas Symposium held in Las Vegas in 1973.

Snyder (1973) demonstrated the complete dosimetry of the noble gases including external dose to the skin from both beta and gamma radiation, external dose to the total-body, gonads and internal organs, dose to the lung from inhaled radioisotopes and dose to the body from noble gases dissolved in the bloodstream and then absorbed into various tissues. Authorities on this subject, including Snyder, agree that the dose from noble gases absorbed in tissue is generally small compared to the dose from direct external radiation and from inhalation (Soldat et al. 1976). Therefore, this analysis will concentrate on the calculation of external and inhalation radiation doses.

External doses resulting from submersion in air are usually calculated assuming that the plume is "infinite" in volume relative to the range of the emitted radiations. Utilizing this assumption the energy absorbed per gram of material is equivalent to the energy emitted per gram. It follows then to convert the average energy per disintegration to dose and to correct for differences in energy absorption between air and tissue and for the physical geometry of the specific exposure. For a person standing on the ground immersed in a large hemisphere of radioactive gas, the geometry for gamma radiation is obviously 2π . For beta radiation with its much shorter range in air, the geometry approaches 4π or infinite. However, since the beta is of limited penetrating power, it will only irradiate the skin from one side, thereby again reducing the geometry back to 2π . Minor exceptions can occur in thin membranes, such as a protruding ear where the geometry approaches 4π .

In some situations when the radiation dose is required for a person very close to the elevated release point, the plume is overhead and "finite." In this situation, the dose calculation becomes more complex requiring the numerical integration of the dose contributions for each segment of the finite

cloud. Such a specialized situation is assumed to contribute an insignificant dose when compared to the calculation of the world population dose considered here. Therefore, the "infinite" cloud was chosen as the model for this study.

The skin dose was calculated at a depth of 7×10^{-3} cm (7 mg/cm²) and the total-body dose at a depth of 5 cm as suggested by the NCRP (NBS-Handbook 85, 1964). Therefore, the radiation dose a man receives while immersed in air containing radioactive krypton is an external dose to both the skin, from the beta and gamma radiation, and the total-body and other internal organs due to the gamma radiation only. The dose to the male gonads was calculated at a tissue depth of 1 cm. Schaeffer's (1976) calculations indicate that beta rays from ⁸⁵Kr are not energetic enough to penetrate through the 1 cm of tissue assumed to be over the male gonad. Therefore, the dose to the male gonad is due to the ⁸⁵Kr gamma. The total-body dose can be used as an upper bound to the female gonadal dose from external radiation (Soldat et al. 1976).

The equations used for calculating the doses from krypton exposure are found in Appendix D.

For ⁸⁵Kr with relatively little penetrating (gamma) radiation, the critical organ will be the skin even considering its less restrictive dose standard. However, where collective doses are calculated, the total-body dose is preferred to enable the estimation of health effects if desired.

APPENDIX D

EQUATIONS FOR CALCULATING DOSES FROM ^{85}Kr EXPOSURE

TABLE D.2. Comparison of Inhalation Dose Factors for ^{85}Kr and the Lung (rem/yr per pCi/m³)

DCF (Lung)	Reference
1.7E-8	Soldat et al. 1976
5.1E-8	Schaeffer 1973
1.4E-8	Whitton 1968
2.6E-8	Russell and Galpin 1972
1.8E-8	Snyder 1973

A constant world population of 6.4 billion persons was assumed. This is the value reported by Killough (n.d.) for the year 2,000 based on a United Nations projection. This is also in good agreement with the value of 6.3 billion people derived from the EPA (Fowler et al. 1976) who based their projections on a 1970 world population of 3.6 billion people and an annual growth rate of 1.9%.

D.1.2 50-Mile Population Dose

The dose to the 50-mile population is the annual dose multiplied by 30 years (Equation 4) plus the additional minute contribution to an individual from the dilute concentration of ^{85}Kr circling the world.

The latter is taken into account by multiplying the results of Equation 3 by the 50-mile population (1.96×10^6 persons, Table D.3) and then adding it to the 30 year dose (see Equation 5).

$$\text{Dose}_{30} = \frac{(\text{Ci/yr}) \sum (\bar{X}/Q')(POP) \frac{\text{sec} \cdot \text{man}}{\text{m}^3}}{3.15 \times 10^7 \text{ sec/yr}} (10^{12} \text{ pCi/Ci})(Df \frac{\text{rem}}{\text{yr}} \text{ per } \frac{\text{pCi}}{\text{m}^3})(30 \text{ yr}) \quad (4)$$

where: $\sum[(\bar{X}/Q')(POP)]$ is the integrated population times annual average atmospheric dispersion factor,
Df is the dose factor,
30 is the operational life time of the FRP

TABLE D.3. 1970 Census Population in Reference Environment (CWM GEIS)

Sector	Miles										Totals
	0.5-1	1-2	2-3	3-4	4-5	5-10	10-20	20-30	30-40	40-50	
N	0	4	4	18	160	210	1,115	3,641	2,137	1,209	8,498
NNE	0	4	4	14	26	157	986	3,350	4,185	1,872	10,598
NE	0	6	18	72	109	232	1,306	4,897	2,848	6,371	15,859
ENE	0	4	12	72	145	333	2,025	2,677	8,743	6,209	20,220
E	0	4	12	145	537	993	1,321	9,094	6,344	14,195	32,645
ESE	0	4	20	353	118	610	3,400	50,482	123,104	163,155	341,246
SE	0	25	245	1,069	194	632	5,063	46,789	581,389	579,114	1,214,520
SSE	0	4	18	45	157	374	3,466	18,642	59,435	32,445	114,586
S	0	4	41	67	112	1,097	5,438	5,844	10,131	7,334	30,068
SSW	4	15	26	67	126	571	3,177	4,809	6,411	7,317	22,523
SW	0	30	65	58	50	423	1,835	4,656	6,106	6,856	20,079
WSW	0	6	55	93	65	414	3,007	1,901	7,515	4,442	17,498
W	0	9	31	78	73	379	1,730	3,600	3,326	4,805	14,031
WNW	4	8	8	44	29	332	1,662	6,495	6,493	5,984	21,059
NW	0	6	9	21	44	293	5,277	47,196	4,061	4,501	61,408
NNW	0	8	15	55	181	165	1,204	2,753	2,480	4,533	11,394
TOTALS	8	141	583	2,271	2,126	7,215	42,012	216,826	834,708	850,342	1,956,232

$$\text{Dose}_{80} = [\text{Dose}]_{30} + [\text{World Dose}]_{80} \quad (5)$$

D.1.3 Maximum Individual Dose

The maximum individual dose assumes that an individual continuously resides at the point of highest ground level concentration (Table D.4) of ^{85}Kr released from the FRP stack. Depending upon the release height, terrain and meteorology, this point is generally within a few miles of the plant. Equation 6 is the annual dose equation multiplied by 30 years to obtain a 30-year cumulative dose.

$$\text{Dose}_{30} = \frac{(\text{Ci/yr})(\bar{X}/Q' \text{ sec/m}^3)(10^{12} \text{ pCi/Ci})}{3.15 \times 10^7 \text{ sec/yr}} \quad (\text{Df rem/yr per pCi/m}^3)(30 \text{ yr}) \quad (6)$$

Upon plant shutdown the individual will continue to be exposed to ^{85}Kr which has become evenly distributed throughout the earth's atmosphere. Therefore, to allow comparison with the 80-year storage scenario, this individual will be exposed to the world atmospheric concentration of ^{85}Kr for an additional 50 years, thus providing an 80-year cumulative dose. Thus, the 80-year cumulative dose to an individual member of the world population (Equation 3) must be added to the dose resulting from Equation 6 to obtain the 80-year cumulative dose to the maximum-exposed individual.

