

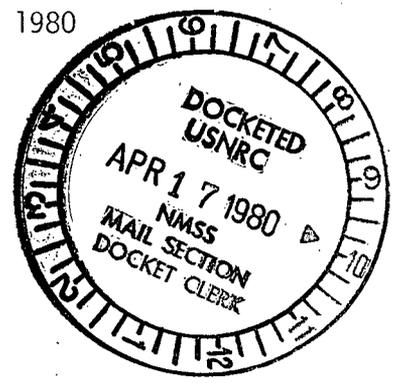


UNION CARBIDE CORPORATION
MEDICAL PRODUCTS DIVISION
P.O. BOX 324, TUXEDO, NEW YORK 10982
TELEPHONE: 914-851-1203

REGULATORY OPERATIONS
FILE COPY

RECEIVED
U.S. NUCLEAR REG.
COMMISSION
MAIL SECTION

April 2, 1980



W.T. Crow, Section Leader
Uranium Fuel Licensing Branch
Office of Nuclear Material Safety & Safeguards
United States Nuclear Regulatory Commission
Washington, D. C. 20555.

License Amendment Request to
Permit Uranium Recovery from Waste
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.

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 certain facets of the application, it is hereby being resubmitted, incorporating agreed-upon changes, superseding Reference 1 in its entirety.

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 recovery step into the existing waste handling process. The uranium recovery 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 process is retained.

FEE EXEMPT
add'l info

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 April 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 Recovery Cell (URC). Raw fission waste solution will be processed in 200 gram U-235 batches and stored in the URC. The end result of the process will be oxidized uranium powder sealed in a 3" diameter cylinder. The liquid remaining from the process with approximately half of the fission products and trace quantities of uranium will be solidified as is presently done, either in the URC or another cell. The 3" diameter cylinders will be stored in an approved array in the URC awaiting shipment to a reprocessing facility.

The uranium recovery process is described in Appendix A. The typical layout of a URC, including the process equipment and the storage configuration, are discussed in Appendix B. The safety analysis of the composite URC, which supports the specific license conditions, is included as Appendix C.

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 Recovery Cell (URC) shall contain no more than 7600 grams of U-235, ≤ 7200 grams being allowed in oxide storage cylinders and ≤ 400 grams in process.
2. Supporting and storage fixtures in a URC 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 minimum 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 URC 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 process shall be restricted to the recovery of uranium from waste solution and shall not include the 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.

Page Four
April 2, 1980

Conclusion

The requested license amendment allows for the conversion of uranium wastes to a usable form. 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 recovery 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: J. Delany, U.S.N.R.C.
W.H. Briner, Society of Nuclear Medicine

