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"LICENSE AMENDMENTS"

#15103

Docket No. 70-687

William O. Miller, License Fee Management Branch, ADM

MATERIALS LICENSE AMENDMENT CLASSIFICATION

Applicant: Union Carbide

License No: SNM-639 Fee Category: 1G

Application Dated: 12/28/79 Received: 1/8/80

Applicant's Classification: safety (amendment - minor)

The above application for amendment has been reviewed by NMSS in accordance with §170.31 of Part 170, and is classified as follows:

1. Safety and Environmental Amendments to Licenses in Fee Categories 1A through 1H, 2A, 2B, 2C, and 4A

 (a) ____ Major safety and environmental

 (b) ✓ Minor safety and environmental

 (c) ____ Safety and environmental (Categories 1D through 1G only)

 (d) ____ Administrative

2. Justification for reclassification: ____________________

3. The application was filed (a) ____ pursuant to written NRC request and the amendment is being issued for the convenience of the Commission, or (b) ____ Other (State reason): ____________________

Signature [signature]
Division of Fuel Cycle & Material Safety

Date 2/1/80

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December 28, 1979

W.T. Crow, Section Leader
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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 Recovery from Waste
License No. SNM-639 Docket No. 70-687

References:

1. American National Standard Guide for Nuclear Criticality Safety in the Storage of Fissile Materials, N16.5 - 1975; published by American Nuclear Society.

2. Nuclear Safety Guide TID-7016, Revision 2, edited by J. L. Thomas, June 1968.

3. Personal discussions with Mr. J. L. Thomas on December 11, 1979.

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Introduction

In our production and separation of medical radioisotopes, a substantial amount of unused fissile target material remains with the radioactive wastes. Our 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. We hereby request an amendment to our SNM-639 license which will allow us to implement a uranium recovery step into our existing waste handling process. The uranium recovery process is simply a conversion of the uranium and other fission products in our 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.

This amendment applies to the contents of waste processing and storage hot cells; fissile materials in forms other than those discussed herein will not be allowed in the work area of the waste processing and storage cell. (They can be transferred through on the inter-cell conveyor which is outside of the processing and storage areas.) The amendment also applies to storage in the transfer canal or reactor pool.

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 December 28, 1979."

Discussion of License Conditions

Processing of wastes will be done within as many of the existing constraints as possible; new steps are identified as described in Appendix A and evaluated for criticality safety in Appendix B. Presently licensed aspects of waste handling which are applicable include:

- An SNM limit of 2000 grams of fully enriched U-235 per waste storage hot cell

- A limit of 200 grams U-235 per aluminum waste container

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- A maximum waste container inside diameter of 5", no restriction on height, and a right cylindrical geometry.

- Storage of waste containers in a linear array, with a minimum center-to-center spacing of 6 1/4" and no restrictions on the total number of containers in the linear array (other than the mass limit for the cell).

- Storage in waste cells of 55 gallon 17H steel drums of radioactive wastes containing waste containers with a maximum of 350 grams of U-235 per drum.

Applicable aspects of the present license basis which are highlighted for clarity or first-time application include:

- More than one hot cell may be classified as a waste processing and storage cell.

- The relative location of 17H waste drums (containing up to 350 g U-235 each) in a waste storage cell is unrestricted; they remain criticality safe even in a tightly packed multi-layered array.

New aspects of waste processing and storage requested by this license amendment include:

- Fissile material will be processed in liquid form in a waste processing and storage cell.

- Processed uranium wastes in dry, powder form will be encapsulated in cylinders described below.

- Waste processing cells may contain SNM in any combination of four forms; liquids in process, reclaimed uranium cylinders, solidified waste solutions in 5" cylinders, and waste drums.

- Cylinders containing reclaimed uranium may be stored in defined arrays in the water-filled transfer canal or reactor pool.

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 typical operations 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. Up to 200 grams of U-235 in the form of reclaimed uranium having H/U $\leq$ 20 may be stored in cylinders of 5.0" maximum internal diameter with no limit on the height of the container. (Typically 3" OD by 14" high containers).

2. The reclaimed uranium containers may be stored in waste processing and storage hot cells, provided they are stored in linear arrays having a minimum center-to-center spacing of 6 1/4", subject to the additional restrictions in items 3 and 4 below.