The raw meteorological data used for these analyses is presented in the Commercial Waste Management Statement (DOE 1979). The annual average atmospheric dispersion factors ($\bar{X}/Q'$) calculated from these data for a 110-m stack height are presented in Table D.4. By inspection of the data, the maximum offsite ground-level concentration ($\bar{X}/Q' = 1.5\text{E-}8 \text{ sec/m}^3$) occurs 2 to 4 miles from the plant in the SE direction.

The external dose factors (Df) used here are calculated as shown by Soldat et al. (1976). However, for comparison, Table D.1 lists published values by several workers. The assumptions used by these authors were not always the same. This is especially true of the depth at which the "skin"

TABLE D.4. Annual Average Atmospheric Dispersion Factors (110-m stack height) (sec/m³ at various miles from release point)

Sector	0.5-1	1-2	2-3	3-4	4-5	5-10	10-20	20-30	30-40	40-50
N	1.3E-8	1.1E-8	1.1E-8	1.1E-8	1.0E-8	7.5E-9	4.3E-9	2.6E-9	1.8E-9	1.4E-9
NNE	5.7E-9	5.6E-9	6.0E-9	6.1E-9	5.8E-9	4.6E-9	2.8E-9	1.8E-9	1.2E-9	9.6E-10
NE	5.7E-9	6.3E-9	6.4E-9	6.3E-9	6.0E-9	4.6E-9	2.8E-9	1.8E-9	1.3E-9	9.8E-10
ENE	4.1E-9	4.2E-9	4.7E-9	4.9E-9	4.8E-9	3.8E-9	2.4E-9	1.5E-9	1.1E-9	8.3E-10
E	5.1E-9	7.3E-9	9.2E-9	9.8E-9	9.4E-9	7.2E-9	4.1E-9	2.5E-9	1.7E-9	1.3E-9
ESE	9.3E-9	1.1E-8	1.2E-8	1.2E-8	1.1E-8	7.6E-9	4.2E-9	2.5E-9	1.7E-9	1.3E-9
SE	1.0E-8	1.3E-8	1.5E-8	1.5E-8	1.4E-8	9.9E-9	5.3E-9	3.0E-9	2.0E-9	1.5E-9
SSE	6.0E-9	7.7E-9	8.5E-9	8.2E-9	7.5E-9	5.3E-9	2.9E-9	1.7E-9	1.2E-9	8.7E-10
S	5.3E-9	6.2E-9	6.8E-9	6.7E-9	6.2E-9	4.6E-9	2.6E-9	1.6E-9	1.1E-9	8.5E-10
SSW	2.3E-9	3.5E-9	4.3E-9	4.4E-9	4.2E-9	3.2E-9	1.9E-9	1.2E-9	8.4E-10	6.4E-10
SW	3.1E-9	4.8E-9	6.0E-9	6.2E-9	5.8E-9	4.4E-9	2.6E-9	1.6E-9	1.1E-9	8.4E-10
WSW	1.4E-9	2.8E-9	4.0E-9	4.5E-9	4.4E-9	3.5E-9	2.2E-9	1.4E-9	9.7E-10	7.4E-10
W	1.6E-9	3.3E-9	5.0E-9	5.5E-9	5.3E-9	4.1E-9	2.4E-9	1.5E-9	1.0E-9	8.0E-10
WNW	2.2E-9	3.5E-9	4.6E-9	4.8E-9	4.6E-9	3.4E-9	1.9E-9	1.2E-9	8.0E-10	6.0E-10
NW	4.7E-9	5.4E-9	6.6E-9	6.9E-9	6.6E-9	5.0E-9	2.9E-9	1.8E-9	1.2E-9	9.4E-10
NNW	5.6E-9	5.3E-9	6.3E-9	6.7E-9	6.5E-9	5.0E-9	3.0E-9	1.8E-9	1.3E-9	9.8E-10

dose was calculated. The dose to the internal organs, including the lung, from external exposure are generally assumed to be the same as the total-body dose. To estimate the lung dose from inhalation, Soldat et al. (1976) calculated dose factors from the ICRP initial lung model (ICRP 1959). The concentration of the noble gas in the air within the lung is assumed to be the same as that in the inspired air. The effective energies of the individual radiations are calculated using an effective radius of 10 cm for the lung.

The inhalation dose factor calculated by Soldat is compared in Table D.2 with values obtained by other researchers. Caution should be used when making a critical comparison of these values. For example, Russell and Galpin (1972) used a lung volume of 5.6 liters, Soldat et al. (1976) based their calculations on a lung volume of 4 liters, and Whitton (1968) did not state the lung volume used for her calculations. When the variations in assumptions are factored into the calculation of the dose factors by these various researchers, there is reasonable agreement among the values.

The total dose would of course be the sum of the external component and the internal contribution. The doses from ^{85}Kr absorbed in tissue is generally small compared to the dose from direct external radiation. It is apparent that for ^{85}Kr the skin is the critical organ for individuals. For collective dose calculations the generally accepted practice is to calculate total-body doses so that health effects could then be estimated if desired.

D.2 OCCUPATIONAL DOSES

This section briefly describes the methodology and computer codes used in this study to calculate the occupational doses, as well as the methods used to verify their accuracy. The two computer codes, discussed in detail in previous PNL publications, are: DACRIN (Houston, Strenge and Watson 1974) for internal doses from the inhalation of ^{85}Kr and ISOSHL (Engel, Greenborg and Hendrickson 1966 and its supplement) for external doses.

The evaluation of occupational exposures received during the operation of ^{85}Kr recovery and storage facilities includes external and internal exposure

calculations. External exposure may be received from discrete sources of ^{85}Kr , such as, gas cylinders, and from airborne ^{85}Kr (air submersion). In the latter case, inhalation of ^{85}Kr leads to internal exposure to the lungs.

As indicated in Section 3.2, ^{85}Kr decays primarily by beta emission, with a 0.514 MeV photon emitted with an intensity of only 0.43%. Much of the occupational dose from shielded ^{85}Kr sources arises from bremsstrahlung radiation from 0.687 MeV beta particles.

The PNL computer code ISOSHL D is thought to be suitable for external exposure evaluation when discrete ^{85}Kr sources could be identified, since it includes the dose contribution from bremsstrahlung. The ISOSHL D program uses a point-kernel integration technique to evaluate exposure rates at a detector location specified by the user. Photon energy flux is calculated for 25 energy groups with average energies ranging from 15 keV to 3 MeV. The contribution of internal bremsstrahlung radiation originating within the krypton source is included in the exposure rate calculations.

In order to verify the codes applicability, however, some initial testing was performed. Thermoluminescent dosimeters (TLDs) were placed at various distances from the top, side and bottom of gas cylinders containing either 20 or 100 curies of ^{85}Kr gas. ISOSHL D runs were then set up to approximate the TLD exposure conditions and locations. The exposure rates as measured by the TLDs were compared to the exposure rates calculated by ISOSHL D. Exposure rates as calculated by ISOSHL D in general showed good agreement (within a factor of two) with measured exposure rates for side and bottom cases. In modeling exposure rates from the top of the cylinders, however, $3\frac{1}{2}$ to 4 inches of steel shielding had to be added in order to give results comparable to the TLD measurements. Having done this, it was felt that ISOSHL D could be used to model the occupational exposure rates from gas cylinders containing 130,000 Ci of ^{85}Kr .

