

PRELIMINARY SAFETY EVALUATION OF A COMMERCIAL-SCALE KRYPTON-85
ENCAPSULATION FACILITY*

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Abstract

This paper demonstrates that a commercial-scale facility for encapsulating krypton-85 in zeolite-5A or glass at a 2000 MTHM per year nuclear fuel reprocessing plant can be designed to contain fragments and the 340 to 800 kCi krypton-85 inventory from an assumed catastrophic failure of the high pressure vessel. The vessel failure was assumed as a worst case and was not based on a detailed design evaluation or operating experience. The process design is based on existing commercial hot isostatic pressing technology operated at up to 40 times the scale required for krypton encapsulation. From the calculated process gas inventory in the pressure vessel and vessel design, the maximum explosive energy of 8.4 kg TNT and resulting vessel plug and fragment velocities were calculated. The facility Containment Cell housing the high pressure vessel was designed to contain the gases, fragments, and the shock wave energy calculated for a hypothetical vessel failure. The Access Cell located directly above the Containment Cell was designed to be a tertiary confinement of krypton-85, should the access hatch be breached.

I. Introduction

Krypton-85 is formed in moderate yield during nuclear fission of uranium or plutonium.¹ Most of it is trapped in the spent fuel and will not be released until the fuel is dissolved during reprocessing.¹ However, Federal regulations prohibit release of more than 50000 Ci of krypton-85 per gigawatt-year of electric power produced from nuclear fuel irradiated after January 1, 1983.² Thus krypton-85 must be recovered during fuel reprocessing,³ and approximately less than 15% of the krypton-85 produced in light water reactors can be released during recovery and storage.²

One of the possible storage methods involves krypton encapsulation in zeolite 5A and low density glass at pressures near 2000 atm and respective temperatures of 600 and 950°C, producing volumes of

* Work performed under USDOE Contract DE-AC07-791001675.

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immobilized krypton equal to volumes of krypton pressurized at about 30 atm.⁴⁻⁶ Leakage rates were estimated to be less than 1% in ten years at storage temperatures of 300°C for zeolite 5A, and low density glass.⁴⁻⁶ Hot isostatic pressing (HIP) technology, which has been developed recently to fabricate ceramic and metal alloy forms on a commercial production scale using active volumes of 30 to 2000 L and pressures and temperatures of 2000 atm and 1400°C, respectively, is directly applicable to the krypton-85 encapsulation process, using less than 50 L active volumes.^{4,7,8,9}

High pressure systems contain a large amount of potential energy in the form of compressed fluids. Catastrophic failure of high pressure components, such as vessels, can result in damage to the surroundings as the potential energy is converted to kinetic energy of fragments.^{10,11} Vessel design, maintenance, and routine inspection are used to help prevent catastrophic failure from occurring.¹¹⁻¹⁴ Barricades are generally used to contain energetic fragments and shock waves and to protect the rest of the facility.^{11,15}

This paper will evaluate the consequences of a worst-case accidental rupture of a pressure vessel used to encapsulate krypton-85 at a commercial fuel reprocessing facility. It will demonstrate that it is feasible to ameliorate the effects of catastrophic vessel failure and to contain vessel fragments and krypton-85 in the facility.

A preliminary design of the encapsulation facility and the effects of a maximum credible accident will be described. The design requirements of the barricade and Containment Cell which will prevent release of krypton-85 will be given. Such requirements can be met within existing high pressure technology.

II. Description of a Commercial-Scale Krypton-85 Encapsulation Facility

Basis for a Commercial-Scale Facility

The reference commercial spent fuel reprocessing plant is assumed to be the one described in the draft environmental impact statement on waste management, with the exception that 100% krypton-85 recovery efficiency instead of 90% is assumed, to provide a conservative estimate.^{16,17} The commercial-scale encapsulation facility will encapsulate 18.7 MCi per year of krypton-85 in zeolite 5A or glass, assuming that the facility operates at 110% of capacity for 300 days per year. The probable krypton-85 compositions obtained from cryogenic distillation and liquid fluorocarbon krypton-85 recovery systems can each be encapsulated.

