70-687

PDR

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Office of Nuclear Material Safety & Safeguards
United States Nuclear Regulatory Commission
Washington, D. C. 20555

License Amendment Request to
Permit Uranium Waste Form Process
License No. SNM-639 Docket No. 70-687

References:

1. License Amendment Request to Permit Uranium Recovery from Waste, letter from M.H. Voth (Union Carbide - Medical Products Division) to W.T. Crow (U.S. Nuclear Regulatory Commission - Uranium Fuels Licensing Branch), December 28, 1979.

2. Nuclear Safety Guide, TID-7016, Revision 2, edited by J.L. Thomas, June 1978.

3. Critical Dimensions of Systems Containing U-235, Pu-239, and U-233, TID-7028, H.C. Paxton, et. al., June 1964.

4. D. L. Summers and M. C. Gashe, Health Physics Journal, Volume 4, Number 3/4, Page 289, 1961.

5. License Amendment Request to Permit Uranium Recovery from Waste, letter from M. H. Voth (Union Carbide - Medical Products Division) to W. T. Crow (U. S. Nuclear Regulatory Commission - Uranium Fuels Licensing Branch), April 2, 1980.

6. Letter from L. C. Rouse (U. S. Nuclear Regulatory Commission) to M. H. Voth (Union Carbide), May 22, 1980.

Preface

An application for an amendment to license SNM-639 on the above subject matter was submitted on December 28, 1979 (Reference 1). Following preliminary review by the NRC Staff and discussion on nuclear criticality safety, it was resubmitted, incorporating agreed-upon changes (Reference 5). Additional information was requested (Reference 6) which has been incorporated into the application. This application supercedes both previous applications in their entirety. For ease of review, substantive changes from the previous version (Reference 5) are sidelined.

FEE EXEMPT

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Introduction

In the production and separation of medical radioisotopes, a substantial amount of unused fissile target material remains with the radioactive wastes. The present practice of disposing of the enriched uranium wastes is an unnecessary burden on our nations radioactive material waste burial sites, an unnecessary waste of a vital natural resource, and an unnecessary expense ultimately affecting health care costs. An amendment is hereby requested to license SNM-639 which will allow implementation of a uranium waste form process into the existing waste handling process. The uranium waste form process is simply a conversion of the uranium and other fission products in the normal waste solution from a sulfate to an oxide form which is compatible with the Savannah River uranium reprocessing operation. At the same time, the option for using the present disposal process is retained.

Amendment Request

The requested amendment to license SNM-639 consists of simply adding a reference to this letter, making paragraph 9 read as follows:

> "9. The special nuclear material is for use in accordance with the statements, representations, and conditions specified in the license's applications dated April 28 and May 21, 1969; November 5, 1970; February 8, June 13, June 29, and August 13, 1973; May 28, 1974; February 11, 1975; August 12, 1976; May 3, October 13 and November 17, 1978; and June 2, 1980."

Clarification of Existing License Conditions

Wastes are presently solidified in 5" diameter cylinders which are placed in 55 gallon drums awaiting off-site burial. Present limits are 200 grams U-235 per 5" cylinder (restricted to storage in a linear array) and 350 grams U-235 per drum, subject to a limit of 2000 grams total U-235 per waste storage cell. There is no stated limit on the number of such waste storage cells in our facility.

The May 3, 1978 letter, referenced in paragraph 9 of the license, includes a figure showing a typical arrangement of drums stacked one high in a waste storage cell. A Region 1 NRC Inspector questioned our practice of storing drums two high, noting that without specific authorization to do so the figure would suggest that we are restricted to storage in a single layer. We agreed to address the subject in our next amendment request.

The safety analysis of fissile material storage in 55 gallon drums shows that a closely packed, multi-layered array of drums remains subcritical when the array is restricted to 2000 grams U-235 with no more than 350 grams per drum. Unless directed otherwise in your action on this amendment request, we will continue to interpret our license as permitting storage of drums in tightly-packed, multi-layered arrays in each waste storage cell.

Discussion of Newly Requested License Conditions

This amendment request defines and analyzes a Uranium Waste Form Process Cell (UWFPC). Raw fission waste solution will be processed in 200 gram U-235 batches and stored in the UWFPC. The end result of the process will be oxidized uranium powder and some incidental mixed fission products sealed in a 3" diameter cylinder. The liquid remaining from the process along with the remaining mixed fission products and trace quantities of uranium will be solidified as is presently done, either in the UWFPC or another cell. The 3" diameter cylinders will be stored in an approved array in the UWFPC awaiting shipment to a reprocessing facility.

The uranium waste form process is described in Appendix A. The typical layout of a UWFPC, including the process equipment and the storage configuration, are discussed in Appendix B. The criticality safety analysis of the composite UWFPC, which supports the specific license conditions, is included as Appendix C. The safety analysis of the uranium waste form process is included as Appendix D. Environmental considerations are discussed in Appendix E.

SPECIFIC ADDITIONAL LICENSE CONDITIONS

The following specific license conditions are designed to give wide margins of safety and, at the same time, maximum flexibility in operations by evaluating bounding conditions and placing appropriate restrictions on the key parameters, allowing process changes to be made within the envelope of the safety analysis and the license conditions:

1. Each Uranium Waste Form Process Cell (UWFPC) shall contain no more than 7600 grams of U-235, $\leq$ 7200 grams being allowed in oxide storage cylinders and $\leq$ 400 grams in process.

2. Supporting and storage fixtures in a UWFPC shall be of substantial structural integrity to preclude a change in geometry under normal operating conditions and credible accident conditions. Sources of pressurized liquid, which could provide moderating material to the storage array, are not allowed in the cell.

3. Oxide storage cylinders shall have a maximum inside diameter of 3" with no restriction on height. Each cylinder shall contain no more than 200 grams U-235 and have a H/U less than or equal to 20.

4. Oxide storage cylinders may be stored two high, centered in a planar unit cell, $\geq$ 12" x 12", which does not include the concrete cell wall or another unit cell.

5. Material in process in each UWFPC shall be restricted to two batches having a maximum fissile material content of 200 g U-235 per batch. Containers used shall be no more than 4 liters each, arranged in a linear array. The separation of selected fission products is permitted provided it is done in sub-batches of $\leq$100 gm U-235 each. The conversion of the sulfate waste stream to an oxide form (wherein incident to the process a mixture of the fission products may be carried over while others may not) does not constitute a separation of selected fission products.

