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REFLECTOR SAVINGS
FOR URANYL NITRATE OF LOW ENRICHMENT

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REFLECTOR SAVINGS FOR URANYL NITRATE OF LOW ENRICHMENT

by

C. A. Rogers, R. C. Lloyd, S. R. Bierman, and E. D. Clayton

Physics Research Department
Physics and Engineering Division

November 1969

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REFLECTOR SAVINGS FOR URANYL NITRATE OF LOW ENRICHMENT

ABSTRACT

Reflector savings for 2.14 and 3.04 wt% ^{235}U enriched uranyl nitrate reflecting a cuboidal core of PuO_2 -polystyrene were experimentally determined and compared with calculations using the HFN and DTF codes. HE^T-computed reflector savings curves for less than 5.0 wt% ^{235}U enriched uranyl nitrate reflecting a spherical, water-moderated, uranium rod lattice are presented with a discussion of the principal parameters which influence reflector savings.

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INTRODUCTION

In the reprocessing of uranium fuels, situations develop, such as in the dissolution of fuel rods, which result in complex fuel configurations. During dissolution, possible systems range from bare fuel to rod lattices under optimum conditions of moderation and reflection, as well as homogeneous systems of dissolved fuel. Criticality safety must be given top priority in all of these situations. An important parameter in establishing criticality safety specifications for such processing steps is the reflector savings of the uranyl nitrate solutions which form during the dissolution.

Reflector savings takes on different significance for various geometries and circumstances. The reflector savings of a sphere is defined as the difference between the radius of an unreflected critical sphere and the radius of a critical sphere of the same material surrounded by a reflector of uniform thickness. For a cuboidal core (rectangular parallelepiped), reflected on one end, the savings is defined as the difference in length of the reflected and unreflected core. For reflection on two opposite ends of a cuboidal core, the savings is one-half the difference in length of the reflected and unreflected core. Values of reflector savings are presented for guidance in criticality safety evaluations involving uranyl nitrate of low enrichment.

In actual practice, it may be difficult to divide the system into a core and a reflector, since the system is continually changing as dissolution progresses. Every choice of core and reflector parameters will yield a somewhat different reflector savings. Spherical geometry probably gives values which are applicable to the largest number of problems. For this study, spherical cores of maximum buckling were used in the calculations of savings for uranium lattices in water. As a check on the accuracy of the reflector savings calculations, a series of

experiments were performed using a reflected cuboidal core composed of 2.2 wt% ^{240}Pu enriched PuO_2 -polystyrene blocks with an H/Pu ratio of 15. This material was used because it was readily available in a form which permitted the construction of clean, critical systems for direct comparison with computer calculations.

SUMMARY

Reflector savings of a 2.2 wt% ^{240}Pu enriched PuO_2 -polystyrene (H/Pu = 15) cuboid, reflected by uranyl nitrate of low enrichment, were measured using the ~~experimental~~ approach method. The effects of the stainless steel in the reflector solution container walls and of the air space at the core-reflector interface, caused by irregularities in the core and reflector surfaces, were experimentally determined. Comparison between measurements and HFN⁽¹⁾ and DTF⁽²⁾ computed reflector savings show these codes to compute values of savings which are slightly too large.

HFN and DTF were used to compute reflector savings for systems of water-moderated uranium-rod spherical lattices reflected by uranyl nitrate of *Less* than 5.0 wt% ^{235}U enrichment. Results for these two codes agreed very well for these systems and it is felt they well represent the true values. Variations of reflector savings as a function of several different parameters are examined. Reflector savings, as a function of k_{∞} , for a constant reflector thickness is nearly independent of the ^{235}U enrichment for the range of enrichments considered.

EXPERIMENT DESCRIPTION

PuO_2 -polystyrene cubes were stacked on both faces of a remote split-table machine⁽³⁾ so as to form a single compact, cuboidal array when the faces were closed. The uranyl nitrate reflector solutions were contained in stainless steel cans placed against the array of blocks. Figure 1 shows the arrangement

of blocks and reflector cans on the split-table machine. The inverse multiplication method was used to determine the number of rows of PuO_2 -polystyrene blocks required for criticality. This procedure was repeated with different uranium concentrations and with different reflector thicknesses. Throughout this experiment, the height and width of the assembly were kept constant; at 30.90 cm.