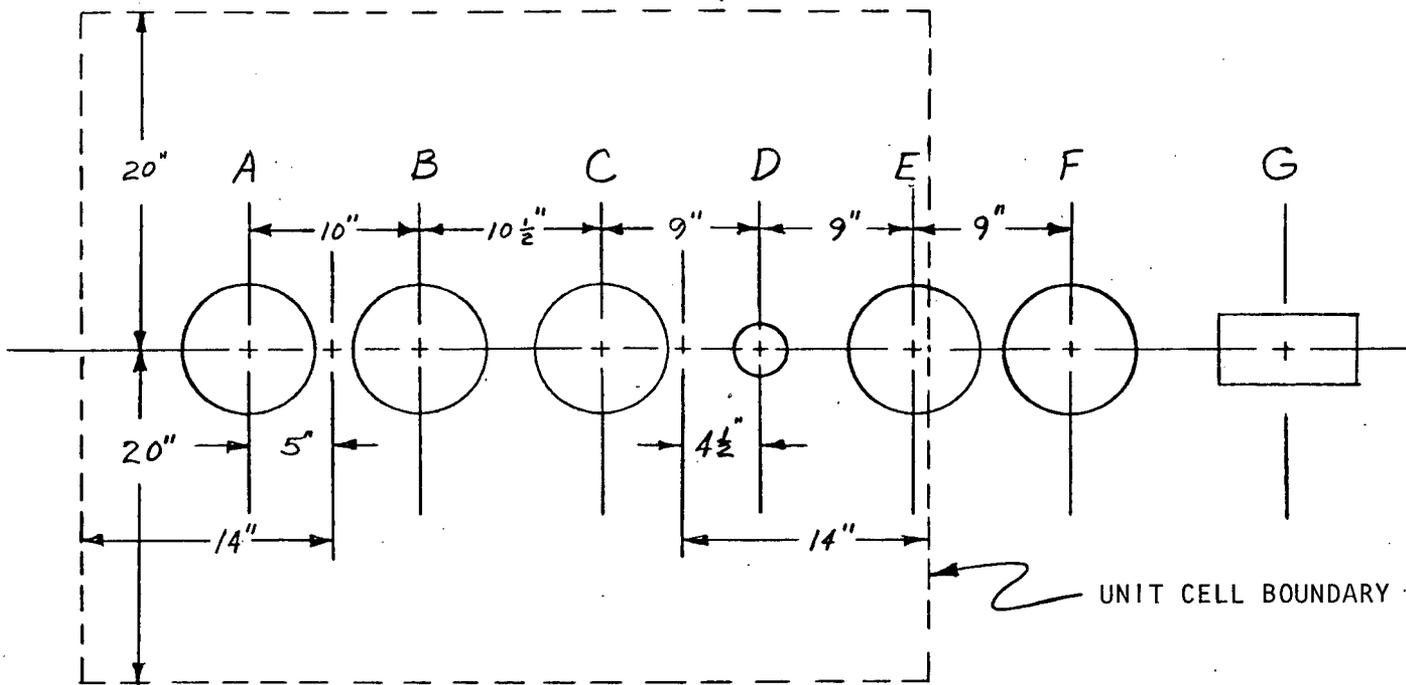
APPENDIX A

DESCRIPTION OF URANIUM RECOVERY PROCESS

Fission product Mo-99 is produced by irradiating targets containing from 10 to 20 grams of uranium enriched to 93% in the U-235 isotope. Raw fission waste solutions, generated by this procedure, consist of uranium and mixed fission products dissolved in a dilute (~ 2 N) sulfuric acid "cocktail" containing $\sim 2\%$ nitric acid. Each process creates approximately 150 ml of such waste, which is then stored in a borosilicate glass bottle, labeled with the appropriate run number, for further disposition. Since only a small portion ($\sim 1\%$) 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. The containers are identified in Figure A-1, which also shows a typical layout along with the minimum unit cell boundary defined by the proposed license conditions. Process steps include:

1. Combine the contents of borosilicate waste bottles containing up to 200 g U-235, (Containers A or B).
2. Precipitate the sulfates from the raw fission waste solution by the addition of barium in the form of a barium acetate solution (Containers A and B), adding a volume of approximately 1300 ml to the 1500 ml of waste solution.
3. Decant and filter the solution to remove the BaSO₄ precipitate (Container A to C or B to C).
4. Measure the filtrate volume and take a sample for assay (Container C).
5. Transfer the solution to an aluminum container while heating to dryness (Container C to D).
6. Continue heating to calcine the uranium (Container D, distillate passing to E and F).
7. Weigh the aluminum container to determine the net weight of uranium content (Container D).
8. Seal the aluminum container and store for subsequent shipment to the reprocessing facility (Container D).
9. Dispose of the precipitate sludge (Container A or B) and the distillate (Container F).

A typical arrangement and container dimensions are shown in Figures A-1 through A-3; however, the license conditions are governing regarding these matters.