A computer code KRDOZE, written for this study, is used to interpolate dose rate and shielding data which have been calculated by ISOSHL D from discrete sources of ^{85}Kr . This is an interactive computer code and allows the

user greater calculational freedom. For example, if the dose rate is found to be excessive, the user can evaluate the effect of increased shielding merely by changing one parameter while on-line at a computer terminal. KRDOZE is a very simplistic code; it assumes point sources of ^{85}Kr and slab shields of ordinary concrete, depleted uranium, lead or water. The chief advantage of the code is the speed of computation. Complex three-dimensional arrays of ^{85}Kr sources, such as gas cylinders in the storage facility behind a concrete wall or the cryogenic distillation columns in the concrete cells, can be mocked up as a series of point sources and the dose rates evaluated at various points within a few minutes at a terminal.

The PNL computer code, DACRIN, is used to assist in the evaluation of internal occupational doses due to the operation of the ^{85}Kr recovery and storage facility.

It is assumed that the principal source of airborne ^{85}Kr is the routine stack release from the ^{85}Kr recovery facility. DACRIN describes the atmospheric dispersion of radionuclides by the use of a bivariate normal distribution model. The standard deviations of the cloud concentration in the cross wind lateral and vertical directions are estimated by Pasquill's curves.

Given a stack (release) height, average wind speed, and Pasquill atmospheric stability class, DACRIN calculates the atmospheric dilution factor ($\bar{X}/Q'$) at user-specified downwind distances. Since the $\bar{X}/Q'$ value is simply the ratio of the air concentration (Ci/m^3) to the release rate (Ci/s), and the release rate is known, the ^{85}Kr air concentration can be evaluated.

Dose calculations for ^{85}Kr in lung are based on the following:

- Assume all "vital capacity" (not tidal volume) of the lung is contaminated to the same concentration as the inhaled air.
- Assume 100% of particulate radiation (β^- , β^+ , e^- , α) is absorbed in the lung with a quality factor of 1.0 for β^- , β^+ , and e^- and 10 for α .
- Assume gamma is absorbed according to exponential energy-dependent formula give in ICRP Publication 2 (ICRP 1959).

Based on these assumptions, a dose equivalent rate factor (mrem/yr per pCi/yr) is derived and used. In order to evaluate the pCi/yr inhaled, the air concentration calculated earlier is multiplied by the ICRP reference man occupational breathing rate and appropriate conversion factors. Thus, a dose equivalent rate to the lung from inhalation is obtained.

APPENDIX E

ALTERNATIVE STORAGE FACILITY DESIGN

APPENDIX E

ALTERNATIVE STORAGE FACILITY DESIGN

About 6% of the krypton present in fuel reprocessing plant dissolver off-gas systems (DOG) is ^{85}Kr . During the recovery process, ^{85}Kr is removed from the DOG by a rare gas recovery system integral to the reprocessing plant, described in Appendix C. A krypton storage facility for gas cylinders has been described in detail (DOE 1979 and 1979a). This facility is co-located with the reprocessing plant in order to avoid accidents involved in transporting large numbers of gas-filled cylinders and is used as the reference storage facility design for Section E.1 of this report. The storage of immobilized ^{85}Kr sputtered unto amorphous metal may slightly alter the facility design alternatives.

E.1 KRYPTON GAS CYLINDER STORAGE

Present technology for storing krypton gas is limited to containing it in standard industrial high-pressure gas cylinders until the ^{85}Kr has decayed to a concentration that would allow its release.

The reference facility is equipped to receive, monitor, inspect and store commercial-type gas storage cylinders filled with ^{85}Kr gas. The facility also provides the capability to retrieve a leaking cylinder and transfer its contents to an empty cylinder.

E.1.1 Operation and Description of the Reference Horizontal Cylinder Storage Facility

The shielded gas cylinder containing ^{85}Kr is received at the cylinder inspection station. The shielded cylinder cask is examined for evidence of damage, surface contamination from FRP-derived fission products and gas leakage. After inspection and monitoring, the cylinder passes from its shielded cask through an air lock into the receiving hot cell. The gas cylinder is

then examined for evidence of leakage. The hot cell ventilation system can be routed back to the FRP DOG system to handle rupture quantities of ^{85}Kr . If leaks are found, the ^{85}Kr is transferred to an empty cylinder. After this determination has been made, the cylinder is passed out of the hot cell and into another shielded transfer cask. This cask is used momentarily to move the filled gas cylinder into the storage area. The cylinder is again removed from its cask as it moves into one of a series of storage cells. The storage cells are equipped to remove decay heat, monitor for ^{85}Kr leaks, and provide shielding and security for the gas-filled cylinders.

A more detailed description of the facility operation, as it relates to occupational radiation protection, is presented in Section 6.3.3.

Figure E.1 is a plot plan of the reference facility (DOE 1979a). The gas cylinder storage facility is located at the reprocessing site and connected by an access road. The facility is enclosed within its own fenced exclusion area.

FIGURE E.1. Krypton Storage Facility Plot Plan

The krypton storage facility has its own heating and ventilation, water treatment and emergency electrical generating equipment. The reprocessing plant, however, does supply primary electrical power, water and sanitary sewage disposal.

The reference storage facility is built in three phases. The initial construction and the second and third phases are shown in Figure E.1. Phase I storage area is a long, narrow building that measures 46 x 542 feet (14 x 165 m) long. The building is designed as a secondary containment to the cylinder storage cells with exterior walls of standard precast concrete and roofs of insulated metal decking with built-up roofing. All exterior walls and ceilings are insulated and caulked to prevent the escape of krypton to the uncontrolled environment. It is expected that the Phase I storage vault will be of sufficient volume to receive about 10 years worth of pressurized steel cylinders containing ^{85}Kr . Therefore, it is estimated that Phase II and Phase III of storage area construction will be ready for occupancy at 10-year intervals.

The interior of the storage vault is comprised of an operating area and a storage area. A gas-tight partition with an air lock is used to separate these two areas.

A central rail track runs the length of each storage building, between two rows of storage cells, to provide for movement and positioning of the shielded container transfer carriage. A 20-ton overhead bridge crane is used for servicing and maintaining each storage cell.

Shielding and Special Handling

The gas-filled pressurized cylinders are radioactive and require the use of shielding and remote handling techniques. Krypton is an inert gas and primarily a beta emitter, thus the radiation dose to man resulting from its release normally results only from external exposure. In addition, ^{85}Kr decays by gamma radiation (0.514 MeV) 0.41% of the time. However, when ^{85}Kr is contained within a cylinder, a significant added hazard occurs. Penetrating gamma radiation called bremsstrahlung is generated within the gas itself and

The hot cell is a Category I structure made of high-density concrete; its walls and roof are 3 feet (1 m) thick. Work stations are equipped with lead glass windows for viewing in-cell operations, a pair of master-slave manipulators and a control station for a remote heavy duty, powered manipulator.