DETERMINATION OF CORE SIZE

An experiment was performed to demonstrate that reflectors on either side of the core act independently of each other for the assemblies being used in these measurements. Insufficient 2.2 wt% ^{240}Pu enriched fuel was available to construct a critical, unreflected, homogeneous core of this material, so a core was constructed with one bottom layer of 8.0 wt% ^{240}Pu enriched blocks and five layers of 2.2 wt% ^{240}Pu enriched blocks. This core could be made critical when unreflected and both the reflected and unreflected core lengths could be measured and the reflector savings determined. Using two identical water reflectors of 9.6 cm thickness, the reflector savings was found to be 4.05 cm for one end reflected, and 4.06 cm for two ends reflected. The close agreement of these values shows negligible interaction to occur between the reflectors.

Since the reflectors act independently, the reflector savings is equal to the difference in core length with one end reflected and with two ends reflected and the value so obtained can be used to determine the unreflected core length. The unreflected critical length of a homogeneous 2.2 wt% ^{240}Pu enriched PuO_2 -polystyrene core was determined to be $32.95 \pm .30$ cm by averaging 21 sets of measurements.

CORRECTIONS TO THE REFLECTOR SAVINGS

Two corrections were made to the measured value of the reflector savings. An adjustment was made to account for the stainless steel walls of the reflector cans and another to compensate for the leakage of neutrons from the air space

between the cope and reflector, Both corrections were evaluated by increasing %hesource of the perturbation, measuring the resulting change in savings, and then extrapolating to the situation where the undesirable element is removed.

The effect of the 50 mil stainless steel can walls was established by inserting up to three extra 50 mil sheets of stainless steel at each can wall and redetermining the core length. These results were used to extrapolate to a system with all stainless steel removed. Figure 3 shows the effect of the stainless steel on the thickness of the reflector. For thin reflectors the reflectivity is enhanced by the stainless steel, while for thick reflectors it is seduced, When the reflector thickness is about 8 cm, 50 mils of stainless steel has no effect on the savings.

The air space resulted primarily from a slight bowing of the reflector can walls and from small irregularities in the core face. In order to determine the effect of this gap, sheets of styrofoam were used as spacers to increase the width of the air space by a known amount. Spacers were used to guarantee that the air gap could be reproduced and also to prevent the cans from being accidentally moved closer to the core than desired, thereby producing a more reactive configuration. Figure 4 shows the dependence of the core size upon the width of the air space. For simplicity, the air space correction to the reflector savings was determined only for 9.6 cm of water reflection, and the value found was assumed to be valid for all concentrations of uranyl nitrate and reflector thickness used in this experiment. For thin air spaces, the reduction in savings was found to be slightly greater than the width of %hespace. The average width of the air space was estimated to be 0.20 cm, and this corresponds to a loss of 0.24 cm to the reflector savings.

Two sizes of reflector cans were used in this experiment: 4.7 cm and 9.6 cm. Multiples of these were use3 to obtain other reflector thicknesses.

Between the two 9.6 cm cans used to obtain a 19.2 cm reflector thickness, a second air space existed of the same size as the space at the core-reflector interface. With 9.6 cm of reflection the savings is more than 90% of its maximum value, and irregularities in the reflector at this distance from the core have a much smaller influence than they would have if they were nearer to the core. The effect of the second air space is several times smaller than the first and is ignored.

EXPERIMENTAL ERROR

TABLE 1

Sources of Experimental Error

1. Stainless Steel Correction	± 0.05 cm
2. Chemical Impurities	± 0.00 cm
3. Concentration Determination	± 0.05 cm
4. Precipitation	± 0.00 cm
5. Block Size and Stacking	3 0.10 cm
6. Reflector Can Positioning	± 0.13 cm
Experimental Error (Square Root of Sum of Squares)	± 0.18 cm

Given above in Table 1 are the sources of experimental error in this experiment. The probable magnitudes were determined as follows:

- (1) The uncertainty in the stainless steel corrections was estimated from a plot of core Length vs. steel thickness (Figure 2) by finding the root mean square variation in the extrapolated values using two points at a time.
- (2) A small amount of chemical impurity was found to be present in the solutions, but transport theory calculations showed the effects to be negligible.
- (3) Uranium concentrations were determined to within 20 g U/liter. For the sake of estimating the uncertainty one can assume the savings are a linear function of concentration between 0 and 200 g U/liter. The measured

difference in savings for reflectors of water and of 200 g U/liter is 0.5 cm.