3. The following quantities and forms of fissile material may be stored in a waste processing and storage hot cell, subject to the 2000 gram U-235 limit per cell and condition 4:

	Form	Maximum Quantity	Maximum Number of Containers, N	Unit Cell Dimension, d*
a.	Waste Drums	350 grams U-235 / drum	20	24"
b.	Waste Cylinders	200 grams U-235 / cylinder	20	24"
c.	Reclaimed Uranium Cylinders	200 grams U-235 / container	20	~~18"~~ 10"
d.	Material in Process	400 grams U-235	N.A.	20"

*The unit cell dimension, d, is a parameter used in determining storage area dimensions as described in Condition 4.

4. When materials of form c or d (above) are present in a waste processing and storage cell, a "storage area" must be defined for each form of fissile material. No two storage areas may overlap. No fissile material of one form may be taken into a storage area of another form; physical or procedural barriers shall effect this requirement. The storage areas shall be defined as follows:

Form	Storage Area
a. Waste Drums	The volume defined by the 55 gallon specification 17H drums when placed in any array.
b. Waste Cylinders and c. Reclaimed Uranium Cylinders	A regular parallelepiped whose volume equals Nd^3, the number of containers allowed in the storage area (N) times the unit cell volume (d^3). The dimensions shall be such that each surface of the storage area has the same minimum separation, s, from the fissile material containers; that is, if the linear array of containers is confined to a minimum volume of x y z, $(x+2s)(y+2s)(z+2s) = Nd^3$. Multiple storage areas are allowed in a hot cell. The storage areas may include concrete cell walls.
d. Material in Process	The storage area shall be the regular parallelepiped enclosed by six planes, each plane having a minimum separation of d/2 from the center of a fissile material batch container. Only one such storage area is allowed in a waste processing and storage hot cell. The storage area may not include concrete cell walls.

5. Material in process in each waste processing and storage cell 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. The process shall be restricted to the recovery of uranium from waste solution and shall not include the separation of selected fission products.

6. Reclaimed uranium cylinders may be placed in storage racks in the reactor pool or the transfer canal. Storage racks shall be designed such that each cylinder is centered in a unit cell no less than 9" x 9" x 14", and such that other fissile material is physically prohibited from being brought to within a 9" center-to-center spacing of a uranium cylinder in the storage array.

Conclusion

The requested license amendment allows for the conversion of uranium wastes to a usable form. While many features of the existing license remain applicable, new conditions have been required and are proposed. Gross conservatisms are included in the license conditions to provide flexibility, ease of analysis, and potential for increasing quantities at a later date without changing geometries.

Pursuant to 10 CFR Part 170.31.1.D, a check in the amount of $1,400 is attached for the Safety and Environmental review necessary for this amendment. Note that the only change which affects materials and plant protection is the storage of irradiated target material in the reactor pool and transfer canal, which has already been evaluated 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: Mr. J. Delany, U.S.N.R.C.
Captain 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 ($\sim$ 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 as follows (refer to Figure A-1):

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 $BaSO_4$ precipitate (Container A to C or B to C).

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

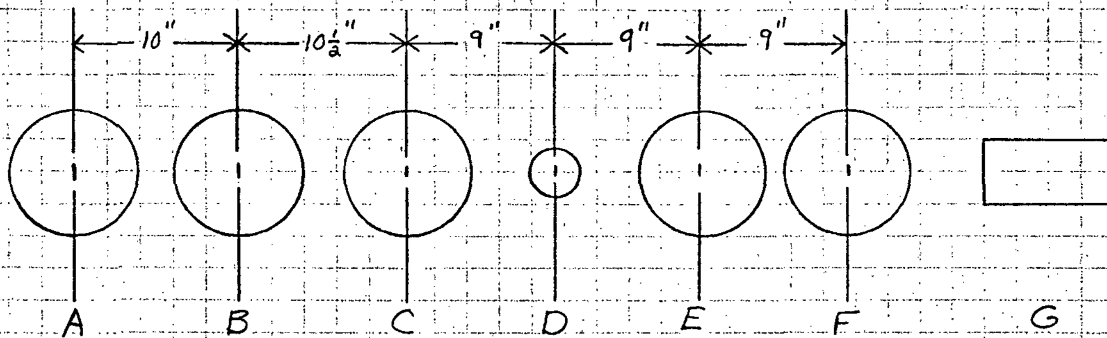
5. Transfer the solution to an aluminum container while heating to dryness (Container C to D).

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).