The pressurized cylinder storage area is normally unoccupied; personnel enter only to deposit or remove cylinders. Entry of personnel into the storage building is rigorously controlled to prevent unnecessary radiation exposure.

FIGURE E.2. Detail of Two Storage Cells Showing Location of 20-Ton Crane and Structure Dimensions

E.1.2 Operation and Description of Alternative Cylinder Storage Facilities

The first alternative considered is a remotely operated horizontal storage facility. Other designs evaluated include vertical cylinder storage and pool storage.

Remote Horizontal Storage Facility

The remote horizontal storage facility is operated in the same manner as the manual facility with one basic difference. In this design, no operator is needed on the forklift which transfers the krypton cylinders between the storage cells and the hot cell.

Vertical Storage Facility

The vertical storage facility as shown in Figures E.4 and E.5 is very similar to the design used for horizontal cylinder storage. The blowers and chillers and other items requiring maintenance are located away from a high radiation zone and 36 inches of concrete is used for shielding.

FIGURE E.4. Vertical Storage Alternatives

FIGURE E.5. Vertical Storage Alternatives

Pool Storage Facility

The last alternative design studied is pool storage using water rather than air as a coolant. There is a possible corrosion problem with water storage, although the gas cylinders could be coated for corrosion resistance. There is also a possible problem with the rubidium daughter of ^{85}Kr decay reacting with water to form hydrogen in the event of a severe leak from a gas cylinder. An inert atmosphere could prevent the remote possibility of a hydrogen explosion.

A plan and elevation view of a pool storage facility is shown in Figures E.6 and E.7. This facility is equipped with storage racks which can accommodate 200 gas cylinders. The gas cylinders have a modified cap which a bayonet which mates with a lifting device suspended from an overhead crane. The crane is remotely operated by a person outside the storage cell using

FIGURE E.6. Pool Storage

mirrors and TV cameras. A second crane is available to retrieve the first crane if it malfunctions. The bottles are moved to the examination hot cell through an underwater transfer channel. The pumps and heat exchangers for cooling are located in a separate equipment bay, which is also equipped with an overhead crane to help in replacement of defective equipment. All items which require maintenance are placed in areas where the radiation dose rate is negligible.

FIGURE E.7. Pool Storage

APPENDIX F

^{85}Kr RECOVERY AND STORAGE FACILITY ACCIDENT ANALYSIS

APPENDIX F

⁸⁵Kr RECOVERY AND STORAGE FACILITY ACCIDENT ANALYSIS

During the recovery and storage operations unforeseen accidents will occur. Therefore a projection of the type, frequency and severity of such accidents is essential.

F.1 BASIS FOR THE ACCIDENT ANALYSIS

The objective of the accident analysis is to estimate the amount of radioactive material which may be accidentally released during krypton recovery and storage operations. These radioactive material releases (called source terms) when added to the other radiation sources provide the basis for estimates of potential radiation exposure to employees and nearby populations.

The accident analysis presented herein is based on generic facilities, processes and inventories. At the time of this analysis, insufficient process and facility specifications were available to allow for an exhaustive examination of all conceivable accidents. However, the postulated accidents, their postulated frequency of occurrence and the amount of radioactive material released, are intended to bracket the spectrum of accidents which could occur at any licensed facility performing cryogenic krypton recovery, immobilization and/or pressurized krypton gas storage.

Before a construction permit would be issued for these types of facilities additional analyses would be completed in compliance with Federal Regulation 10 CFR 50 (NRC 1979). 10 CFR 50 requires an environmental impact statement and a preliminary safety analysis report based on preliminary design details. In addition, the facility operating license requires a final safety analysis report based on final design details. These safety analyses define the facility safety hazards and describe the safety systems that will keep accidental releases within regulatory limits.

A complete source term includes information such as type and quantity of nuclides, chemical form, release point, rate of release, and quantity of diluent (air and water) accompanying the release. However, the point of release and diluent flow rates are the same for most accidents at a particular facility; for example, the main stack and stack effluent at a fuel reprocessing plant (FRP). In these cases the necessary source term data reduces to released material quantities and the rate of release.

Accident frequency estimates were developed for the identified accidents. These estimates are based on whatever previous experience with similar equipment exists; or are engineering judgments based on review of the conceptual designs. The given frequency values should be considered order of magnitude estimates. A listed frequency for a specific event is believed to be an upper limit estimate; however, they have a substantial uncertainty since accident frequencies are sensitive to the type, manufacture, installation, and operation of the equipment.

Following source term and frequency definition, the lists of representative accident scenarios were classified into three accident severity groups:

- Minor: relatively frequent occurrences involving process interruptions without potential for significant release of radioactive or other hazardous materials.
- Moderate: infrequent events with potential for small material release, major equipment damage, or the creation of radiation fields in occupied zones which could result in occupational exposures exceeding 10 CFR 20 limits (5 rem/yr) (NRC 1979a), but not to exceed 25 rem/yr.
- Severe: unlikely events with a potential for significant radiation hazards; these events are postulated to establish performance requirements for plant safety systems. Accidental releases of sufficient radioactive materials to cause occupational exposures which could result in detectable clinical effects (assumed to be >50 rem) are included in this category. Since the safety systems are designed to confine and mitigate the impact of these severe accidents, the actual release may be no larger than the "minor" or "moderate" class accidents.

The three accident classifications cover the spectrum of design-basis accidents. Nondesign-basis accidents include all accidents which exceed site criteria^(a) or involve concurrent independent failure of process and multiple containment system barriers. By virtue of plant design and operational techniques, the possibility of a nondesign-basis accident is extremely unlikely during the 100-year, or shorter, design life of the Kr recovery and/or storage facility.

Releases of radioactive material to the environment result from two different events: 1) normal operational releases and 2) releases resulting from accidents. Operational releases result from routine handling or processing of radioactive materials and are limited by the containment system design and performance. They are expected to occur at a relatively uniform rate over the life of the plant. Accidental releases occur intermittently because of operational errors or system component and containment failures, and releases are generally inversely proportional to their frequency.

The accident descriptions for the krypton recovery and storage systems are presented in Tables F.1 through F.8 by severity category in the following four column tabular format. This format concisely describes the sequential nature of the accident propagation and the action of the plant safety systems to mitigate the potential accidental releases:

- Column 1 - accident number; - a two-digit number that identifies each accident by technology, descriptive title, and estimated frequency of occurrence.
- Column 2 - sequence-of-events information that shows the dynamic path of the accident.
- Column 3 - safety system description that explains how an accident can be prevented by design, discovered by alarms, and/or mitigated by a confinement system and/or operator action.

(a) Site criteria include: 1) definition of the maximum credible earthquake, surface faulting, floods, and wind velocities based on historical evidence, local and regional geology, and expert judgment; 2) local and regional demography; and 3) proximity and definition of hazards caused by man.