An uncertainty of 20 g U/liter in concentration results in an uncertainty in reflector savings of about one-tenth this value, or 0.05 cm.

- (4) Precipitation was discovered in one of the 9.6 cm cans of 2.14 wt% ^{235}U enriched uranyl nitrate at a concentration of 600 g U/liter. The maximum change in reflector savings as a result of this precipitation is 0.10 cm. No other precipitate formed during these experiments.
- (5) The plutonium blocks vary slightly from each other in size, and small irregularities in the core resulted when the blocks were stacked. An estimated uncertainty of 0.10 cm in core size resulted from the variations in block size and in the stacking of the blocks.
- (6) Using the average core size, one can find two values of the reflector savings. One value can be determined from the core size with a reflector on one end, and the other value can be found from the core size with reflectors on two ends. The difference between corresponding values of the savings is an estimate of the uncertainty in measuring them. Using the measured data, the root mean square uncertainty in the reflector savings is ± 0.16 cm. This is a combination of the errors resulting from the stacking of blocks and from the positioning of reflector cans. Since the estimated error in block stacking is ± 0.10 cm, the uncertainty from the positioning of the cans would be approximately ± 0.13 cm. This agrees with the error estimated by considering the possible variations in the width of the air space between the core and reflector. Since the maximum fluctuation of the space width is estimated to be ± 0.10 cm from the mean value, the uncertainty in the reflector savings would be ± 0.12 cm, as determined from Figure 4.

COMPARISON OF EXPERIMENTAL AND COMPUTED VALUES OF REFLECTOR SAVINGS

COMPUTATIONAL METHODS

Five computer programs were used in obtaining the reported analytical results: HFN,⁽¹⁾ DTF,⁽²⁾ THERMOS,⁽⁴⁾ GAMTEC,⁽⁵⁾ EGGNIT,⁽⁶⁾. Each calculation required the use of at least two programs: one to generate the cross sections and the other to solve the diffusion transport equations. GAMTEC generated 18 group cross sections for the homogeneous uranyl nitrate reflectors and punched them onto cards for use in HFN and DTF. For the uranium-water lattices, EGGNIT, a modified version of GAMTEC, was used because it performs a better resonance integral for lattice calculations. Since EGGNIT uses only one thermal group, THERMOS, which uses 30 thermal groups, should produce better thermal parameters. THERMOS data was substituted for the thermal parameters in the punched EGGNIT cards,

Cross sections from the above three codes were used in HFN and DTF to calculate the critical dimensions of the reflected systems. HFN solves the multigroup, multiregion diffusion equation in one space dimension while DTF solves the corresponding transport equation with anisotropic scattering.

COMPARISON OF HFN, DTF, AND REFLECTED CUBOID EXPERIMENTAL DATA

Both HFN and DTF overestimate the uranyl nitrate reflector savings of an end-reflected PuO_2 -polystyrene core. Table 2 gives values for comparison of calculated and experimental results. For 19.2 cm of reflection HFN values are 0.4 to 0.9 cm (10-20%) larger than the experimental values. DTF does better by giving values only 0.1 to 0.5 cm (5-10%) high. Much of this discrepancy probably results from improper leakage corrections by the codes.

A difficulty exists in the interpretation of the experimental data for 2.14 wt% ^{235}U enriched uranyl nitrate - the reflector savings at 394 g U/liter was slightly less than the value at 216 g U/liter (see Figure 6). In comparison

TABLE 2

Uranyl Nitrate Reflector Savings for a 2.2 wt% ^{240}Pu Enriched PuO_2 -Polystyrene Cuboidal Core ($\text{H}/\text{Pu} = 15$)