6. A minimum planar unit cell for uranium in process shall be defined as that area enclosed by lines drawn 20" on either side of and parallel to the centerline of the linear array of process containers and 14" from the center of mass of the two end batches and perpendicular to the centerline of the linear array of process containers. This unit cell shall not include the concrete cell wall or another unit cell.

Conclusion

The requested license amendment allows for the conversion of uranium wastes to a form acceptable for reprocessing by Savannah River. Gross conservatisms are included in the license conditions to provide flexibility, ease of analysis, and potential for increasing quantities through another license amendment at a later date without changing geometries.

Pursuant to 10 CFR Part 170, a license amendment fee was submitted with Reference 1. Note that the materials and plant protection evaluation of this process has already been addressed in Amendment MPP-3 to License SNM-639, which was issued on January 30, 1979.

We consider the uranium waste form process to be a significant step in relieving our nation's radioactive waste disposal problem, especially as it affects the medical community. We, therefore, request an expeditious review of this license amendment application.

Yours very truly,

Marcus H. Voth

Marcus H. Voth
Manager, Nuclear Operations

cc: L. C. Rouse, U.S.N.R.C.

APPENDIX A

DESCRIPTION OF URANIUM WASTE FORM PROCESS

Fission product Mo-99 is produced by irradiating targets containing fully enriched uranium. Waste solutions, generated by this procedure, consist of uranium and mixed fission products dissolved in a dilute (~2 N) sulfuric acid containing ~2% nitric acid. The gaseous fission products (Xe, Kr, I_2) are removed during the fission product Mo-99 process. Each process creates approximately 150 ml of such waste, which is then stored in a borosilicate glass bottle and labeled with a sequential process number, for further disposition. Since only a small portion (~1%) of the U-235 present in each target tube fissions during irradiation, each of these bottles may be assumed to contain essentially the same amount of U-235 as was originally present in the targets. The basic process steps are listed below and shown in Figure A-4. Figures A-1 through A-3 show the processing equipment dimensions and arrangment inside the hot cell, along with the minimum unit cell boundary defined by the proposed license conditions; however, the license conditions are governing regarding these matters. Process steps include:

1. Combine the contents of borosilicate waste bottles containing up to 100 g U-235 in Container A. Repeat for Container B.

2. Precipitate ruthenium from the raw fission waste solution as a sulfide at 90°C by the use of thioacetamide added to Container A and B. This adds a volume of approximately 25 ml to the 750 ml of waste solution in each flask. (This is a precautionary step; its need will be evaluated during the startup program. If it is found unnecessary it will be eliminated.)

3. Precipitate the sulfates from the raw fission waste solution at 90°C by the use of barium acetate solution added to Containers A and B. This adds a volume of approximately 650 ml to the 775 ml of waste solution in each flask. (Steps 2 and 3 may be reversed or done simultaneously.)

4. Decant and filter the solution to remove the $BaSO_4$ precipitate (Container A to C or B to C).

5. Measure the filtrate volume and take a sample for assay (Container C).

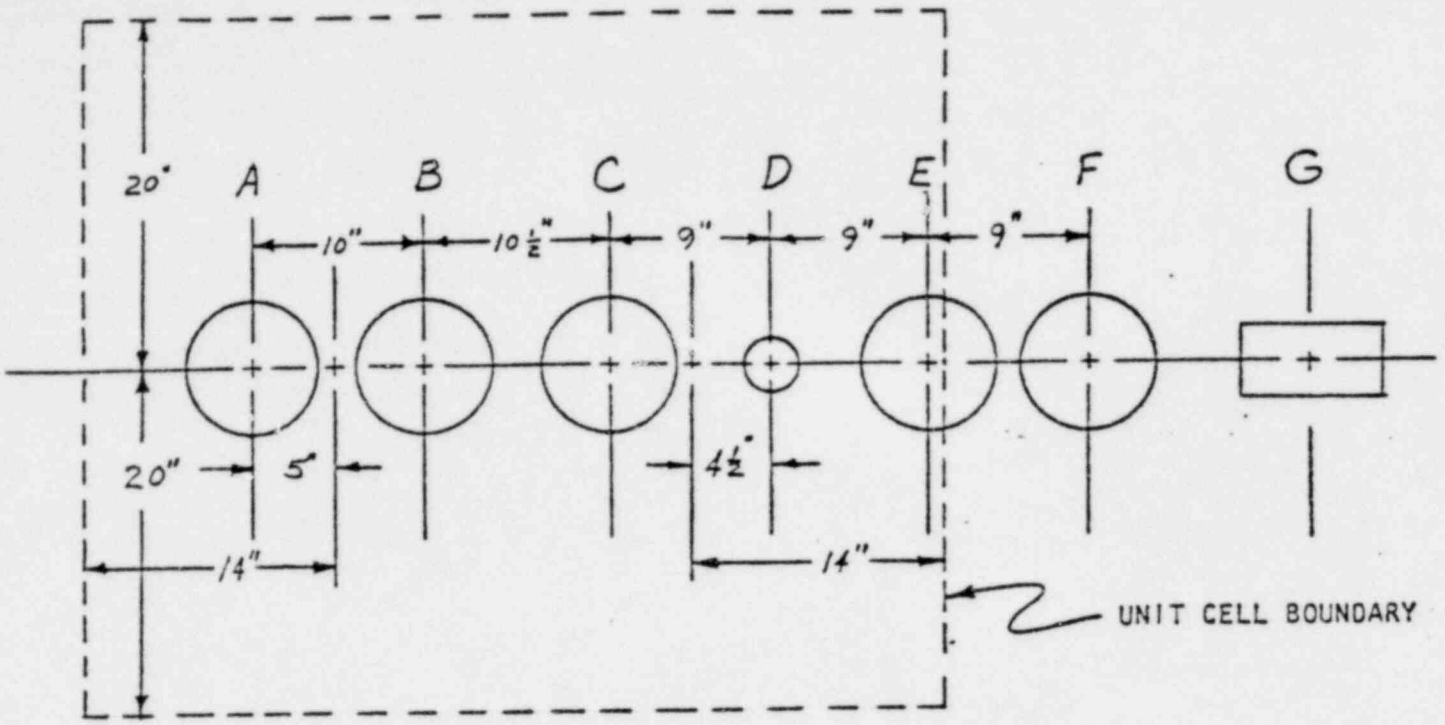
6. Transfer the solution to an aluminum can placed in an electrically heated furnace and heat to dryness. The vacuum differential distillation is run at approximately 50°C. At the end of the vacuum drying step, the temperature of the dry Uranyl Acetate will be approximately 280°C. (Heating done in Container D with distillate passing to containers E and F.)