TABLE F.1. Minor Accidents - Cryogenic Kr Recovery System

Accident No. and Description	Sequence of Events	Safety System	Release
1.1 Freezing. Expected frequency: ~6 per year.	<ol style="list-style-type: none"> 1. Line or process unit (condensers, dryers, separation columns in cold box) plugs due to accumulation of ice, nitrogen oxides, and/or frozen process gas. 2. Loss of gas flow is identified by instrumentation. 3. Dual process unit (if it exists) is switched into service, or process gas is temporarily vented to FRP Stack. 4. The plugged system is warmed and purged to remove the ice plug. 5. The process unit is returned to service within 8 hours. 	<ol style="list-style-type: none"> 1. Process gas flow monitoring system provides continuous information. 2. Some portions of the cryogenic system have redundant units installed in parallel, or the gas flows are routed to the FRP stack. 3. Process gas lines and vessels contain pressure relief valves which are vented to FRP Stack. 	Minor release of purge volumes - and feed gas while system is down. (This cold box process inventory is stored in an appropriately sized storage tank.) ⁸⁵ Kr is released continuously from the FRP stack at a rate of 2400 Ci/hr for 8 hours.
1.2 Plugged lines or system malfunction resulting in process gas routed to FRP stack (e.g., Process gas dryer vents process gas during drying cycle). Expected frequency: ~1 per year.	<ol style="list-style-type: none"> 1. H₂O condenser, demister of other process unit plugs. 2. Loss of gas flow noted by instrumentation. 3. Identified bed is isolated from system. 4. Dual system switched into service, or process gas temporarily vented to FRP Stack. 5. Plugged bed replaced. 6. The process unit is returned to service within 48 hours. 	<ol style="list-style-type: none"> 1. Equipment upstream to control particulates. 2. Radiation and air flow monitoring systems provides continuous information. 3. Loss of product output. 4. The adsorber and reactor systems have redundant units installed in parallel. 5. The beds are capable of being remotely replaced while in service. 	Minor release of process gas via plant stack. ⁸⁵ Kr is released from the plant stack at a rate of 2400 Ci/hr for 48 hours.

TABLE F.1. Minor Accidents - Cryogenic Kr Recovery System (contd)

Accident No. and Description	Sequence of Events	Safety System	Release
<p>1.3 System leak to process cell atmosphere.</p> <p>Expected frequency: 3 per year.</p> <p>This accident description is expected to cause 1 system outage per year with 48 hours down time. More frequent occurrences of this type will cause down times averaging 12 hours. One accident in 2 years will require venting of process gas inventory. One accident in 5 years will also require venting of a half full Kr receiver.)</p>	<ol style="list-style-type: none"> 1. Process line, valve or process unit (e.g., compressor) develops leak. 2. Krypton detected in process cell atmosphere. 3. Operators isolate portions of process line to locate leak. 4. Suitable portion of system bypassed (switched to redundant system if available) and purged to allow access for repair. 5. If leak occurs within cold box, system is purged to temporary storage tank (tank is sized to hold cold box inventory plus Kr receiver inventory). 6. Repair is completed. 7. If leakage is minor, repair may be delayed. 	<ol style="list-style-type: none"> 1. Radiation detector continuously monitors cell exit ventilation air flow. 2. Similar detector checks inert gas in cold box. 3. High maintenance redundant items are located in separated cells to avoid process shutdown. 	<p>Process inventory plus 48 hours FRP output up stack once per 2 years. Process inventory is released in a 12 hour period in addition to the normal production rate. Release rate for first 12 hours is 8,600 Ci/hr followed by 36 hours at 2,400 Ci/hr.</p> <p>Once per 2 years this accident will release 2,400 Ci/hr for 48 hours.</p> <p>Once per 2 years this accident will release 2,400 Ci/hr for 48 hours.</p> <p>Twice per year, leaks will cause down times averaging 12 hours (releasing 2,400 Ci/hr for 12 hours).</p>
<p>1.4 Loss of cooling supply to cryogenic separator.</p> <p>Expected frequency: 0.2 per year</p>	<ol style="list-style-type: none"> 1. Loss of LN₂ supply and/or loss of refrigerated cooling to precooler. 2. Condition promptly detected by loss of flow and/or process gas pressure surge. 3. Operators promptly restore cooling (w/o process interruption) or shutdown the system. 4. Process gas inventory is routed to the emergency holding tank. 5. Process gas flow is routed to FRP stack. 	<ol style="list-style-type: none"> 1. Temperature monitors on all process units. 2. Pressure monitors on all process units. 3. Flow monitor on LN₂ supply. 4. Temperature sensor on precooler outlet. 5. Pressure relief valves (vented to FRP stack) on all process units. 6. A holding tank is available which is sufficiently large to hold the cold box process inventory plus the liquid Kr receiver inventories. 	<p>Treated DOG routed to FRP Stack for 24 hours (2,400 Ci/hr for 24 hrs).</p>

TABLE F.2. Moderate Accidents - Cryogenic Kr Recovery System

Accident No. and Description	Sequence of Events	Safety System	Release
<p>1.5 Extended process shutdown, with ^{85}Kr venting</p> <p>Expected frequency: ~0.2 per year</p>	<ol style="list-style-type: none"> 1. Recovery system shut down because of equipment failure. Radiation and airflow monitoring systems record venting of Kr directly to APS and FRP main stack. 2. Dissolver and main FRP continues to process for 30 days. 3. Offsite Kr concentrations monitored to assure adequate dispersion. 4. Equipment repaired and returned to service. 	<ol style="list-style-type: none"> 1. Redundant equipment installed. Radiation and airflow monitoring systems provide continuous information. 2. If repairs are not completed within 30 days the FRP is shut down. 3. Offsite monitoring system installed. 	<p>The DOG would be vented to the stack after treatment. 5.8×10^4 Ci of ^{85}Kr would be released per day (2,400 Ci/hr for a maximum of 30 days. Current practice allows complete Kr release. Short-term releases in future reprocessing plants would have lesser consequences.</p>
<p>1.6 Oxygen recombiner explosion; with or w/o building breach.</p> <p>Expected frequency: ~0.1 per year w/o building breach ~0.02 per year with building breach.</p>	<ol style="list-style-type: none"> 1. Gases in recombiner reach detonation limit (4 vol% H_2) owing to failure of exit gas recycle or a poisoned bed. 2. Gases ignite. 3. Rupture disk vents to relief piping. Choked flow develops. 4. Vessel ruptures. 5. If the explosion is sufficiently violent the cell explosion panel is breached. 6. Off-gas stream routed to redundant system. 7. Failed recombiner replaced and adjacent facilities repaired. 	<ol style="list-style-type: none"> 1. Instrumentation and alarms monitor H_2 levels. Exit gas piping designed to assure adequate recycle. 2. No ignition sources present. 3. Rupture disk sized for deflagrations and minor over-pressures. 4. Explosion barriers and breakaway building panels provided to mitigate explosion energy. 5. Redundant recombiner installed in parallel. 	<p>56 Ci ^{85}Kr released to cell atmosphere and out FRP stack (1.4 minute release period), Kr released at ground level in restricted zone (1.4 minute release).</p>
<p>1.7 Cryogenic system or cell explosion with or w/o building breach.</p> <p>Expected frequency: ~0.05 per year w/o cell breach. ~0.005 with cell breach.</p>	<ol style="list-style-type: none"> 1. Recombiner function becomes ineffective resulting in H_2/O_2 or NO_x/O_2 into the cryogenic system. (Other combustible gas sources entering process stream or process cell can be postulated as freak accidents.) 2. Gases ignite. 3. One or more vessels rupture. 4. Process cell remains intact causing process inventory to be vented through plant stack, or cell containment is lost and process inventory is vented at ground level. 	<ol style="list-style-type: none"> 1. Items 1 to 3 in accident 2 above also apply to cryogenic separation system. 2. Cryogenic system would not behave normally with significant quantities of foreign gases present. 3. The cold box structure is a blast shield. 4. The cell access door will be designed to preferentially vent a noncontainable explosion to the recombiner cell and/or breakaway panel. 5. Liquid Kr product receiver is located in a separate shielded cell. 	<p>If the accident does not result in cell breach, the process inventory will be vented to the plant stack (245,000 Ci in one hour) followed by the 30-day extended process shut down as in accident 1.5 above, or with cell breach - cryogenic process inventory plus 15 minutes of normal process flow released at ground level. 245,000 Ci released in a 4.6 minute burst to a restricted zone, followed by 2,400 Ci/hr release for 30 days from FRP stack.</p>