Thickness (cm)	Concentration g U/l	Density (g/cc)	H/U	REFLECTOR SAVINGS (CM)		
				Experimental	HEU	DTF
<u>Water</u>						
4.7	Water	1.00	15.0	3.27	3.60	4.00
9.6	"	"	"	3.80	4.50	4.17
19.2	"	"	"	3.93	4.58	4.22
<u>2.14 Wt% ²³⁵U Enrichment</u>						
4.7	216 ± 20	1.27	105	3.48	3.55	3.60
9.6	" "	"	"	4.34	4.75	4.39
19.2	" "	"	"	4.50	5.00	4.59
4.7	394 ± 10	1.52	59	3.08*	3.75	3.60
9.6	" "	"	"	4.13	5.05	4.70
19.2	" "	"	"	4.38	5.54	5.07
4.7	596 ± 10	1.79	36	3.12	3.65	3.60
9.6	" "	"	"	4.50	5.28	4.30
19.2	" "	"	"	5.00	5.76	5.23
4.7	1390 ± 20	2.67	3.8	2.22	2.27	2.30
9.6	" "	"	"	3.17	3.29	3.30
19.2	" "	"	"	3.62	3.67	3.79
<u>3.04 Wt% ²³⁵U Enrichment</u>						
4.7	550 ± 20	1.73	40	3.04	3.60	3.60
9.6	" "	"	"	4.67	5.00	4.60
19.2	" "	"	"	5.17	6.22	5.74
4.7	870 ± 15	1.65	8.2	1.86	2.20	2.20
19.6	" "	"	"	2.42	2.60	2.60
19.2	" "	"	"	3.00	3.00	3.00

*Values for 394 g U/l appear low relative to other measurements.

with the reflector savings for water and the values of reflector savings for concentrations of 216 g U/liter and 596 g U/liter, the reflector savings for the 394 g U/liter solution appears too small, although the precise reason has not been determined. Calculations do not show a dip in the savings vs. concentration curve at this concentration.

Richey⁽⁷⁾ examined water-reflected, well-moderated $\text{Pu}(\text{NO}_3)_4$ spheres and cylinders of low ^{240}Pu isotopic enrichment and concluded that the critical parameters could be reproduced using HFN with a good degree of accuracy. Hansen⁽⁸⁾ studied well-moderated plutonium systems of ^{240}Pu isotopic enrichment $\leq 25\%$ and found HFN to give quite accurate size determinations for all geometries. Calculations of undermoderated systems of ^{240}Pu isotopic enrichment $\leq 12\%$ using DTF give reasonably good agreement (to about 1.0%) for all geometries, except reflected slabs. This failure with reflected slabs is the result of GAMTEC-II not accounting for the spectral shift for this geometry when averaging the energy-dependent cross sections.

DISCUSSION OF SPHERICAL LATTICE CALCULATIONS

DESCRIPTION OF SYSTEMS STUDIED

The most reactive form of slightly enriched uranium is a lattice of uranium rods in water with the proper diameter and spacing. Table 3 gives a list of the parameters of the cores used in the reflector savings calculations. These configurations are very near to the most reactive configurations. Using each of the lattices given in Table 3, a set of curves was generated using HFN giving the savings for reflectors of thicknesses of up to 50 cm and of uranium concentrations of up to 1000 g U/liter. Table 4 lists the reflector compositions used. The enrichment of reflector and core are kept the same. Using 3.0 and 5.0 wt% ^{235}U enriched uranyl nitrate, several DTF checkpoints were computed and agreement with HFN is very good. With 20.3 cm (8-inch) reflectors the two codes agree with each other to less than 0.3 cm for all concentrations of uranium in the reflectors.

TABLE 3

Parameters Used in Calculations for Spherical Lattices

^{235}U Enrichment (wt%)	Rod Diameters (cm)	Center-to-Center Spacing (cm)	Buckling* (cm^{-2})	k_{∞} *
2.0	1.270	2.238	0.0110	1.29
3.0	1.016	2.144	0.0152	1.41
4.0	0.762	1.706	0.0182	1.48
5.0	0.762	1.706	0.0200	1.52

TABLE 4

Composition of Uranyl Nitrate Reflectors
Used in Spherical Lattice Calculations

^{235}U wt%	g U/l	H/U	k_{∞} *		^{235}U wt%	g U/l	H/U	k_{∞} *
2.0	200	125	0.430		4.0	200	125	0.710
"	400	58.5	0.654		"	400	58.5	0.972
"	600	36	0.791		"	600	36	1.106
"	800	24	0.881		"	800	24	1.170
"	1000	17.5	0.932		"	1000	17.5	1.215
3.0	200	125	0.586		5.0	200	125	0.813
"	400	58.5	0.836		"	400	58.5	1.077
"	600	36	0.976		"	600	36	1.202
"	800	24	1.060		"	800	24	1.267
"	1000	17.5	1.103		"	1000	17.5	1.293