The typical arrangement, separation 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. Condensor
F. Distillate - 4 liter flask
G. Vacuum Pump

FIGURE A-1

ARRANGEMENT OF COMPONENTS IN HOT CELLS

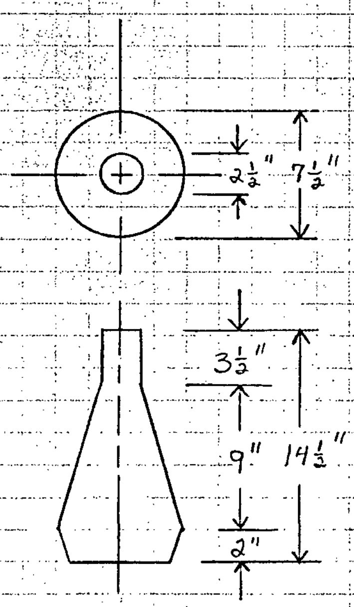


FIGURE A-2

FOUR LITER FLASK

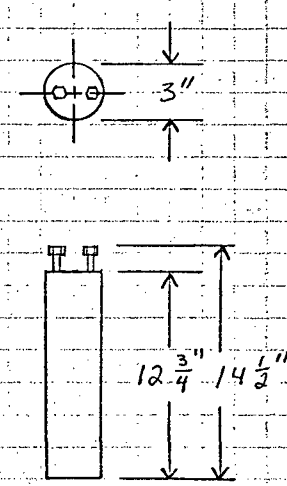


FIGURE A-3

URANIUM CONTAINER

APPENDIX B - CRITICALITY SAFETY EVALUATION

Criticality safety considerations of the proposed license changes are handled in a two-step process:

I. Describe each form of fissile material, its normal storage configuration and the most severe conditions assuming an operator error or equipment malfunction. Evaluate the criticality safety of each configuration.

II. Evaluate the interaction between the various adjacent configurations.

Part 1 - Evaluation of Arrays

Waste Drum Storage

The license already allows the storage of up to 350 grams of U-235 enriched to 93% in each 55 gallon specification 17H drum in a waste processing and storage cell. This form of storage remains unchanged. It should be noted that this is 1.32 grams per liter, less than 12% of the single parameter limit of 11.5 g U-235/liter (Reference 2). Since the fissile material is not distributed homogeneously throughout the drum and moderation is not necessarily optimum, even greater margins of safety exist. Due to the low concentration of fissile material, an infinite array of closely stacked drums remains subcritical.

Waste Cylinders

The present license allows storage of waste cylinders along with waste drums in a waste processing and storage cell, provided the cylinders are no greater than 5" inside diameter and are stored in a linear array having a minimum center-to-center separation of 6 1/4", and contain no more than 200 grams U-235 enriched to 93% per cylinder with no restriction on the H/U ratio (typical concentration is 50 grams U-235/liter or H/U = 500).

Reclaimed Uranium Cylinders

The reclaimed uranium cylinders fall within the envelope of analysis for the waste cylinders. The license conditions for container size, storage configuration, and quantity of fissile material are the same. In addition, a restriction of $H/U \leq 20$ is included to allow (in Part II) a smaller storage area of reclaimed uranium cylinders. Three abnormal conditions are evaluated:

1. Interchanging waste cylinders and reclaimed uranium cylinders in storage arrays.

2. Exceeding the maximum allowed H/U.

3. Double batching.

Interchanging a waste cylinder for a reclaimed uranium cylinder is not likely. The majority of the uranium from process wastes will be reclaimed in cylinders destined for reprocessing; a small amount remaining in the sludge will be solidified in waste cylinders. Because of the economic value of reclaimed uranium cylinders and the problems associated with sending a waste cylinder to reprocessing, special constraints are implemented to prevent an interchange. Waste cylinders are presently the full 5" diameter allowed by the license, while reclaimed uranium cylinders are only 3" diameter with receptacles in the storage racks sized accordingly. Even if the cylinders and racks were identical and cylinders were exchanged, the linear array would remain subcritical since it is not dependent on H/U.

Measures will be taken, as discussed below, to assure that processing is done properly and, therefore, that the H/U and the net uranium weight is within the license conditions. In addition, post-process weighing of each cylinder will verify the H/U in the following manner. In processing a batch of uranium solution, a 20% excess of barium acetate will be added for the reaction with the known amount of uranium. The filtrate will consist of uranyl acetate and barium acetate after moisture has been driven off. At this point in the process of a batch containing 200 gram U-235 enriched to 93%, there will be 392 grams of uranyl acetate and 61 grams of barium acetate, the total weight being 453 grams. The H/U at this point would be:

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

With heating, the acetates convert to water vapor and other volatile gases while the uranium is oxidized, further reducing the H/U and the total mass. Experiments show that for an initial 200 g of U-235 batch, the final net contents weigh approximately 350 g. An acceptance criteria for storage restricting the net weight to less than two times the starting batch weight of U-235 will verify that contents of reclaimed uranium cylinders have an acceptable H/U and are not double batched.