TABLE F.3. Minor Accidents - Kr Gas Cylinder Filling, Transportation, Interim Storage, Long Term Storage and Decommissioning

Accident No. and Description	Sequence of Events	Safety System	Release
<p>2.1 Krypton cylinder develops small but detectable leak (or cylinder leakage is detected on receiving inspection).</p> <p>Expected frequency: Indoor: ~3 per year. Outdoor: ~0.02 per year.</p>	<ol style="list-style-type: none"> 1. Valve stem leakage or pinhole develops in cylinder or valve structure (assume nominally detectable leak rate). 2. Beta sensitive detector located in the storage cell or ventilation duct alarms, or if leakage occurs during acceptance inspection alarm in the inspection cell indicates the leakage, or Kr cylinder develops a small leak while being transported (assume ten times leak rate that would alarm indoor detectors). 3. If leakage is detected during acceptance inspection, the Kr is rebottled. 4. If cylinders are stored under water, significant leaks will be observed (bubbling). 5. If leakage is detected in storage, the leaker is isolated using sniffing probes. The leaking cylinder is recovered and taken to the hot cell for rebottling. 	<ol style="list-style-type: none"> 1. Beta sensitive detector in each storage vault to detect Kr released to the vault atmosphere. 2. Facilities (protective clothing, breathing apparatus, etc.) to allow safe handling of leaking gas cylinders. 3. Cylinder pressure is insufficient to rupture cylinder. 	<p>Detection capability is very powerful. Indoor release rate equals 0.2 Ci/hr for one day.</p> <p>Outdoor release rate equals 2.0 Ci/hr ground level for one day.</p>
<p>2.2 Minor direct exposure from Kr cylinder</p> <p>Expected frequency: ~2 per year.</p>	<ol style="list-style-type: none"> 1. Misalignment of transfer cask, failure to properly close transfer cask, improper storage vault closure sequence, etc. Results in short term direct exposure from 1 or more Kr gas cylinders. 2. Facility radiation alarms activate, or operator detects the improper sequence which results in exposed cylinders. 3. Operator(s) move out of direct radiation zone. 4. Shielding is corrected. 	<ol style="list-style-type: none"> 1. Facility radiation detectors located and designed to alarm when radiation level exceeds specification. 	<p>None</p>
<p>2.3 Loss of storage cell cooling system.</p> <p>Expected frequency: ~1 per year.</p>	<ol style="list-style-type: none"> 1. Power to substation supplying krypton storage facility interrupted, and/or refrigerator system fails. 2. Emergency power generators are activated and/or alternate cooling system is activated. 	<ol style="list-style-type: none"> 1. Emergency power diesel generator (several redundant emergency power supplies). 2. Redundant cooling systems. 3. Kr cylinders will remain at safe temperature indefinitely w/o aid of the storage cell cooling system. Cooling will take place by convection and conduction through the vault and building walls. 	<p>None</p>

TABLE F.4. Moderate Accidents - Kr Gas Cylinder Filling, Transportation, Interim Storage, Long Term Storage and Decommissioning

Accident No. and Description	Sequence of Events	Safety System	Release
<p>2.4 One krypton cylinder ruptures in cell (filling, inspection or storage).</p> <p>Frequency of occurrence: ~0.05/yr.</p>	<ol style="list-style-type: none"> 1. Krypton cylinders rupture while filling, ring during inspection in the hot cell, or in a storage vault. Cylinder rupture could be caused by overfilling, introduction of impure gas (O, N, air, H₂O, hydrocarbons. Ozone and nitrogen oxides will be formed catalyzed by radiation), and/or corrosion. 2. Leak detected by radiation monitoring system. 3. Vault isolation valves are closed. 4. Contaminated air is contained in the vault or purged to a holding tank. If neither is possible, it is released through the storage facility stack. 5. Holding tank contents are released to the stack at a controlled rate or sent to FRP for reprocessing. 	<ol style="list-style-type: none"> 1. Radiation monitoring system provides continuous information as to radiation levels. 2. Damage to other gas cylinders in the storage vault is minimized by restraints in the storage racks and the protective valve caps. 3. The vault is isolated from the remaining storage facility. 4. The operations cells and the storage vaults have individual radiation detectors (air sampling for beta activity) and ventilation control. The cells are leak tight. 5. The contaminated air is pumped into a holding tank and released to the stack or returned to the FRP for reprocessing. 	<p>Controlled release will not exceed normal FRP operational release rate of 58 kCi/day through the FRP stack or facility stack. Up to 130 kCi may be released.</p>
<p>2.5 Krypton cylinder corrodes due to Rubidium.</p> <p>Frequency of occurrence: ~0.05/yr.</p>	<ol style="list-style-type: none"> 1. Valve seat and stem destroyed due to radiological decomposition, or stress corrosion cracking of cylinder body. 2. Radiation monitoring system detects leaking cylinder. 3. Leaking cylinder is identified using sniffing probes. 4. The cylinder is recovered and the gas is transferred to new cylinder. 	<ol style="list-style-type: none"> 1. Monel or equal quality valve. 2. Radiation monitors for leak detection. 3. Handling and detection equipment are available to identify and recover the leaking cylinder. Facilities are provided for safe gas transfer to new cylinder. 4. Long term cylinder inspection should detect any general corrosion process that develops allowing re-bottling of the Kr prior to development of leaking cylinders. 	<p>Controlled release will not exceed normal FRP operational release rate of 58 kCi/day through the FRP stack. It is assumed that this type of failure will not occur prior to one half life. Up to 65 kCi of ⁸⁵Kr may be released.</p>
<p>2.6 Moderate direct over-exposure from mishandled Kr Cylinder.</p> <p>Expected frequency: ~0.1 per year.</p>	<ol style="list-style-type: none"> 1. Handling error (with or w/o failure of radiation alarms to function) exposes Kr cylinder. This condition exists long enough that operational over-exposure occurs. 2. Condition is recognized and corrected. 	<ol style="list-style-type: none"> 2. Facility radiation alarms, spot surveys, shipping surveys, personnel radiation badges and pocket dosimeters all will contribute to detection of high radiation field. 	<p>None</p>

TABLE F.5. Severe Accidents - Kr Gas Cylinder Filling, Transportation, Interim Storage, Long Term Storage and Decommissioning