*Calculated by GAMTEC-II

HFN computed critical radii of fully water-reflected spheres shown in Table 5 are up to 5% larger than the values reported by Clark⁽⁹⁾. This difference reflects the difficulty of obtaining proper energy and space averaged cross sections for a lattice. Lattice cross sections must be first averaged over a unit lattice cell before they can be used in HFN.

Table 5
Comparison of HFN-Calculated
Water-Reflected Radii and Reported Radii

^{235}U Enrichment <u>(wt%)</u>	Bare Critical Radii (cm)	Water Reflected Critical Radii (cm)	
	<u>HFN</u>	<u>HFN</u>	<u>DP-1014⁽⁹⁾</u>
2.0	27.65	23.96	22.94
3.0	23.05	19.60	19.21
4.0	20.98	17.53	---
5.0	20.02	16.56	15.79

PARAMETERS AFFECTING REFLECTOR SAVINGS

Parameters influencing reflector savings fall into three categories: (1) core parameters, (2) reflector parameters, and (3) geometry. Core composition determines the core's diffusion coefficient and buckling. A large diffusion coefficient produces a greater leakage probability and consequently more neutrons leaving the core to be returned by the reflector. A large buckling signifies a more reactive core material whose replacement would require a greater number of reflected neutrons. Reflector composition determines the diffusion coefficient, absorption cross section, and multiplication of the reflector. A large diffusion coefficient in the reflector allows the neutrons to penetrate well within the reflector and decreases the chances of returning to the core. Multiplication increases the worth of the reflector and correspondingly increases the savings. Absorption in the reflector decreases savings. Geometry has a considerable

effect on reflector savings, since leakage from the system depends greatly on the surface area, shape and arrangement of the core material.

Reflector thickness is usually chosen as the independent variable and all other parameters are held constant. It is also instructive to choose other independent variables, such as k_{∞} , of the reflector and examine their effect on the reflector savings. The more important variables are the following:

(a) Reflector Thickness

Variations of reflector savings with reflector thickness depend greatly on the uranium concentration and enrichment (see Figures 7 - 10). Ten centimeters of water has a reflector savings which is more than 95% of its maximum value. When enriched uranyl nitrate is added to the water, a greater thickness of reflector is required for the reflector savings to reach 95% of its maximum value. When k_{∞} of the reflector is greater than unity, there is no longer an asymptotic approach to a constant value. Instead, an inflection point is reached on the reflector savings curve beyond which the slope of the curve increases with increasing thickness. At one particular thickness, the core is no longer needed and the reflector becomes critical by itself.

(b) Reflector Uranium Concentration

Uranium concentration influences both the moderation and the neutron multiplication of the reflector. Variations in moderation affects both the spectrum of the neutrons returning to the core and the neutron spectrum within the reflector. Neutron multiplication within the reflector is affected by the ^{235}U concentration and by the neutron spectrum.

(c) Reflector Uranium Enrichment

For uranium concentrations up to 1000 g U/liter and reflector thickness less than 10 cm, the reflector savings is an insensitive function of enrichment

(see Figure 13) in the range of enrichments considered, larger thicknesses of reflectors show a much greater influence on reflector savings as the enrichment is changed (see Figures 14-16).

(d) Reflector Infinite Multiplication Constant

The similarity of curves of k_{∞} plotted against reflector savings (with reflector thickness held constant) for different enrichments can be readily seen in Figure 17. A single curve could be used to approximate all of these curves to a fair degree of accuracy. Curves of lower enrichment lie slightly above those of higher enrichment, primarily as a result of differences in moderation.