Material in Process

The processing of fissile waste material in a waste processing and storage hot cell is a new license condition; the safety evaluation of the process shows substantial margins of safety. While this specific process is new, the facility and its staff have years of experience with similar processes. Safety matters evaluated include:

1. Criticality (including oversized batches)
2. Operator errors
3. Process equipment malfunctions

The lines of defense against accidental criticality are threefold:

1. Batches are restricted to small sizes
2. Containers restrict the volumes of fissile material to small quantities
3. Procedures administratively control the process

The maximum batch size of 200 grams U-235 represents about one weeks through-put. To double batch would mean skipping a process one week, an event not likely to be ignored. Nevertheless, the limit for material in process is 400 grams which allows two batches in process, one assumed to be double-batched, making the total 600 grams, which is below the single parameter limit of 760 grams U-235.

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 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. The final vessel contains the distillate driven off the uranium solution as it is converted to an oxide form in the reclaimed uranium cylinder. Should any uranium pass through the system, it will be collected in this container. Should any container or connection break the maximum amount of fissile material that can escape is one batch of up to 200 grams U-235. Even though this is unlikely and unexpected, each glass container is placed in a metal catch basin, plus a pan is provided which holds the entire process. Any spill will be collected by the pan; in addition, the geometry of the pan is such that the material will be dispersed rather than concentrated, making the potential for neutron interaction insignificant.

The design of the process and the related equipment is such that credible operator errors and equipment malfunctions can be tolerated without a significant impact on criticality safety. Integrated with these features as a backup will be step by step procedures which administratively 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.

Pool Storage Racks

The pool storage racks for reclaimed uranium cylinders are designed based on the single parameter limit of 11.5 grams U-235 per liter. The cylinder, having a maximum of 200 grams U-235 with H/U $\leq$ 20, would normally be allowed to reside in much closer array than allowed by this limit. However, it is conservatively assumed that, despite the sealing and leak testing of the cylinders, they all fill with water, placing the uranium oxide in a more reactive homogeneous mixture. The unit cell of 9" x 9" x 14" represents an average concentration of 10.76 grams U-235 per liter. An infinite array will, therefore, remain subcritical in this abnormal condition since, with all cylinders at their maximum loading, the 11.5 gram/liter criteria is met. Individual cells are likewise criticality safe because the 200 grams U-235 per cell is well below the single parameter limit of 760 grams.

PART II - EVALUATION OF INTERACTION BETWEEN ARRAYS

The evaluation of interaction between arrays for criticality safety is the Criticality Indicator (CI) technique discussed in Reference 2, using data presented in Reference 1. An entire waste processing and storage hot cell will be considered a single large storage area subdivided into smaller storage areas, wherein fissile material of only one form may be taken. By the Criticality Indicator Technique, each of N fissile material units having a unit cell dimension d is assigned a CI, the total of which must not exceed 100. The analyzed configuration remains subcritical when either dry or flooded. When concrete is present in a storage area, the allowed mass is reduced to 60% to account for reflection by the concrete.

While the CI technique requires separation into defined unit cells, other analysis allows closer spacing of units in a storage area. Therefore, the CI method is used only to evaluate interaction. To make the CI method valid, a storage area is defined equivalent in volume to the aggregate volume of the unit cells. In this manner, there is a greater separation between each of the forms of fissile material. The geometry of storage areas is defined to provide the maximum separation.

Reference 1 states the limits for unit cells as a function of H/U. Experiments have shown (Reference 3) that if the volume of fissile material remains the same while H/U is increased (that is, replacing U with H atoms), the H/U $\leqslant$ 20 data is valid for any H/U. This factor is used below where H/U exceeds or could exceed 20 with the following results:

Form	Number of Units, N	Unit Cell, d	CI
Waste Drums	20	24"	2
Waste Cylinders	20	24"	31
Reclaimed Uranium Cylinders	20	~~18"~~ 10"	2
Material in Process	NA	20"	3
		Total CI =	38

The number of units selected is the maximum number of units of any form of material, filled to approximately half of the licensed limit for fissile material per unit, that it would take to fill the waste processing and storage cell to its 2000 gram U-235 limit. Because the total inventory of the cell must be less than 2000 grams, the analysis is very conservative. Since the aggregate CI is less than 100, the storage area is considered criticality safe.