7. Continue to raise the temperature of the dry power to calcine the uranium to a final temperature of 320°C. During this stage, the imput and output of the aluminum can (Container D) are vented through glass wool particulate traps to the hot cell atmosphere.

8. Weigh the aluminum container to determine the net weight of mixed uranium oxides content (Container D).

9. Seal the aluminum container and store for subsequent shipment to the reprocessing facility (Container D).

10. Dispose of the precipitate sludge (Container A and B) and the distillate (Container F) by solidifying in concrete for burial.



A. Collection/Precipitation - 4 liter flask
B. Collection/Precipitation - 4 liter flask
C. Filtrate - 4 liter flask
D. Uranium Container
E. Condenser
F. Distillate - 4 liter flask
G. Vacuum Pump

FIGURE A-1

ARRANGEMENT OF COMPONENTS IN HOT CELLS

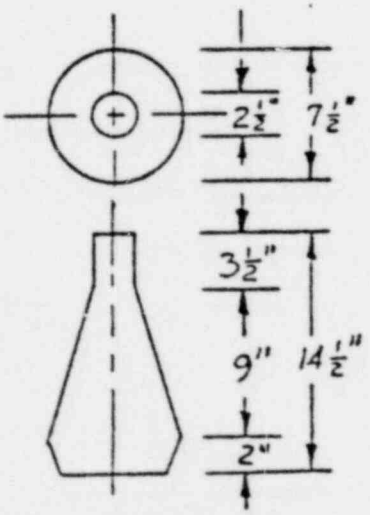


FIGURE A-2

FOUR LITER FLASK

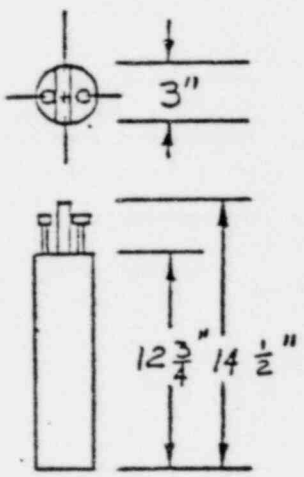


FIGURE A-3

URANIUM CONTAINER

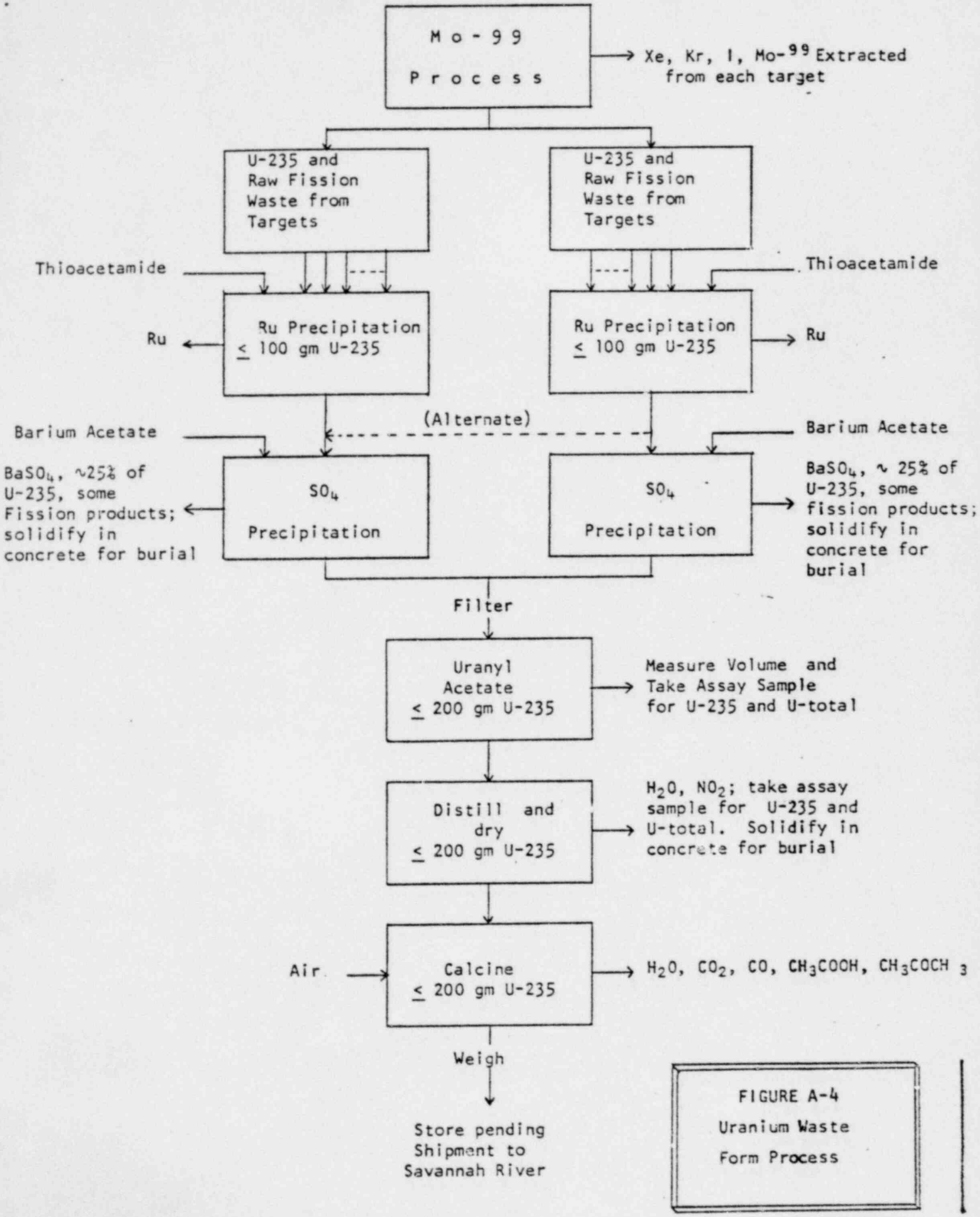


FIGURE A-4
Uranium Waste
Form Process

APPENDIX B

TYPICAL UWFPC LAYOUT

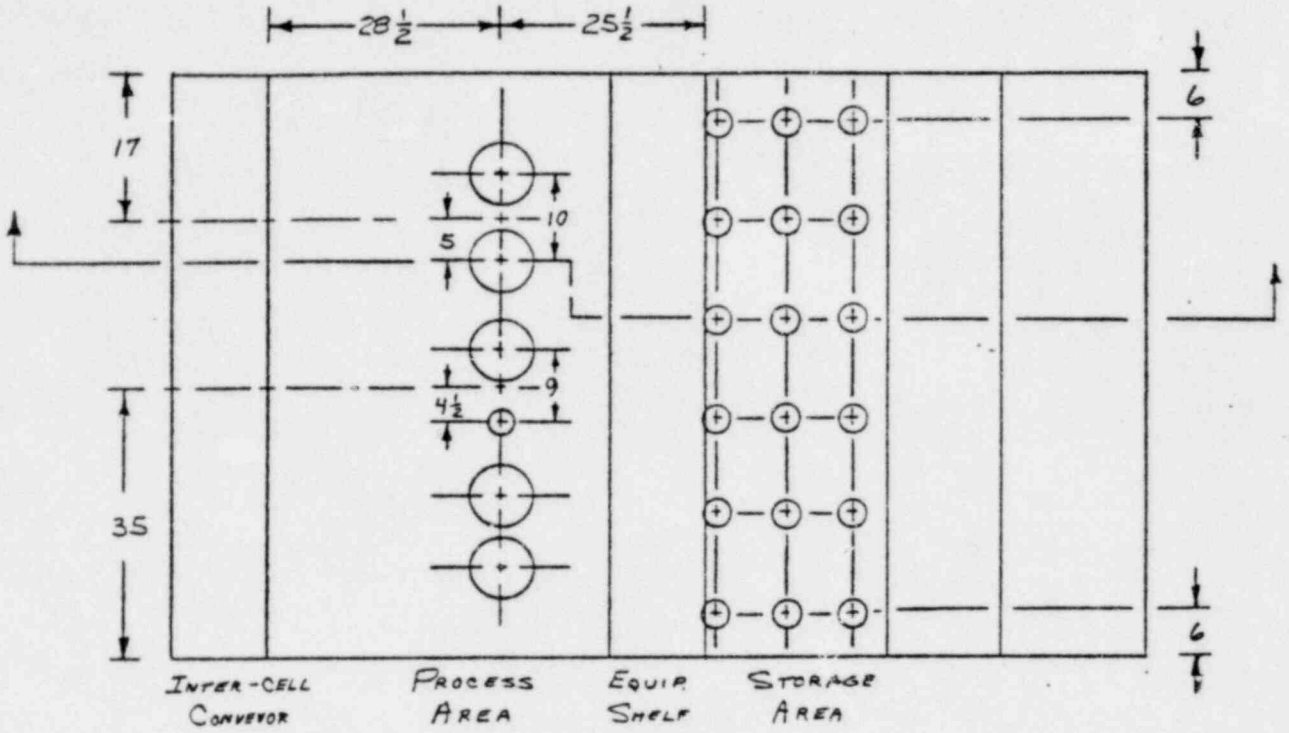
To better visualize the general license conditions stated in the foregoing letter and the supporting safety analysis presented in Appendix C, a typical Uranium Waste Form Process Cell (UWFPC) apparatus is shown in Figure B-1 and is discussed in this section. Figure A-1 shows the minimum proposed license dimensions of a typical unit cell boundary for material in process. A standard 6' wide x 10' deep x 10' high hot cell is divided into two distinct zones, one for processing and another for storage.