Process Description

A simplified schematic of the encapsulation process is shown in Figure 1; more detail can be found in ENICO-1011.⁴ The major equipment includes an internally heated pressure vessel similar to a HIP vessel, compressors, and vacuum pumps. The pressure vessel is

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filled with a capsule containing the solid encapsulation substrate (zeolite 5A or glass), pressurized with krypton-85, and heated for a time sufficient to load the solid with krypton. After the encapsulation time is reached, the remaining krypton-85 is recycled to the storage containers using the compressors and vacuum pumps. The vessel is then opened, and the encapsulated sample removed in its container and placed in interim storage prior to removal to a permanent storage facility. A fresh batch of zeolite or glass is placed in the vessel and the process repeated. Based on laboratory studies, the process can be completed at a rate of one or more batches per day.⁴⁻⁶

Facility Design

The preliminary design for a krypton-85 encapsulation facility is shown in Figure 2; more detail can be found in ENICO-1055⁷. The building consists of three levels: a ground level, a first basement level, and a second basement (or subbasement) level. The pressure vessel and barricade are contained in an air-tight cell - the Containment Cell - in the second basement level; all of the other process equipment is located in the first basement level. The ground level is used for access to the process cells and for offices and support laboratories.

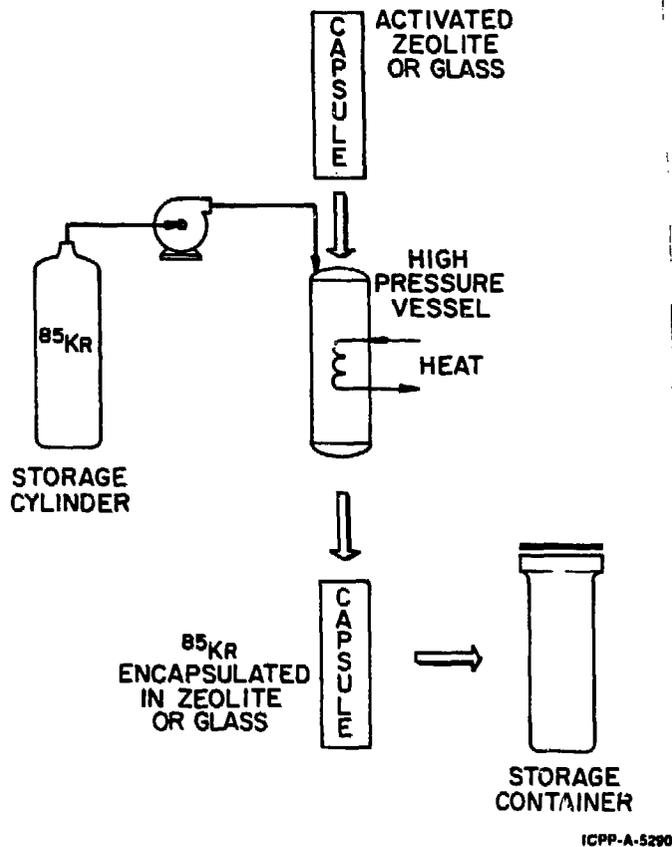


Figure 1. Simplified schematic of krypton-85 encapsulation process.