Waste Drums

The fissile material per waste drum is restricted to 350 grams U-235, contained in waste cylinders. Since the concentration of fissile material is only a fraction of the single parameter limit, it can be stored in an infinte array and the CI would not apply. However, to evaluate potential interaction when an array of waste drums is stored in the vicinity of other fissile material containers of a defined CI, the following analogy is used to derive an appropriate CI. In the Density Analog, described in Reference 2, a unit cell for storage is determined by equation 4.8, which is a function of the allowed surface density in equation 4.5. The allowed surface density, as a function of H/U and uranium concentration in water, is plotted in Figure B-1. The fissile concentration in a tightly packed array of drums, with each drum having its limit of 350 grams U-235, is 0.00132 grams U per cubic centimeter. Since the allowed surface density for concentrations less than 0.00132 g U/cc is much greater than that for concentrations of 1.22 g U/cc (H/U = 20), the CI data of Reference 1 for H/U $\geq$ 20 will be used. From Table 5.2 up to 1000 units of 3.4 kg U each can be stored in 24" unit cells. Therefore, 20 waste drums would have a CI of:

$$CI = 20 \frac{(100)}{1000} = 2$$

The volume occupied by the drum is 7% greater than that of a 24" cubic unit cell. Therefore, no larger storage area need be defined beyond the confines of the drum. Concrete reflectors can also be ignored.

Waste Cylinders

Before being placed in waste drums, the waste cylinders may be stored in the specified storage areas. Each cylinder has up to 200 grams U-235 in an effective volume of 3 liters. If the 3 liter volume contained fissile material of H/U = 20 or 1.22 grams U per cubic centimeter, the container would have 3.66 kg U. However, concrete reflectors are assumed, making an equivalent fissile content of 6.1 kg. From Table 5.2 of Reference 1, up to 64 units of 6.5 kg U each can be stored in 24" unit cells, making the CI:

$$CI = 20 \frac{(100)}{64} = 31$$

Since this material is stored in a linear array, the storage area designated for the material is defined to give maximum separation from other fissile materials. Namely, the minimum separation, s, from the container to the boundary of a storage area shall be such that:

$$(x+2s)(y+2s)(z+2s) = Nd^3$$

For example, a linear array of 5 waste cylinders of 5" diameter x 10" high on 6 1/4" spacing would mean:

$$(5+2s)(30+2s)(10+2s) = 5 \times (24)^3$$

or s = 13.7" with a storage cell defined having dimensions of 32.4" x 57.4" x 37.4", centered about the linear array.

Reclaimed Uranium Cylinders

The content of a reclaimed uranium cylinder may be a maximum of 200 grams of U-235 of 93% enrichment and H/U less than or equal to 20. Since concrete may be present as a reflector, the effective mass is 0.36 kg U which, by Table 5.2 of Reference 1, can be stored in 1000 unit arrays of 10" cubic storage units. For 20 units, the CI is:

$$CI = 20\frac{(100)}{1000} = 2$$

A storage area of:

$$(x+2s)(y+2s)(z+2s) = Nd^3$$

will be defined for reclaimed uranium cylinder storage in the same manner as for waste cylinder storage.

Material in Process

The process vessels are restricted to less than or equal to 4 liter containers. Using the experimental data (Reference 3), and allowing for H/U = 20, the 4 liter containers could contain 4.9 kg U each. Since the storage area for material in process may not include concrete, there is no adjustment. According to Table 5.2 of Reference 1, up to 64 units of ≤ 5.2 kg U can be stored in 20" cubic storage units. Since only 2 batches can be in process concurrently,

$$CI = 2 \times \frac{(100)}{64} = 3$$

The definition of a storage area is actually larger than $2d^3$, since the material can move in the process. The storage area is conservatively defined as the area enclosing the material in process with a minimum distance of d/2 from the center of the process containers (items A, B, C, and D in Figure A-1) to the extremity of the storage area. The storage area is in practice approximately 20" x 20" x 50" in the layout shown or approximately $2.5d^3$.

Pool Storage Racks

The only forms of fissile material normally handled in the reactor pool and transfer canal will be the reclaimed uranium cylinders, fueled targets, and reactor fuel elements. Interaction between the storage array and other fissile materials has been evaluated. The proposed license conditions require a minimum 9" center-to-center physical separation between a reclaimed uranium cylinder stored in the array and other fissile materials. The uranium cylinder represents the greatest concentration of fissile material of any form handled in the pool or canal. Therefore, by maintaining the required spacing, any fissile material brought close to the storage will not interact with the array to a greater extent than if it were placed in the array which is shown to be criticality safe as an infinite array.