Accident No. and Description	Sequence of Events	Safety System	Release
<p>2.7 Krypton cylinder ruptures indoors or outdoors.</p> <p>Expected frequency: Indoor: ~0.02 per year. Outdoor: ~0.001 per year.</p>	<ol style="list-style-type: none"> 1. Krypton cylinder ruptures during handling in the operating area or storage corridor. Rupture could be caused by penetrating and/or crushing accident, overfilling, impure gas and/or corrosion (See accident #2.4). 2. Leak detected and alarmed by monitoring system. (Beta activity in air.) 3. Personnel are evacuated from the contaminated area. Fresh air masks are used if necessary. 4. Krypton is vented to the facility stack. Ventilation control is used to restrict the rate of release. 5. Personnel reenter facility and repair damages. 	<ol style="list-style-type: none"> 1. Transfer cask minimizes damage to the cylinder. 2. Monitors and alarms provide continuous information. 3. Personnel trained for emergency response. 4. Fresh air masks are immediately available at all work stations. 5. Cylinders pressurized to 500 psi vs 2000 psi design operating pressure. Each cylinder is pressure checked to 3000 psi. 6. Nuclear facility is located within an exclusion zone sufficient to limit plant boundary maximum transient dose to 25R. 	<p>Indoor rupture - 130 kCi of ⁸⁵Kr is released to the krypton storage facility stack in 0.5 hr. Worker located within 10 ft of cylinder at time of rupture. Worker trained in emergency action and in evacuation procedure.</p> <p>Outdoor rupture - 130 kCi of ⁸⁵Kr is released at ground level in a 1.4 minute burst. Release is centrally located within the facility exclusion zone. A worker is located (randomly) within 50 ft of cylinder at time of rupture.</p>

TABLE F.6. Minor Accidents - Immobilized Kr Loading, Transportation, Interim Storage, Long Term Storage and Decommissioning

Accident No. and Description	Sequence of Events	Safety System	Release
<p>3.1 Pinhole leak in Kr canister.</p> <p>Expected frequency: Indoor: ~1 per year. Outdoor: ~0.01 per year.</p>	<ol style="list-style-type: none"> 1. Event sequence is identical to Accident #2.1. 	<ol style="list-style-type: none"> 1. Safety systems as described in accident #2.1 are applicable to immobilized Kr. 2. The immobilized Kr matrix provides a restraint to the release of Kr. 3. Container leakage (if in excess) will initiate slowly. 	<p>The quantity of Kr which builds up at normal storage temperature (assume 90°C) for the first month after encapsulation would be available for release. Assume leak rate is similar to Krypton gas cylinder leak rate (which is based on detection threshold).</p> <p><u>Indoors -</u> 3.2×10^{-3} Ci/min for one day (0.2 Ci/hr).</p> <p><u>Outdoors -</u> 2 Ci/hr ground level for 2.5 hrs. (Assume 1 occupational worker is located statistically within 50 ft of the cylinder for 2 hours.)</p>
<p>3.2 Direct exposure from Kr Canister.</p> <p>~2 per year.</p>	<ol style="list-style-type: none"> 1. Event sequence identical to Accident #2.2. 	<ol style="list-style-type: none"> 1. Safety systems identical to Accident #2.2. 	<p>None</p>

TABLE F.7. Moderate Accidents - Immobilized Kr Loading, Transportation, Interim Storage, Long Term Storage and Decommissioning

Accident No. and Description	Sequence of Events	Safety System	Release
<p>3.3 Kr Canister ruptures in loading cell, hot cell, or storage cell.</p> <p>Frequency of occurrence: ~0.05 per year.</p>	<p>1. Kr canister rupture in controlled atmosphere facility. Rupture could occur from crushing/shearing/puncturing accident, corrosion, improper fabrication or stacking resulting in a temperature which exceeds the release point.</p> <p>2. Events 2 through 5 are identical to Accident #2.4.</p>	<p>1. Safety systems identical to Accident #2.4.</p>	<p>Inventory of krypton which has been released from the solid matrix will be released, 5 Ci (in 2 min to storage facility stack).</p>
<p>3.4 Corrosion</p> <p>Frequency of occurrence: ~0.05 per year.</p>	<p>1. Kr solid matrix and/or container corrodes due to rubidium attack, action of humidity or other outside agent, or matrix material degrades from radiolysis or other unanticipated</p> <p>2. Events 2 to 4 identical to Accident #2.5.</p>	<p>1. Kr solid matrix extensively tested to minimize corrosion mechanisms.</p> <p>2. Safety systems 2 to 3 identical to Accident #2.5.</p>	<p>Due to ⁸⁵Kr decay - the quantity available for release never significantly exceeds the amount released by 1 month (available for release - 5 Ci).</p>
<p>3.5 Moderate Direct Exposure</p> <p>Expected frequency: ~0.5 per year.</p>	<p>1. Sequence of events identical to Accident #2.6.</p>	<p>1. Safety systems identical to Accident #2.6.</p>	<p>None</p>
<p>3.6 Canister ruptures indoors or out (no confinement).</p> <p>Expected frequency: ~0.2 per year.</p>	<p>1. Canister rupture due to one of the mechanisms described in Accidents #3.4 and #3.5.</p> <p>2. Sequence of events are identical to those described in Accident #2.7.</p>	<p>1. Safety systems identical to Accident #2.7.</p>	<p>5 Ci of Kr is released (over 2 min period). A worker is located (randomly) within 10 ft of the canister at time of rupture.</p>

TABLE F.8. Severe Accidents - Immobilized Kr Loading, Transportation, Interim Storage, Long Term Storage and Decommissioning

Accident No. and Description	Sequence of Events	Safety System	Release
<p>3.7 Kr/zeolite autoclave rupture.</p> <p>Expected frequency: ~0.1 per year w/o loss of confinement.</p> <p>~0.001 per year with loss of confinement.</p>	<ol style="list-style-type: none"> 1. Zeolite loading autoclave rupture due to operator error, equipment malfunction or introduction of explosive gas mixtures. Impurities or explosive agents introduced with the zeolite is a conceivable explosion source. 2. Kr released (inventory in autoclave and feed lines) to operating cell, or operating cell is breached by explosive force and autoclave inventory is released in a single burst at ground level. 3. The undamaged operating cell atmosphere is contained and sent to the FRP for Kr recovery. 4. A fraction of the Kr inventory will be released in a controlled mode at the FRP stack. 5. The autoclave facility is decontaminated, repaired, and returned to service. 	<ol style="list-style-type: none"> 1. The autoclave process is inherently safe due to the noble process gas. 2. Overpressurization prevention by virtue of relief valves and blow out disks. 3. Weak fittings which might cause a blow out will tend to leak first. The radioactive property of Kr will provide leak detection and alarm at very small leak rates. 4. The autoclave will be located in an operating cell designed to contain an autoclave explosion. 	<p>288,000 Ci Kr inventory in autoclave assembly. Rupture w/o loss of confinement will lead to 58 KCi/day release from FRP stack (for 5 days).</p> <p>With loss of confinement 280,000 Ci is released at ground level in 4.7 minutes.</p>

- Column 4 - amount of release, the expected source term, i.e., the material specification, location, and duration of the expected potential release.

F.2 ACCIDENT ANALYSIS OF THE CRYOGENIC ^{85}Kr RECOVERY SYSTEM

The krypton recovery system will process feed gas consisting of air, small amounts of noble gases, nitrogen oxides, water and hydrocarbons (Brown, Knecht and Thomas 1978). There would be too much gas for practical temporary storage; a tank greater than 50 feet in diameter would be required for an 8-hour inventory. Thus, in event the cryogenic system is down, the feed gas must be routed to the FRP stack. To minimize down time, the corresponding loss of the ^{85}Kr process stream, two redundant cryogenic systems are assumed. The ^{85}Kr concentration in the feed gas will be the range 11 to 14 Ci/m³.