For improved visibility and ease of operation, the storage rack is on an incline. Since the safety analysis treats the entire UWFPC as a planar array, representing individual mass units as spheres, the minimum separations are reflected onto an imaginery plane which is perpendicular to the storage rack tubes. The following comparison of minimum allowable to actual separation shows that in all cases the minimum criteria are satisfied:

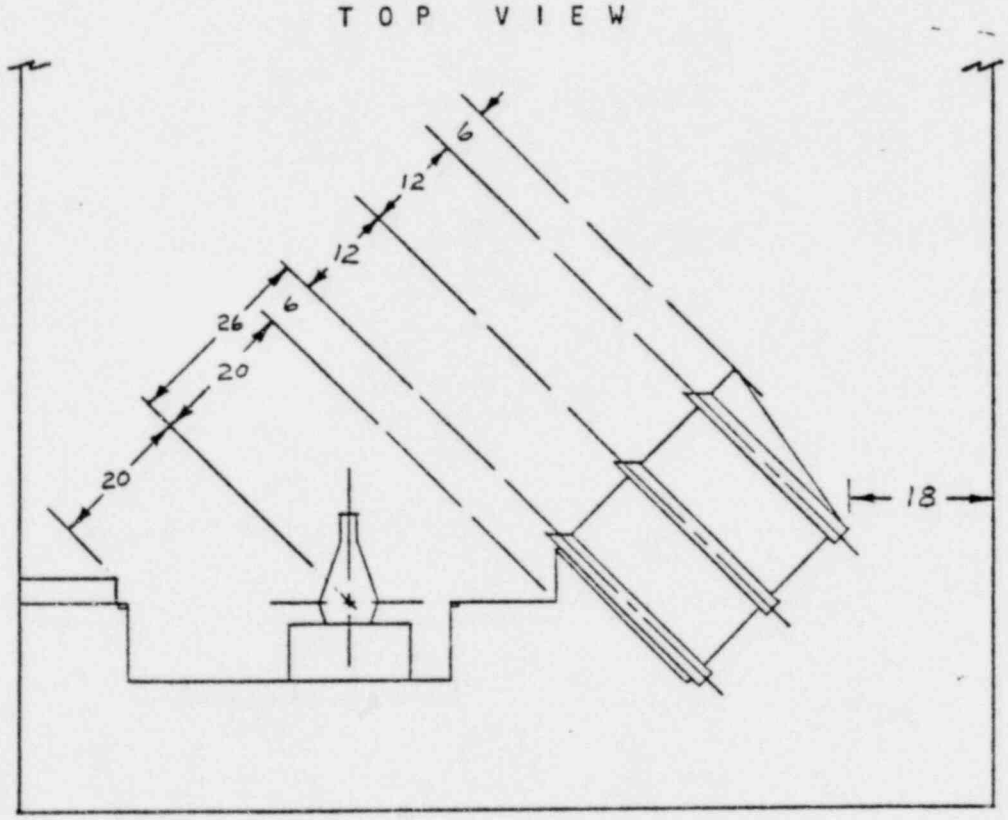
Parameter	Minimum Allowable Separation	Actual Separation
3" uranium cylinders in storage (centers)	12"	12"
3" cylinder (center) to cell walls	6"	6" sides
		18" back
3" cylinders to process containers (centers)	26"	26"
process containers (center) to edge of UWFPC (intercell conveyor)	20"	24"
Two end processing containers (center) to cell wall	14"	17" left
		35" right

Each vessel which holds material in process is fixed in a rigid metal structure. Since material in solution is vacuum pumped from one container to another there is no need for containers to be moved during a process. The support fixtures stand in a stainless steel pan. In the unlikely event that one or more of the fixtures fails, the contents will be confined to the horizontal 30" x 60" pan which by design is a less reactive array than either the process containers or the sphere assumed in the safety analysis. Another precaution taken in the equipment design is a catch basin which holds each of the glass flasks. Should a flask leak or rupture, the catch basin will contain the contents of the flask in approximately the same geometry.