(e) Core Composition

Reflector savings depends on core composition, Figure 11 shows the savings for 600 g U/liter reflectors of 3.0 and 5.0 wt% ^{235}U enriched uranyl nitrate to be greater with a 3.0 wt% ^{235}U enriched core than for a 5.0 wt% ^{235}U enriched core. Both of these reflectors have a hydrogen to uranium ratio of 36, indicating that H/U of 36 is a more optimum reflector moderation relative to the 3.0 wt% ^{235}U enriched core than relative to the 5.0 wt% ^{235}U enriched core. A water reflector produces the same savings for both of the above cores,

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FIGURE 1
Split Table Machine Showing the Arrangement of Plutonium Blocks and Reflector
Cans Prior to a Critical Approach

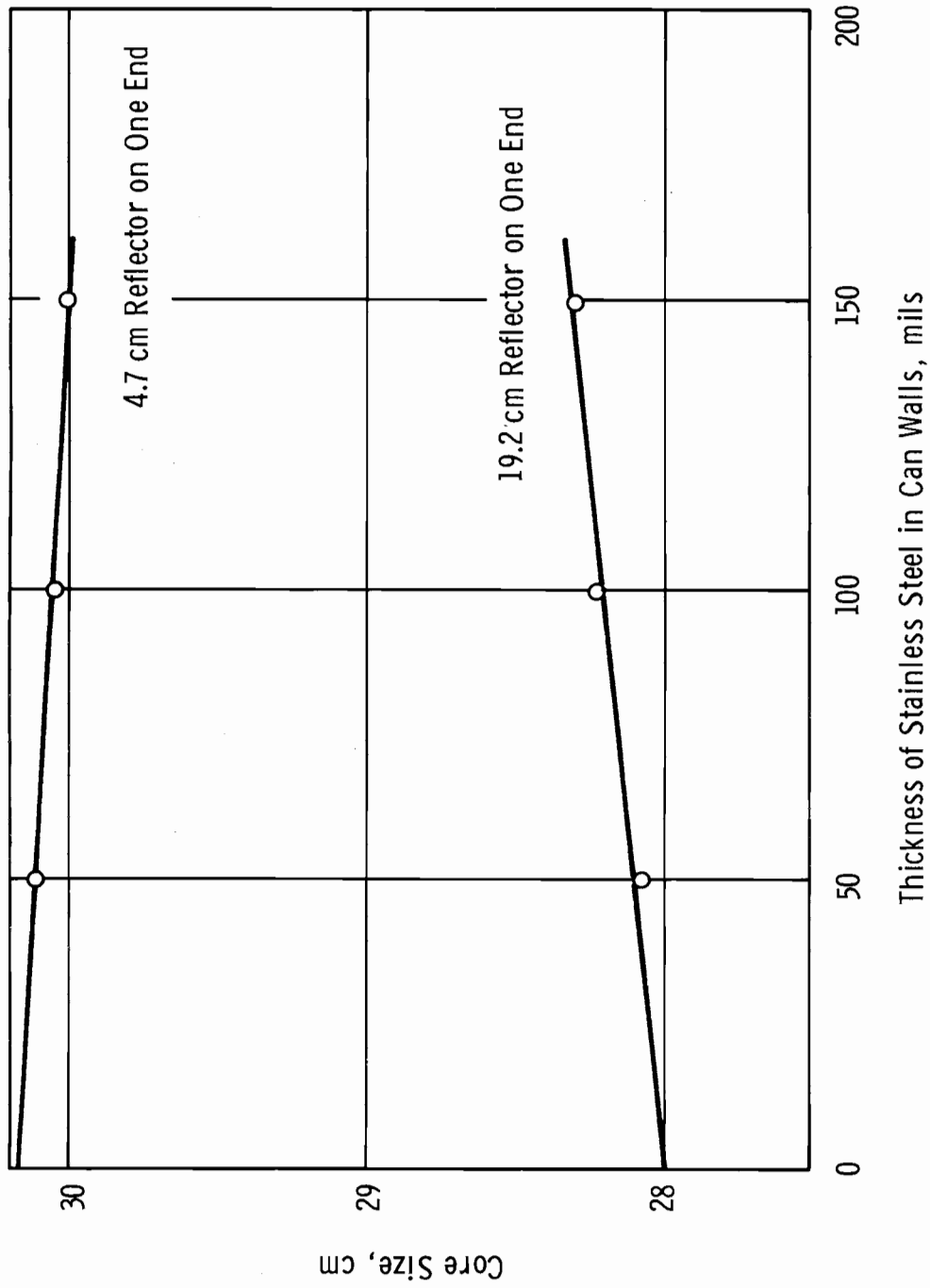


FIGURE 2
Stainless Steel Thickness at Core-Reflector Interface Plotted Against Core Size