The cryogenic system liquefied gas inventory is about 20 liters which would expand to 20 m³ of gas at STP. The accident analysis assumes that a tank is available for temporary storage of this gas. This process inventory of ^{85}Kr in the column reboilers and final product column will be in the range 50 to 75 kCi (Brown, Knecht and Thomas 1978, and Bendixsen 1976).

The cryogenic system has several inherent safety features: 1) there are no explosive materials normally present in the system (e.g., natural gas, other fuels and/or flammable solvents), 2) the cryogenic components are in a cold box which doubles as a blast shield and a containment shroud which is continuously monitored for beta activity, and 3) it is remotely located in a shielded cell with ventilation confinement and control.

Potential operational and safety problems with the recovery system include: 1) a high probability of freezeups (either process gas or foreign substances: e.g., NO_x) since the boiling points and freezing points of the process gases are only a few degrees apart and the system is small and lacks significant heat capacity, 2) system leaks due to expansion and contraction, and 3) requirement for close observation and control of the oxygen recombiner. A freezeup frequency of 6 per year is based on experience with the ICPP Pilot Plant and other similarly sized units. A higher frequency is expected during startup; experienced operating crews will have fewer freezeups.

The cryogenic system minor accidents described in Table F.1 have occurred at the ICPP ^{85}Kr recovery pilot plant. The frequency of these accidents at ICPP have been somewhat higher than those listed; however, a production facility will be designed to minimize down time. The listed values should be achievable.

The oxygen recombiner explosion and the cryogenic system or cell explosion (accidents 1.6 and 1.7, Table F.2) are not expected to occur; however, they are postulated as maximum credible accidents (containment breach with prompt release of the entire system inventory). The consequences of these postulated accidents fall in the moderate category. The frequency of occurrence values listed are based on maximum acceptable frequencies; that is, if such an event occurred, the system would be modified to prevent any future occurrences.

F.3 ACCIDENT ANALYSIS FOR ^{85}Kr STORAGE IN CYLINDERS: FILLING, TRANSPORTATION, INTERIM STORAGE AND LONG-TERM STORAGE

Compressed gas cylinder technology is well established, extensively used throughout the industrial world and has an excellent safety record. Compressed gas cylinders are designed for zero leakage. Current valve designs use diaphragms to eliminate valve stem leakage. If the compressed gas is toxic or valuable, the valve is customarily capped. There are compressed gas cylinders in use which are more than 60 years old.

Compressed gas cylinders are manufactured under specifications contained in DOT regulations. Specifications for cylinders are found in 49 CFR part 178.36 to 178.68 (DOT 1978). Hydrostatic tests, flattening tests, and leak testing are required. Standard steel cylinders are tested to 5/3 service pressure; aircraft tanks are tested (1 in each lot of 200) to burst pressure which must exceed service pressure by a factor of 3. Leak testing requires under water examination while pressurized (service pressure) with air.

Gas cylinders are inspected and retested at 5-year intervals (more frequently if used for corrosives) per specification 49CFR part 173.34

(qualification, maintenance and use of cylinders). This accident analysis assumes that the gas cylinder design selected and manufactured for krypton storage will conform to the technology cited above.

The accident analysis assumes that stored cylinders will not leak, i.e., the airborne beta activity in the storage cells will be normal background. A small leak of 3.2×10^{-3} Ci/min (0.038 STP cm³/min) will be readily detected by the continuous air sampling counter, or a detectable ⁸⁵Kr air concentration of 100 dpm, a cell size of 10 x 10 x 15 ft at 10 air changes/hour. The cylinder would leak at this rate for 75 years; long after the ⁸⁵Kr had decayed. This leak rate would also be observed by water immersion inspection. Clearly, krypton activity is very sensitive to detection. In event of krypton cylinder leakage, the facility alarms and/or receiving/shipping inspection would detect leakage at rates in the range of 3.2×10^{-3} Ci/min (or less).

In practice, compressed gas cylinder leakage (and/or rupture) is a rare event and when it occurs, it is due to an operator error. No compilation of historical leak data is available per discussions with the Compressed Gas Association, 500-5th Ave., New York, NY, 10036. Krypton cylinder handling would involve less errors than would be expected in the general industry due to specialized and limited handling activity. The listed frequency of krypton cylinder minor leakage (accident 2.1) is believed to be substantially higher than would be observed in practice. A minor ⁸⁵Kr release did occur October 8, 1977, at Harrisburg, PA (NRC 1978). This incident occurred during shipment of low specific activity ⁸⁵Kr gas (5 mCi ⁸⁵Kr/l). Approximately 0.5 Ci ⁸⁵Kr was released; it was estimated that the exposed individual received a 150 mrem skin dose. The leak rate that occurred at this accident is similar to the leak rate specified in this analysis.

In event storage cell cooling is lost (accident 2.3), the cylinders will remain below the temperature required to lose strength (370°C). Heat transfer calculations (Foster and Pence 1975) indicate cylinder wall temperatures of 60°C for natural convection in room temperature air (21°C ambient; 45°C at 10 years). Storage vaults could be equipped with an optional emergency natural convection ventilation system if the safety analysis for such a facility indicates need.

Cylinder rupture is viewed as an extremely unlikely event which would only occur if a filling or handling error occurs. Periodic inspection of the ^{85}Kr cylinders should detect external corrosion potential long before leakage occurs. Internal corrosion potential will have to be handled by other means. The indoor and outdoor krypton cylinder ruptures (accident 2.7) are postulated to occur while inserting a cylinder in the vault or caisson (a shift in the loading machine results in crush or shear).

F.4 ACCIDENT ANALYSIS FOR IMMOBILIZED ^{85}Kr ; LOADING, TRANSPORTATION, INTERIM STORAGE AND LONG-TERM STORAGE

The immobilized ^{85}Kr material assumed in this analysis is $\text{Fe}_{0.79}\text{Y}_{0.12}\text{Kr}_{0.09}$, an amorphous deposit prepared by sputtering the metal in krypton gas. The krypton release from the various immobilization materials tested (Tingey et al. 1979) suggests that this accident analysis is generally valid for all material options. These materials initially outgas, releasing small amounts of the ^{85}Kr inventory in 30 days. Based on data reported by Tingey et al. (1979), 5 Ci of ^{85}Kr gas would accumulate in a canister containing 130,000 Ci, at 100°C , of immobilized ^{85}Kr . Thus, if a canister is a threshold process, the leak rate will be similar to a leaking gas cylinder. Source terms for the immobilized ^{85}Kr minor accidents are thus similar to those for the ^{85}Kr gas cylinders. The expected frequencies of immobilized ^{85}Kr storage accidents are estimates based on a review of the type of operations involved. For choosing these frequencies, it was assumed that immobilized krypton would be stored in a dry caisson field (Klett 1978) while krypton gas cylinders would be stored in above ground vaults.

Moderate accidents involving immobilized ^{85}Kr have lower source terms than corresponding accidents for gas cylinders owing to the immobilized form. Severe accidents for this material in storage do not exist. One severe accident for loading ^{85}Kr on zeolite was postulated (accident 3.7 - Kr/zeolite autoclave rupture); although it is not expected to occur. The consequences of this accident are similar to several other accidents postulated for other portions of the ^{85}Kr recovery system (e.g., accidents 1.7 and 2.7).

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