The storage rack for 3" cylinders consists of 36 storage tubes on 12" centers. The diameter and fabrication tolerance of storage tubes is such that the 3" cylinder will be centered within 1/4" of the cell center. Despite the fact that flooding is not considered a credible event, the tubes are designed with drain passages that provide a greater drainage area than the area available for in-leakage of liquid. The upper grid plate of the storage rack serves to confine the tubes to the 12" centers, keep moderating material from being placed between storage tubes, and prevent other containers from being suspended in the storage array. In the same manner, a lower grid structure maintains the required spacing of the bottoms of the storage tubes. The structure is supported and braced so as to support the dead weight of the fully loaded rack, plus the combined operational and accidental forces that could possibly exist in the cell with a margin of safety greater than 3.



TOP VIEW



SECTION VIEW

FIGURE B-1. Typical Uranium Waste Form Process Cell Layout

APPENDIX C

CRITICALITY SAFETY ANALYSIS

General Approach

The criticality safety analysis uses the surface density analog presented in Section 4.23 of Reference 2. The individual mass units are assumed to be spherical masses in an ordered array on a single plane. Each mass unit is centered in a planar unit cell with the boundaries of adjacent unit cells touching. This method constitutes a conservative treatment since the third dimension is ignored. In the actual geometry, the third dimen- sion provides a great deal of dispersion and, therefore, a reduction in the reactivity of the array.

Numerous factors of conservation are built into the safety analysis for the following reasons:

- To give a wide margin of safety
- To allow for contingencies
- To make simplifying assumptions, thereby avoiding lengthy calculations or complicated experiments.
- To provide margins that, at a later date, can be converted to increased limits in the existing geometries with the appropriate analysis and license approval.

Table C-1 is a summary of the proposed license conditions, assumptions which form the basis for the safety analysis, and the actual conditions expected to exist in a Uranium Recovery Process (UWFPC).

Material in Process

The H/U of material in process will vary over a wide range; it is therefore analyzed as having the H/U resulting in the minimum allowed surface density. The proposed license conditions restrict batch sizes to 200 grams U-235. In addition, the process vessels are limited to 4 liters each. As discussed earlier, a batch may be distributed in one or more process vessels which are arranged in a linear array. Two batches or 400 grams U-235 maximum may be in process in a UWPRC at one time.

Double batching is a common concern in handling fissile material. The potential for double batching and other operational errors is minimized by the following features:

- The facility and staff has extensive experience with processes of a similar complexity performed in hot cells.

- Through test runs using unirradiated natural uranium, laboratory technicians are familiar with the process.

- Step-by-step procedures control the process. The uniformity resulting from repetitive, identical processes will provide results consistent with those demonstrated by experiments, reduce the probability of operator error, and give prompt identification of any deviation from normal conditions.

- Batching is done by transferring the contents of 200 ml borosilicate bottles into a 4 liter flask. This step is similar to waste handling currently done routinely and safely. A normal batch will occupy about 2.8 liters, leaving inadequate room in the 4 liter container for a complete double batch. Once combined, the process solution remains in an enclosed system. Transfer from one vessel to another is done by vacuum pumping.

Despite the unlikelihood of exceeding the 200 gram per batch limit and the impossibility of achieving a double batch, that condition is considered.

The entire amount of material in process is first considered as a single unit. If one of the two batches were somehow doubled, the total material in process would only be 600 grams U-235, well below the 760 gram single parameter limit (Reference 2, Section 2.2). The material in process will therefore be subcritical when considered as a single unit.

The analysis of the array of material in process is done next. The physical arrangement of containers is restricted to a linear array with a limit of 400 grams divided among 2 or more of the containers. For the purpose of analysis, the entire mass is considered a single unit. Intuitively, there should be a greater side separation (between the unit cell boundary and the linear array of process containers) than end separation (between the unit cell boundary and the end of the linear array). The approach used is to first determine the minimum end separation using a set of conservative assumptions. Then an additional conservative assumption (double batching) is added, giving the design bases for the minimum side separation.

The design bases for minimum end separation are:

- The entire licensed limit of 400 grams U-235 from two adjacent process vessels is represented as a bare sphere centered between those two vessels. (Data for a sphere confined by 1/16" stainless steel is used.)

- The H/U is at the concentration resulting in the most restrictive allowable surface density.

- The unit cell is reflected on all sides by thick concrete, reducing the allowable surface density to 60% of that calculated in Table C-2.

The results presented in Table C-2 show a unit cell requirement of 28" x 28" or a half-cell spacing of 14" which becomes the license limit for the separation between the center of mass of two end process vessels and the boundary of the array.

It is conservative to treat the two vessels as a single mass because, as can be seen from the data in Table C-2, when a uniform slab is segmented into a planar array of spherical units the allowable surface density decreases.

The side separation design bases are the same as listed above except that one batch is assumed doubled, making the spherical mass 600 grams. The unit cell requirement is 40" x 40", giving the license limit of 20" between the boundary of the array and the centerline of the linear array of process vessels.

Uranium Storage (3") Cylinders

Analysis of uranium storage cylinders is based on the fact that the $H/U \leq 20$. Measures are taken, as discussed above, to assure that processing is done properly and, therefore, that the H/U is within the license conditions. In addition, process monitoring instrumentation verifies the H/U in the following manner.