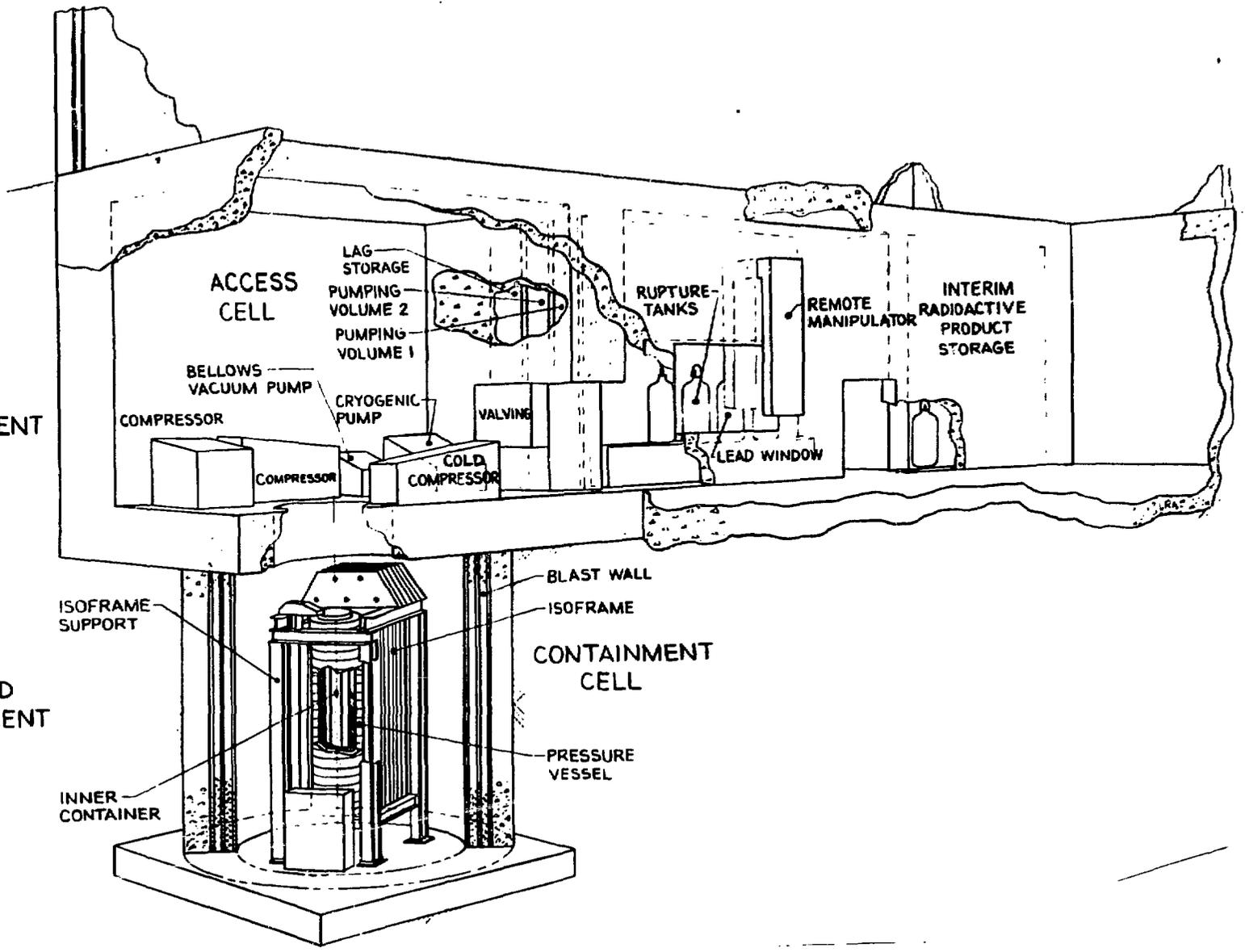


Figure 2. Preliminary isometric lay-out design of a commercial-scale krypton-85 encapsulation facility.

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III. Preliminary Safety Evaluation

This preliminary safety evaluation assumes that the worst possible consequence of operating a krypton-85 encapsulation facility would be the catastrophic release of krypton-85 to the surroundings. Based on the facility design shown in Figure 2, a number of potential sources of krypton-85 release exist in the facility. A detailed safety analysis of the probability of krypton-85 release from each of the sources has not been made. It is assumed that the high pressure vessel is the major source of a large, catastrophic release of krypton-85 because of the large inventory of krypton-85 in the vessel during an encapsulation run and because of the possibility that a vessel rupture could produce high energy fragments which could damage the facility and thus lead to release to the environment.

Postulated Maximum Credible Accident

Based on this preliminary assessment, the maximum credible accident for a commercial-scale krypton-85 encapsulation facility is assumed to be catastrophic rupture of the high pressure vessel during an encapsulation run. Modes of pressure vessel failure include: 1) failure of the plug or closure mechanism, 2) pressure vessel fracture in a radial plane (perpendicular to the longitudinal axis) producing two large fragments, and 3) pressure vessel fracturing into equal longitudinal strip fragments. The most severe effects will be used to design a containment barricade.

A detailed design study combined with a periodic inspection program of an operating vessel for a commercial-scale encapsulation system may show that the probability for vessel rupture is so low that it is not credible.¹²⁻¹⁴ If other accidents are found to have a more significant risk than vessel rupture, some of the facility design criteria required to contain krypton-85 from vessel rupture should also be sufficient to contain krypton-85 from another source.