- 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

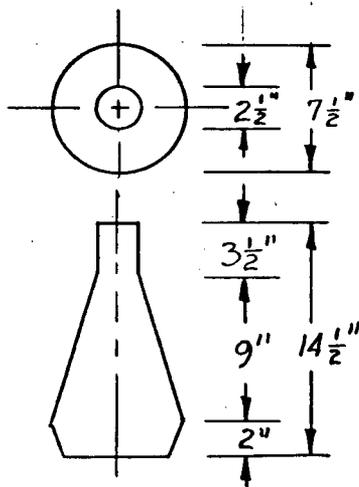


FIGURE A-2

FOUR LITER FLASK

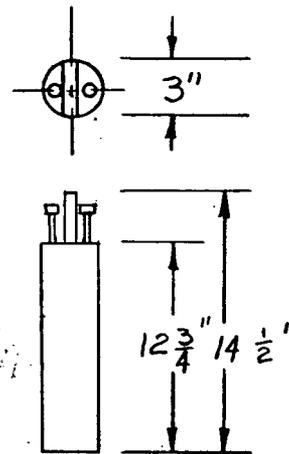


FIGURE A-3

URANIUM CONTAINER

APPENDIX B

TYPICAL URC LAYOUT

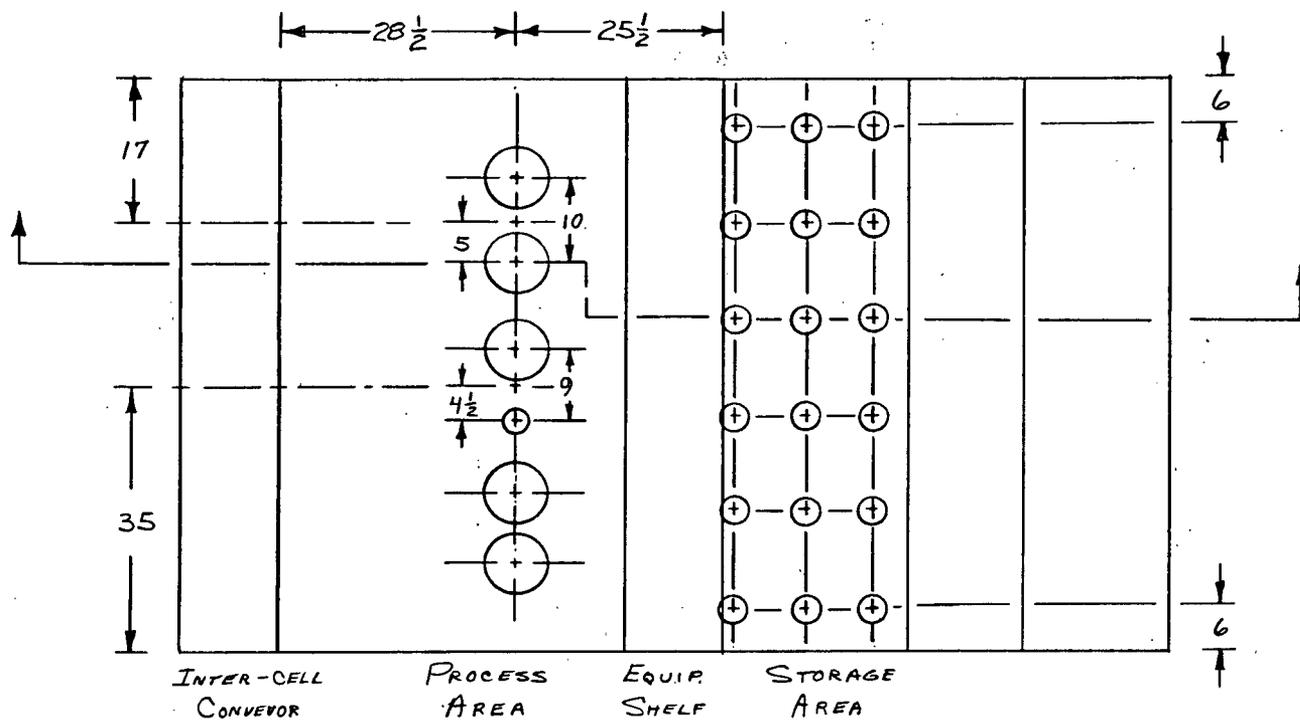
To better visualize the general license conditions stated in the foregoing letter and the supporting safety analysis presented in Appendix C, a typical Uranium Recovery Cell (URC) is shown in Figure B-1 and discussed in this section. Figure A-1 shows the minimum proposed license dimensions of a typical 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 URC as a planar array, representing individual mass units as spheres, the minimum separations are reflected onto an imaginary plane which is parallel to the storage rack surface. The following comparison of minimum allowable to actual separation shows that in all cases the minimum criteria are satisfied:

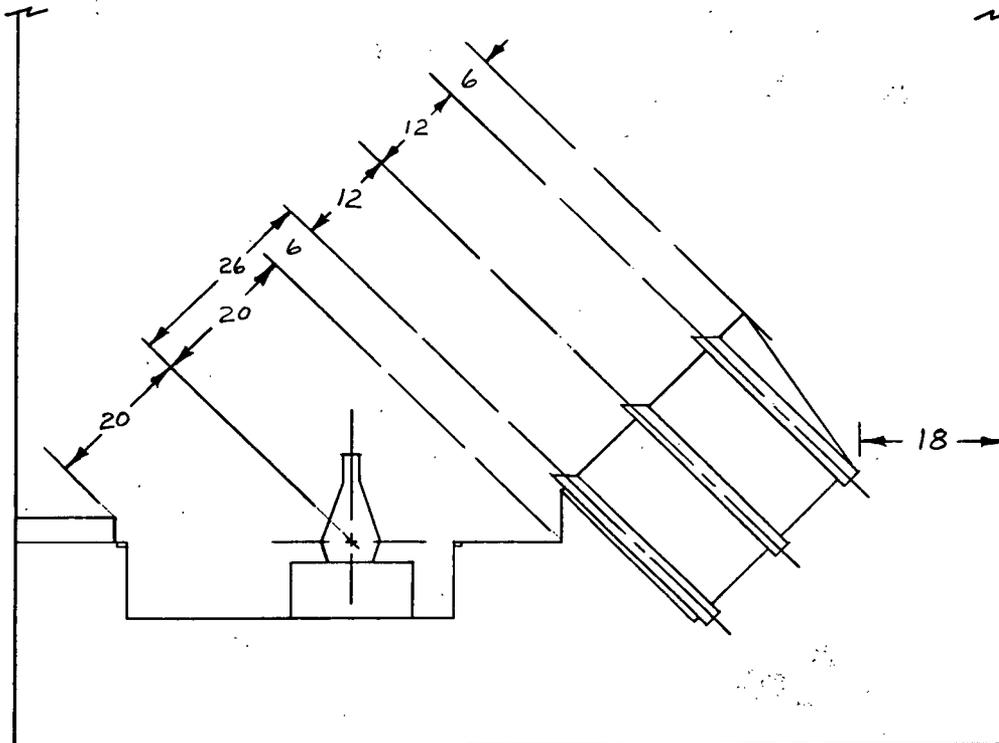
<u>Parameter</u>	<u>Minimum Allowable Separation</u>	<u>Actual Separation</u>
3" uranium cylinders in storage (centers)	12"	12"
3" cylinder (center) to cell walls	6"	6" sides 18" back
3" cylinders to process containers (centers)	26"	26"
process containers (center) to edge of URC (intercell conveyor)	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 RECOVERY 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 dimension 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 Cell (URC).

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 URC 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:

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

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 $H/U \leq 20$ 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:

$$\begin{aligned}C &= 1.22 \text{ g U/cm}^3 \text{ (for } H/U = 20) \\T &= 3.7 \text{ cm (Figure 2.4 of Reference 2)} \\ \sigma_0 &= 4.51 \text{ g U/cm}^2 \\M &= 8500 \text{ g U (Figure 2.1 of Reference 2)} \\m &= 800 \text{ g U} \\f &= \frac{m}{M} = .094 \\ \sigma &= 0.54 \sigma_0 (1 - 1.37 f) = 2.12 \text{ g U/cm}^2 \\d &= \sqrt{\frac{m}{0.60\sigma}} = 25 \text{ cm} = 9.9''\end{aligned}$$

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

Flooding of URC

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 the 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 (magnite) 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-1: Comparison of License Conditions
Basis of Safety Analysis, and Actual Conditions

<u>License Condition</u>	<u>Basis of Safety Analysis</u>	<u>Actual Condition</u>
Material enclosed in concrete hot cell.	Tightly fitted concrete around all sides of the planar array.	Less than half of the perimeter has tightly fitted concrete.
Two batches, 200 g max. each, in process. Separation between center of combined end containers and cell boundary ≥ 14 ".	400 g sphere centered 14" from the cell boundary having H/U resulting in minimum critical mass.	Two batches, 200 g max. each in process. Separation of 14" when in position nearest to the boundary.
Two batches, 200 g max. each, in process. Separation from center of material in process to boundary of storage array ≥ 20 ".	600 g sphere centered 20" from the cell boundary having H/U resulting in minimum critical mass.	Containers restricted in size making complete double batching impossible. Batch vessels separated, making less reactive array than the assumed sphere. Separation from center of process vessels to boundary is 20".
3" max. ID cylinders containing ≤ 200 g each stored 2 high on vertical centerline of planar unit cell ≥ 12 " x 12".	800 g sphere centered in a 9.9" square unit cell.	Two 3" diameter cylinders containing ≤ 200 g each stored one on top of another with the possibility of a third cylinder in transit over the two, effective height of the 600 g being 48" in a 12" x 12" unit cell.
H/U ≤ 20 in storage cylinder.	H/U=20, container wall simulated by a 25 mm water reflector.	H/U < 2 in a metal container.

TABLE C-2

DETERMINATION OF MINIMUM ALLOWABLE
SURFACE DENSITY AND UNIT CELL DIMENSION

C (g U/cm ³)	T slab (cm)	σ_0 (g/cm ²)	M (kg)	m = 400 g		m = 600 g	
				f	σ (g/cm ²)	f	σ (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