In processing a batch of uranium solution, a 20% excess of barium acetate is added for the reaction with the known amount of uranium. The filtrate consists of uranyl acetate and barium acetate in solution. The bulk content temperature and the wall temperature are continuously recorded as the cylinder is heated. Following distillation of the liquid and drying of the liquid a wet powder remains. As the powder reaches dryness the bulk temperature approaches the wall temperature, indicating that all moisture has been driven off leaving the uranyl acetate and barium acetate which has H/U = 11.6 as shown:

$$\frac{\text{H in } (UO_2(C_2H_3O_2)_2 \cdot 2H_2O) + 20\% \text{ of H in } (Ba(C_2H_3O_2)_2 \cdot H_2O)}{\text{U in } UO_2(C_2H_3O_2)_2 \cdot 2H_2O} = 11.6$$

After the moisture is driven off, the material continues to be heated for 5 hours at temperatures up to 320°C. With heating, the acetates convert to water vapor and other volatile gases while the uranium is oxidized, further reducing the H/U. Analyses of processed material show that $H/U < 2$ using the processing technique described.

The proposed license conditions allow for two 3" cylinders to be stored one on top of the other. In addition, when transferring cylinders a third cylinder may pass over two in storage, making a column of 3 cylinders having 200 grams U-235 each or a total of 600 grams. For conservatism, a total mass of 800 grams was assumed in every planar unit cell, despite the fact that only one can be transferred at a time. The metal container wall is conservatively represented in the analysis as a 25 mm water reflector confining the 800 gram U-235 mass. This is much less than the critical mass of a single unit either bare (8500 grams U) or reflected (5100 grams U) based on Figure 2.1 of Reference 2.

The planar array of unit cells was analyzed using the surface density analog of Reference 2. The license conditions allow two, 200 gram containers of H/U ≤ 20 centered in a unit cell of 12" x 12" minimum. The licensing bases conservatively assume 800 gram spheres of H/U = 20 in an array tightly fitted with concrete. The minimum unit cell spacing is then calculated to verify that 12" cells are adequate. Using Reference 2 and the nomenclature defined in Table C-2, the following values result:

$C = 1.22$ g U/cm^3 (for H/U = 20)

$T = 3.7$ cm (Figure 2.4 of Reference 2)

$\sigma_o = 4.51$ g U/cm^2

$M = 8500$ g U (Figure 2.1 of Reference 2)

$m = 800$ g U

$f = \frac{m}{M} = .094$

$\sigma = 0.54\,\sigma_o\,(1 - 1.37\,f) = 2.12$ g U/cm^2

$d = \sqrt{\frac{m}{0.60\sigma}} = 25$ cm $= 9.9$"

Since the minimum allowed unit cell dimension (9.9") is less than the license limit (12") the licensed array is conservative.

Flooding of UWPRC

The potential for flooding has been evaluated and determined not to be a credible event; moderation and reflection by water, therefore, need not be considered. The hot laboratory floor elevation is about four feet above grade level of the plateau on which the hot laboratory is located. This plateau is 30 to 40 feet higher than grade level of the remainder of the site. The site is well drained, leaving no possibility of external flooding. The proposed license conditions prohibits pressurized liquid inside the cell, eliminating tne possibility of flooding from within.

Despite the fact that flooding of fissile material storage arrays is not considered credible, storage racks are designed to assure drainage should moisture accumulate. Each hot cell door has approximately a 20 square inch drainage pathway at floor level that likewise guarantees no liquid accumulation in the cell.

Vault Pairs

Individual hot cells are separated by four feet of high density (magnitite) concrete, providing effective isolation between cells. Interaction between vault pairs, therefore, need not be considered.

Conclusion

The individual units and the arrays of both material in storage and material in process have been analyzed and determined to be criticality safe with wide safety margins under the proposed license conditions. Errors and malfunctions have been allowed for in the license conditions; the analysis shows that criticality safety is assured in the proposed license amendment.

TABLE C-2

DETERMINATION OF MINIMUM ALLOWABLE

SURFACE DENSITY AND UNIT CELL DIMENSION

				m = 400 g		m = 600 g	
C	T slab	σ_0	M	f	σ	f	σ
(g U/cm^3)	(cm)	(g/cm^2)	(kg)	—	(g/cm^2)	—	(g/cm^2)
.013	127	1.65	150	.003	.888	.004	.886
.02	20.3	.406	2.8	.143	.176	.214	.155
.03	12.7	.381	1.7	.235	.139	.353	.106
.04	10.2	.408	1.5	.267	.140	.400	.100
.05	8.4	.420	1.5	.267	.144	.400	.103
.06	7.6	.457	1.6	.250	.162	.375	.120
.1	6.1	.610	1.9	.211	.234	.316	.187
.2	5.0	1.0	3.0	.133	.441	.200	.392
.5	4.6	2.3	7.2	.056	1.147	.083	1.100
1.0	4.6	4.6	16.0	.025	2.399	.038	2.356

$d = \sqrt{\frac{m}{0.6\,\sigma_{min}}}$ = 28" 40"

d/2 = 14" 20"

C = U-235 concentration

T = minimum water reflected slab thickness, Figure 11 of Reference 3

σ_0 = Surface density for water-reflected infinite slab, C x T

M = Critical mass of an unreflected sphere, Figure 8 of Reference 3

m = mass of individual units of fissile material being analyzed

f = ratio of the mass of a unit in the array to the critical mass of an unreflected sphere, m/M

σ = allowed surface density

APPENDIX D

SAFETY ANALYSIS OF PROPOSED URANIUM WASTE FORM PROCESS

Introduction

The purpose of this safety analysis is to demonstrate that routine operation of the proposed uranium waste form process will not introduce additional significant radiation hazards to the public or hot laboratory operations personnel, and to describe and analyze the design basic accident.

Routine Operation

When considering the safeguard aspects of this process, it is convenient to divide the process into two parts termed "low temperature solution processing" and "high temperature solids processing". Each part has been examined to identify and evaluate potential hazards involving mechanical safety and release of radioactivity during routine operation.

Low Temperature Solution Processing

This part encompasses the sequential steps of precipitation, filtration, and distillation, which are arbitrarily considered together because they all take place at a temperature below 100°C.

Since the entire process is accomplished in a hot cell designed for processes of this type, there will no additional mechanical safety hazard associated with this part of the process.

The possibility exists that all of the solution in any of the glass containers could be spilled if the flask is broken or overturned. To prevent this from happening, the flasks are all fixed firmly to the main equipment rack. In the event that a flask breaks, all the solution will be maintained by a catch basin placed below each flask. If the basin were to fail to contain the solution, it would be contained in the large lipped pan that is under all the equipment.

It should be noted that prior to the waste form process most of the volatile fission products (Xe, Kr, I), which pose the greatest potential for radioactive effluents, have been removed. The present process for solidifying raw waste solutions involves openly transferring from individual bottles to an aluminum waste container; experience shows no significant release of gaseous activity from this source.