Critical Process Component

Based on the postulated maximum credible accident of catastrophic vessel rupture, the critical process component is the 318.6-L pressure vessel, shown in Figure 3. It is located in the Containment Cell of Figure 2, and uses a balanced pressure concept: the strength-bearing wall of the outer vessel is pressurized with an inert, non-radioactive gas, such as He or Ar, to balance the pressure of krypton-85 in the 50.5-L inner vessel.¹⁸ Using standard HIP technology, the inner vessel containing the zeolite or glass and krypton-85 is heated to the encapsulation temperature of 500-1000°C, while the strength-bearing outer vessel is kept at less than 200°C. The tubing connection to the inner vessel would be closed remotely by crimping and welding after an encapsulation run, and the inner vessel could become the final storage canister.

Consequences of Postulated Maximum Credible Accidents

Using the design of the pressure vessel shown in Figure 3, it is possible to calculate the amount of gas contained in the vessel during an encapsulation run, the explosive pressure-volume energy of the compressed gas, and the resulting missile velocity, shock wave energy, and equivalent static pressure after a failure.

Amount of Compressed Gas in Vessel. The volume surrounding the inner canister contains argon gas and the furnace, consisting of a heater and a heat shield. Since the inner vessel is heated to the run temperature and the outer pressure vessel wall is kept below 200°C, the argon gas is at some intermediate temperature. Assumptions of the average temperature can be made for various regions of the vessel.⁷ The inner canister contains zeolite 5A spheres or porous glass rods, 1 cm in diameter, with the respective void volumes of 25 L at 500°C and 13 L at 1000°C. The amounts of argon and krypton-85 in the pressure vessel during an encapsulation experiment at 500°C for zeolite 5A and 950°C for glass and 2000 atm can be calculated using the above assumptions and the Redlich-Kwong equation of state and are shown in Table I.^{4,19,20}

Table I. Results of Calculations of Gas Compression Energy^a

	<u>Glass Encapsulation</u>	<u>Zeolite 5A Encapsulation</u>
p_1 , ^a atm	2000.	2000.
v_1 , cm ³ /mole	55.1	47.5
T_1 , K	782.	632.
p_2 , atm	1.3	1.2
v_2 , cm ³ /mole	1800.	1200.
T_2 , K	40.	35.
Argon, m ³ at STP	111.0	130.1
Krypton, m ³ at STP	3.8 ^b	9.5 ^c
Total Gas Volume, m ³ at STP	114.8	139.6
ΔU , MJ	37.8	29.3
ΔU , kg TNT	8.4	6.5

a Subscripts 1 and 2 refer to initial and final states, respectively.

b Assuming a 13-L void volume in the 50.5-L inner vessel; this is equivalent to 340 kCi of 6% ⁸⁵Kr in krypton.

c Assuming a 25-L void volume in the 50.5-L inner vessel; this is equivalent to 850 kCi of 6% ⁸⁵Kr in krypton.

Explosive Characteristics of Vessel Rupture. Using the vessel design shown in Figure 3 and the calculated volume of gas in the vessel at high pressure, the explosive energy, ΔU , of the vessel rupture can be calculated by assuming that the gas expands reversibly

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and adiabatically to atmospheric pressure. The initial and final values of pressure, volume, and temperature as well as the calculated energies are shown in Table I.^{11,21} The details of the calculations are shown in ENICO-1055.⁷

The maximum total energy which can be released from the compressed gas was calculated to be 8.4 kg TNT by assuming the conditions corresponding to glass encapsulation.¹¹ If Zeolite 5A encapsulation is assumed, the value is 6.5 kg TNT. Since the safety code developed by the High Pressure Technology Association states that above 100 atm considerable error in the energy results from assumption of the ideal gas behavior,¹¹ the Redlich-Kwong equation of state was assumed in the calculation. Using the methodology in reference 21, 5.7 to 22.2 kg TNT is obtained.²¹ The Sandia Pressure Safety Manual uses the real gas properties of argon for calculating the 8.1 kg TNT energy, but assumes argon to be at room temperature.²² Thus, the value used in this report is 37.8 MJ or 8.4 kg TNT, which is calculated for the vessel design in Figure 3, based on real gas properties of argon.