Conclusion

Each of the storage arrays and also the configuration of material in process has been analyzed and determined to be criticality safe. Interaction between arrays has been analyzed and shown 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.

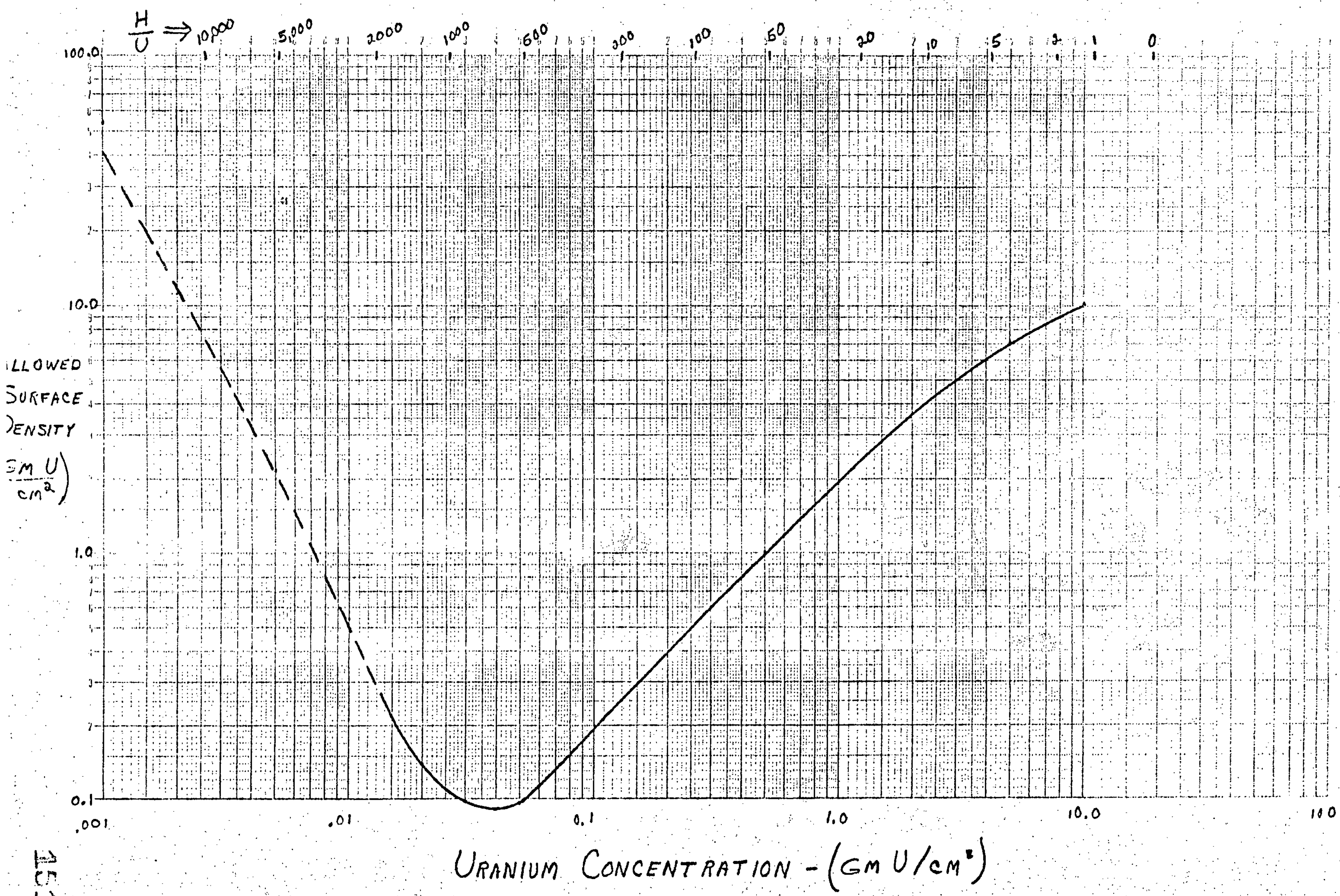


FIGURE B-1. ALLOWED URANIUM SURFACE DENSITY vs. CONCENTRATION FOR AQUEOUS SOLUTIONS