The chemistry of ruthenium during these low temperature steps has been examined. The valence state is no higher than +4 since it exists in a reducing environment throughout; ruthenium may become volatile at the +8 valance state, but is a particulate and therefore more controllable at valance states less than +8. Although full scale sulfate precipitation experiments, using actual raw fission waste at 90°C, have not released any ruthenium, a process step is included for evaluation in the startup program to precipitate the ruthenium out of solution in small batches (not to exceed 100 gm U 235 each) at the front end of the process.

In summary, no additional off site release is expected due to the routine operation of low temperature solution processing.

High Temperature Solids Processing

This part encompasses the sequential steps of drying and calcining in the aluminum shipping container. After all the solution has been transferred to the container and distilled to a cake of wet solids, the temperature is slowly raised to approximately 320°C until the uranium has been thoroughly dried and converted from the acetate to the oxide form. The drying and calcining process requires approximately 10 hours for completion.

The dry powders within the can in the presence of air begin decomposing in the latter part of the drying step. The major components to be decomposed are uranyl acetate, barium acetate, and small amounts of uranyl sulfate and nitrates. By carefully controlling the rate of temperature rise during this step, the uranyl acetate decomposition rate is controlled, avoiding excessive pressure buildup within the can. As discussed in the next section, even the design basis accident of an over-pressurized and ruptured container will not result in unacceptable releases.

The noble gas and iodine are removed prior to entering the heat treating steps. If ruthenium is present, it is expected to be only in the +3 and +4 valance states because of the reducing environment, resulting in a particulate rather than a volatile form.

In summary, no additional off site release is expected, due to the routine operation of high temperature solids processing.

Design Basis Accident

This accident is defined as the release of the entire contents of an aluminum waste container into the cell as a cloud of dust during the heat treatment step. The accident would be the result of a thermal excursion, due to improper temperature control. A rapid temperature rise to a temperature in excess of 500°C could possibly create pressure inside the vented can in excess of the can burst pressure, since the decomposing uranyl acetate would generate gases faster than the venting rate, releasing some of the uranium and fission product particulates into the cell.

The following conservative assumptions were assumed for the design basis accident:

- Maximum allowed batch size of 200 grams U 235
- Minimum practical decay time of 10 days
- Maximum irradiation time of 320 hours (average time is approximately 240 hours)
- Average neutron flux seen by targets (since multiple targets are required for a batch, the average flux is representative)

The results of these conditions is a theoretical batch inventory of 11,000 curies. In reality, the inventory based on the above conservative assumptions is approximately 3,000 curies because the following materials are removed prior to the calcining step:

- The Mo 99 which is separated for radiopharmaceutical purposes
- The iodine and noble gases which are trapped during the Mo 99 separation process, many of these byproducts likewise having radiopharmaceutical applications
- Approximately 60% of the remaining mixed fission product activity which precipitates or co-precipitates with the addition of barium acetate prior to the drying and calcining step

The main concern with the design basis accident is the potential offsite particulate release. Noble gases and iodines are removed earlier. The potential for each of other remaining fission products to volatilize has been evaluated and determined not to be consequential, a conclusion which can be readily verified during the startup program. In the event that particulate material were dispersed in the hot cell and entered the ventilation system (see description in Appendix E), it would be filtered by two HEPA filters in series, each having a rated efficiency of 99.97% for particles larger than 0.3 microns, which is a much smaller particle size than expected for a dust dispersal. Assuming no plating out of activity in the system and the theoretical batch inventory of activity, the maximum release would be:

$$11{,}000 \text{ curies} \times (3 \times 10^{-4})^2 = .001 \text{ curie} = 1{,}000 \text{ uci}$$

Reference 4 presents MPC values for mixed fission products as a function of decay time. For 10 day old mixed fission products in air, the environmental MPC is 1.33×10^{-9} uci/cc. Assuming no atmospheric dispersion, dilution by the normal 50,000 cfm exhaust ventilation flow from the facility results in a potential release of 0.4 MPC-day for the design basis accident.

Discussion

While the potential release of the postulated design basis accident is very small, numerous conservativisms should be pointed out which, in a more realistic assessment, would make the potential consequences even smaller:

1. The calculation is based on a release of 11,000 curies; in reality only 3,000 curies would be expected in the most restrictive conditions.

2. The dust must first pass through roughing filters in the hot cell into a plenum below the table. Then it must pass through approximately 10 feet of 10 inch diameter duct and 50 feet of three foot diameter duct to reach the first HEPA filter. It must then pass through three charcoal beds containing a total of 4 3/4" of activated charcoal and additional ducting before reaching the final HEPA filter. Some fraction of the dust would settle or plate out before the final HEPA filters, as well as in the 400 foot long, four foot diameter duct which goes up the hill from the hot laboratory to the exhaust stack.

3. No credit is taken for atmospheric dispersion due to the elevated release of the fine particles escaping the HEPA filters; the 0.4 MPC-day result is based on the concentration in the stack itself. The average measured dilution factor for the nearest residential area is 101,000.

4. In the model for the design basis accident, it is postulated that the temperature rises to 500°C instantly so that all the uranyl acetate would decompose at this temperature, leading to a high pressure and can rupture. If, in fact, some of the uranyl acetate had already decomposed prior to the temperature rise, the pressure developed at 500°C might be low enough so that the can could vent instead of rupturing and much smaller amounts of powder would be dispersed. In addition, it has not actually been proven under actual operating conditions that the container can be ruptured at 500°C.

5. The assumption of instant temperature rise is an essentially impossible condition for two reasons. First, the thermal inertia of the furnace/can system is such that it would take approximately 45 minutes to reach 500°C from the normal operating temperature of 300°C at full power. Second, there would have to be multiple simultaneous failures in redundant electrical and mechanical safety devices associated with the temperature control circuitry for the temperature excursion to occur.

APPENDIX E

ENVIRONMENTAL CONSIDERATIONS

Effluent Treatment

The uranium waste form process will be done in one of five hot cells which make up the main hot cell bank. The hot cell bank is maintained at a negative pressure at all times by the gaseous effluent treatment system. Exhaust fans draw suction from the cells through a common duct to filters; the discharge flow from the exhaust fans is released via an elevated stack. Instrumentation and controls measure radiation in the effluents, monitor cell pressure, alarm abnormal conditions, and automatically start backup equipment on loss of negative pressure. No liquid or solid radioactive effluents enter the environment surrounding the facility; liquids are solidified in the hot cells and shipped offsite for burial along with other solid wastes.