Facility Design Required to Ameliorate Effects of Maximum Credible Accident

The design of a commercial-scale krypton-85 encapsulation facility must include engineered safety features to contain fragments and krypton-85 released from the pressure vessel rupture assumed as the maximum credible accident. The vessel must be located within a sealed Containment Cell with provisions for recovery of released krypton-85. The Containment Cell must itself be protected from fragment or blast damage. Secondary restraint on the vessel closure by a passive yoke structure around the vessel would further limit fragment damage and would make launching of the plug an incredible event.

The calculations of fragment energy and shock energy for several modes of fragmentation are shown in Table II. The facility design features required to ameliorate the explosive effects described in the following section are based on the values shown in Table II. Detailed calculations are given in ENICO 1055.⁷

When a small number of fragments is formed, most of the energy is removed by the kinetic energy of the fragment. When a large number of fragments is formed, most of the energy is removed by the shock wave energy. Thus the Containment Cell must be designed with wall and missile shield thicknesses which are large enough to contain both a launched plug fragment and the maximum shock energy obtained from a multiple fragmentation mode.

Pressure Vessel. The pressure vessel (Figure 3) is a multi-walled shrunk-fit 318.6-L vessel with threaded end closures containing a 50.5-L active volume. This type of vessel is commonly used in commercial high pressure and temperature applications, such as Hot Isostatic Pressing.^{8,9} The vessel contains a heater/heat shield package in the annulus between the inner wall of the pressure vessel and the inner vessel containing the zeolite or glass.

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Table II. Explosive Energy of Vessel Fragmentation^a

	Fragmentation Mode			
	<u>Plug</u>	<u>2 Parts</u>	<u>10 Parts</u>	<u>100 Parts</u>
Fragment Quantity:				
Mass, kg	548	5933	1186	119
Area, m ²	0.15	0.49	0.49	0.49
Kinetic Energy, MJ	22.7	11.3	0.76	0.08
Velocity, m/s	288	61.8	35.7	35.7
Concrete Thickness, m	1.6	0.55	0.04	0.004
Steel Thickness, m	0.13	0.05	0.003	0.0003
Shock Wave Energy, MJ	15.1	15.1	30.2	30.2
Equivalent Static Pressure				
in Containment Cell, ^b atm	3.6	3.6	5.9	5.9
Equivalent Static Pressure				
in Containment Plus Access				
Cells, ^b atm	0.7	0.7	1.1	1.1

^a See ENICO-1055 for details of the calculations.⁷

^b Equivalent static pressure is the pressure measured relative to atmospheric pressure which, when maintained indefinitely in the contained volume, will produce the same deflections in the walls as the shock wave.¹¹

Penetrations for gas lines and instrument and electrical leads are located on the bottom closure plug. The internal volume and pressure-bearing wall of the pressure vessel are instrumented with pressure and temperature sensors to provide indication and/or alarm for operating conditions.

Safety Yoke and Support. The safety yoke shown in Figure 3 is a massive band structure that surrounds the pressure vessel during high pressure operation. The yoke provides secondary closure force in the event of failure of the threaded closure, and its sides also provide missile barriers on two sides of the vessel.

The vessel and yoke are mounted on a support structure which has rails for moving the yoke out of position for loading and unloading the vessel. Microswitches and interlocks are provided to prevent pressurization of the vessel when the yoke is not in the operating position. A hydraulic system is used for remote positioning of the yoke.

Calculations of the static and dynamic load on the yoke which would result from plug failure give safety factors, based on the ultimate strength, of 7.4 for the static load and 1.4 for the dynamic load assuming ASTM A-242 as the yoke material.

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Containment Cell. The Containment Cell shown in Figure 2 is a cylindrical cell, 3.7 m diameter by 5.5 m high, constructed of 1.6-m thick reinforced concrete with an airtight steel liner and is located in the second basement level. Access to the pressure vessel for loading and unloading the zeolite or glass container is through a sealed overhead cell access cover located in the Access Cell in the first basement level. The Containment Cell will contain any krypton-85 released. Piping to and from the cell is used for purging the cell and recovering krypton-85.

The values of fragment mass and velocities and shock wave energy resulting from pressure vessel rupture shown in Table II were used to design the concrete thickness of the walls, floor, and ceiling shown in Figure 2. The access cover requires twelve 3.8-cm diameter bolts to contain the shock wave and equivalent static pressure.

Explosive Barrier and Missile Shield. The explosive barrier is a cylindrical composite structure which lines the cell wall, shown in Figure 2 and surrounds the pressure vessel.^{2,3} It is constructed of screen materials sandwiched between perforated steel plates and is designed to suppress a 20-kg TNT blast. Structural angles mounted on the inside and outside walls serve as missile and shock wave deflectors. The missile barricade also serves as a shock wave suppressor. This type of barricade is a safety-approved suppression shield for fragment containment, blast suppression, and flame attenuation.^{2,3}

The 13-cm steel missile shield located above the high pressure vessel (Figure 2) is suspended from the ceiling of the containment cell and protects the cell liner from missiles resulting from vessel rupture. The 1.6-m thick concrete ceiling wall is also thick enough to stop the vessel plug, if necessary.

Quality Assurance. The development of a Quality Assurance Program Plan is essential for control of design, material fabrication, testing, and operation of the krypton-85 encapsulation facility. The plan should define responsibilities for establishing requirements and assuring compliance with the requirements.

Design, fabrication, and inspection of the pressure vessel should be in accordance with the rules of the ASME Boiler and Pressure Vessel Code Section III or Section XIII Division 2. Other applicable codes should also be consulted.^{1,1}

An explosive model test program should be carried out to determine or verify vessel failure modes, fragmentation, missile velocities, and adequacy of barriers and containment designs.

IV. Conclusions

This paper demonstrates that a commercial-scale krypton-85 encapsulation facility can be designed to contain fragments and the 340-850 kCi krypton-85 inventory from a hypothetical catastrophic failure of the high pressure vessel.

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The pressure vessel, containing an active volume of 50 L, is designed to encapsulate the 17 MCi annual krypton-85 production from a 2000 MTHM commercial spent fuel reprocessing plant in 300 days/yr, assuming 110% production overcapacity. The vessel design is based on existing commercial hot isostatic pressing vessel technology in which the active volumes range from 30 to 2000 L.⁸ Although vessel rupture is an unlikely event if modern design and inspection procedures are used,¹¹⁻¹⁴ it is assumed to be the maximum credible accident for the evaluation in this report.

The facility, which is designed to contain vessel fragments and krypton-85 from a hypothetical vessel rupture, consists of one above-ground and two underground levels of cells. The process vessel is located in a sealed second basement level Containment Cell, which is designed to be a secondary containment for the process gases, argon, krypton, and krypton-85. The first basement level houses process equipment such as manipulators, cranes, compressors, vacuum pumps. Feed gas storage, and interim product storage for encapsulated krypton-85 are also on this level. The Access Cell, which contains the access to the second basement level Containment Cell, is also sealed and is designed to provide tertiary containment of the krypton-85 if the connecting wall to the Containment Cell is breached. The above-ground level houses offices, a support laboratory, and access hatches to the basement cells.

The inventories of argon and krypton-85 in the high pressure vessel during an encapsulation run would produce a maximum explosive energy in the Containment Cell of 37.8 MJ or 8.4 kg TNT. The explosive barrier is designed to suppress a 20 kg TNT blast.²³ The 1.6-m thick ceiling and the bolted hatch cover of the subbasement Containment Cell are designed to contain the 30 MJ or 6.7 kg TNT shock wave.¹¹ A 13-cm thick steel missile barrier and support structure above the pressure vessel is the required size to contain a launched 548-kg vessel plug at a velocity of 288 m/s estimated for plug failure.¹¹ However, a yoke surrounding the vessel would make plug launching an incredible event. Other smaller fragments would be contained by the surrounding explosion barrier and the second basement level cell walls supported by the surrounding ground.

After a more detailed design of the krypton-85 encapsulation facility is made, further safety evaluations should be carried out to identify all accidents and determine probability and consequences of each. Such an evaluation may show that vessel rupture is not a credible accident and that facility design requirements to contain krypton-85 from the maximum credible accident will not be as stringent. However, the preliminary evaluation shown in this report should give a first approximation to the design requirements for a commercial-scale krypton-85 encapsulation facility.

Acknowledgment

The assistance of R. C. Green, L. H. Jones, and J. P. Sekot in the preparation of this report is gratefully acknowledged.

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Work supported by the U.S. Department of Energy Assistant Secretary Energy Technology, Nuclear Waste Management under DOE Contract Number DE-AC07-79ID01675.

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