The gaseous hot cell effluents pass through two filter systems. They first pass through the hot cell filter system which consists of a roughing filter and HEPA filter followed by two banks of activated charcoal filters in series, each bank consisting of 2" charcoal beds of sufficient area, to assure low velocity - long resident time for maximum absorption. The HEPA plus two charcoal system is in parallel with an identical system to provide for maintenance and redundancy in emergency situations. Sample ports allow samples to be drawn at the various stages of filtration. Monthly test results have consistently shown charcoal filter efficiencies well in excess of 99.5%, which is expected since water vapor, organic fumes, and other contaminant substances are not present in significant quantities in the cells. The filter banks have an automatically actuated CO_2 fire extinguishing system. After leaving the hot cell filter system, the effluents pass through the main laboratory exhaust filter system which consists of a 3/4" activated charcoal bed, followed by a HEPA filter, the overall charcoal efficiency of this system being well in excess of 95%. The HEPA filter removal efficiency in each system is rated at 99.97% for particles 0.3 micron and larger.

The main exhaust fan operates continuously. In the event of loss of offsite power, it draws power from the emergency electrical generator which serves vital loads in the reactor and hot laboratory. In the event of high hot cell pressure (reduction in negative cell pressure), an auxillary exhaust fan will start automatically and maintain negative cell pressure. The auxillary fan is also fed by the emergency bus.

The negative pressure in each hot cell is indicated on manometers in the operating area. In addition, electrical devices sense pressure for the purpose of (1) initiating the above automatic action, and (2) alarming in the reactor control room.

A side-stream sample is drawn from the facility effluent stream before entering the vent line leading to the exhaust stack. This continuous sample is analyzed for radiation as particulates, noble gases or iodines. Any of the three will initiate a high alarm in the reactor control room. The alarm setpoint for the stack gas monitor is set below a value that would result in an exposure greater than 2 mrem hour assuming a dilution factor of 2000 and the isotope mixture determined by the most recent analysis. The alarm setpoint for the stack I-131 and stack particulate monitors is set below a value corresponding to that listed in Appendix B, Table II, Column I of 10 CFR Part 20 assuming a dilution factor of 2000 and averaging over one week.

Should these setpoints be reached, corrective action is initiated immediately. In addition to the continuous stack monitor, grab samples are regularly collected and analyzed to verify that routine emissions do not approach the instantaneous release setpoints.

The only effluents from the facility are those trace gaseous quantities remaining after extensive filtering. A high degree of reliability is designed into the gaseous effluent treatment system. Continuous monitors sense and alarm abnormal performance of the system. The salient result is a highly effective and reliable system for the treatment of gaseous effluents.

Airborne Effluent Releases

Many years of environmental monitoring by Union Carbide and a variety of regulatory agencies has resulted in the identification of the major exposure pathways associated with effluent releases. The nuclides identified as causing this exposure are Iodine 131, Iodine 125, and noble gases. These are all released from a single exhaust stack which is equipped with continuous monitors and recorders. The environmental monitoring program continuously samples air from five different locations. Two of these are within the 100 acre site and three are at strategic locations offsite. One of these offsite stations is located in the nearest residential area. Measured iodine concentrations at this station are used to calculate infant thyroid dose to this population. These measured concentrations, along with measured iodine concentrations in the exhaust stack, are also used to determine the gaseous dispersion factor. The noble gas concentration measured in the exhaust stack and the gaseous dispersion factor are then used to calculate the population exposure from noble gas. This method of analysis is necessary because the concentrations of noble gas at the monitoring station is too low to measure. These calculations are performed routinely in accordance with the requirements contained in the reactor Technical Specifications.

Using this technique and the dose factors contained in Regulatory Guide 1.109, Revision 1, October 1977, the present mode of operation is determined to result in the following population exposures at the nearest residential area:

Iodine 131	=	0.32 mrem per year (infant thyroid)
Iodine 125	=	0.48 mrem per year (infant thyroid)
Noble Gas	=	0.22 mrem per year (total body)

Operation of the proposed uranium waste form process should not have any effect on these offsite exposures. Iodine 125 is not involved in the process at all and Iodine 131 and Noble Gas are removed from the process prior to the waste form process step. A detailed analysis of effluent releases will be made during startup testing to confirm the prediction that the waste form process will not result in any measureable increase.

Occupational Radiation Exposure

The four foot thick high density concrete shielding of the hot cells minimizes occupational radiation exposure during in-cell uranium waste form processing. The major component of exposure from in-cell processing is caused by waste disposal procedures after completion of the processing. At the current production rate, approximately 50 waste shipments are made per year. The total annual employee exposure resulting from these shipments is 1.5 man-rem with a maximum individual exposure of 400 mrem.

With the addition of the proposed uranium waste form process, approximately 50% of the fission product inventory will remain in the cell for a considerably longer period of time; 120 days vs. 30 days. This in-cell decay will result in lower exposure dose rates on routine waste shipments. Since there is a slight waste volume increase as a result of the uranium waste form process, the projected number of shipments will not decrease, but the exposure resulting from these shipments will be less. The net reduction in occupational exposure is estimated to be as much as 10 to 20%.

Startup Plan

Startup and evaluation of the uranium waste form process will be done by R&D personnel with Health Physics supervising the effluent sampling and analysis. Production technicians will be present as part of an ongoing operator training program. Initial runs will be 1/4 scale, followed by 1/2 scale runs, and ultimately full scale runs. During small scale tests the need for the ruthenium precipitation step will be evaluated. If no volatile ruthenium is found in the absence of the precipitation step, the step will be eliminated.

During initial operation of the process, the continuous effluent radiation monitoring system will be checked carefully to detect any changes resulting from the procedure. In addition, evacuated containers will be used to obtain gas samples during the calcining step. The radioactivity content of these samples will be evaluated by the Health Physics group, using a calibrated multichannel Pulse-Height Analyzer. If a significant increase in activity is detected over present release rates, additional trapping devices will be investigated; development and prototype work done to date indicates that such action will not be necessary.

Conclusions

All potential effluents from the uranium waste form process will be subject to the existing effluent treatment system which has been demonstrated to be a highly effective and reliable system. No change is expected in offsite exposure as a result of the process. Because of the more effective waste packaging, a reduction of 10-20% is expected in the occupational exposure due to waste handling on site. A startup program is planned to verify the conclusion that there will be no significant environmental impact from the process. Not included in this determination is the global benefit derived in sending the uranium to a reprocessor, rather than a waste burial site, thereby reducing land use and the environmental impact and occupational exposures associated with waste burial, mining, milling, and enriching the equivalent amount of material.