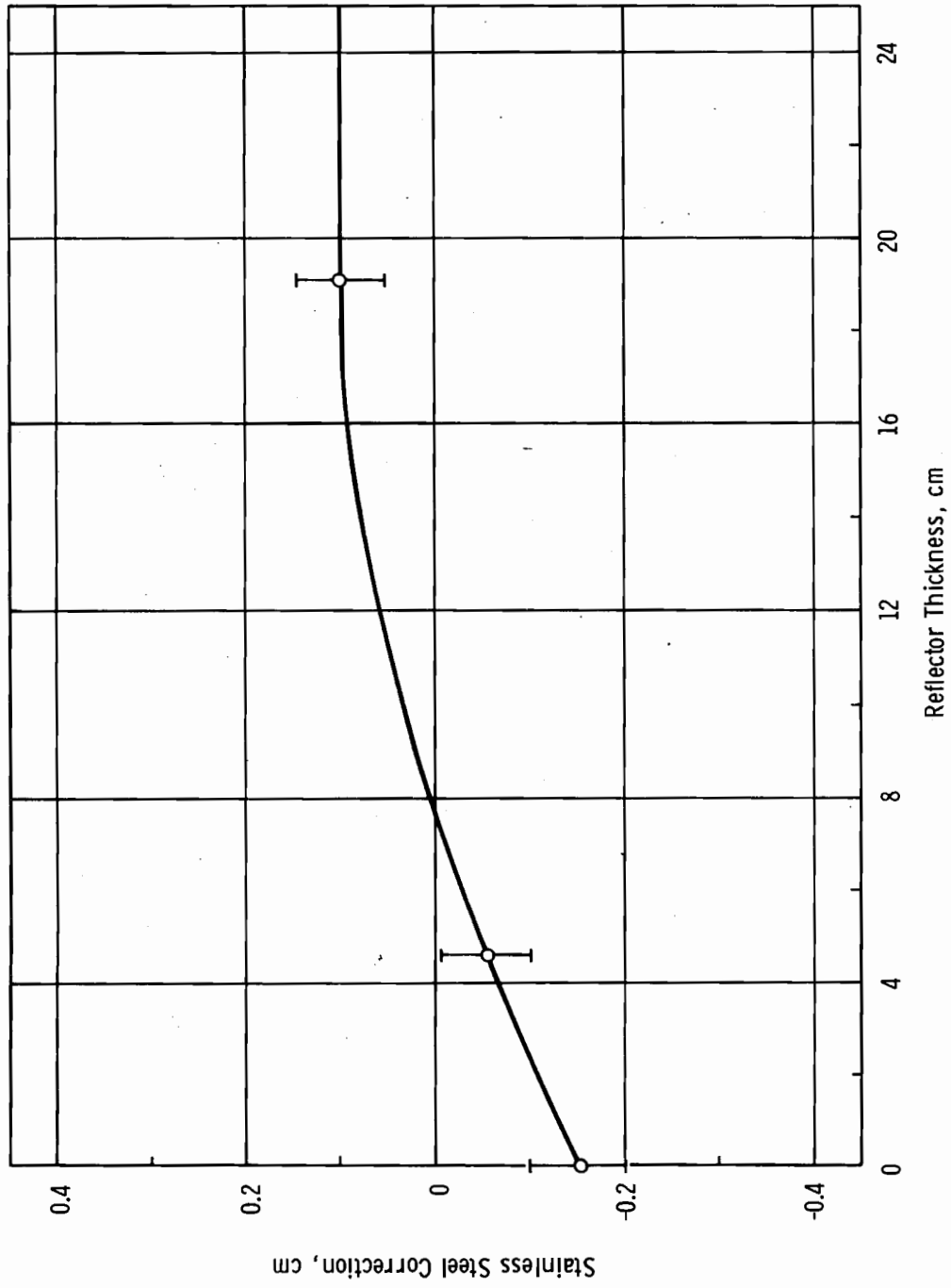


FIGURE 3
Correction for Stainless Steel in Cans Plotted Against Reflector Thickness

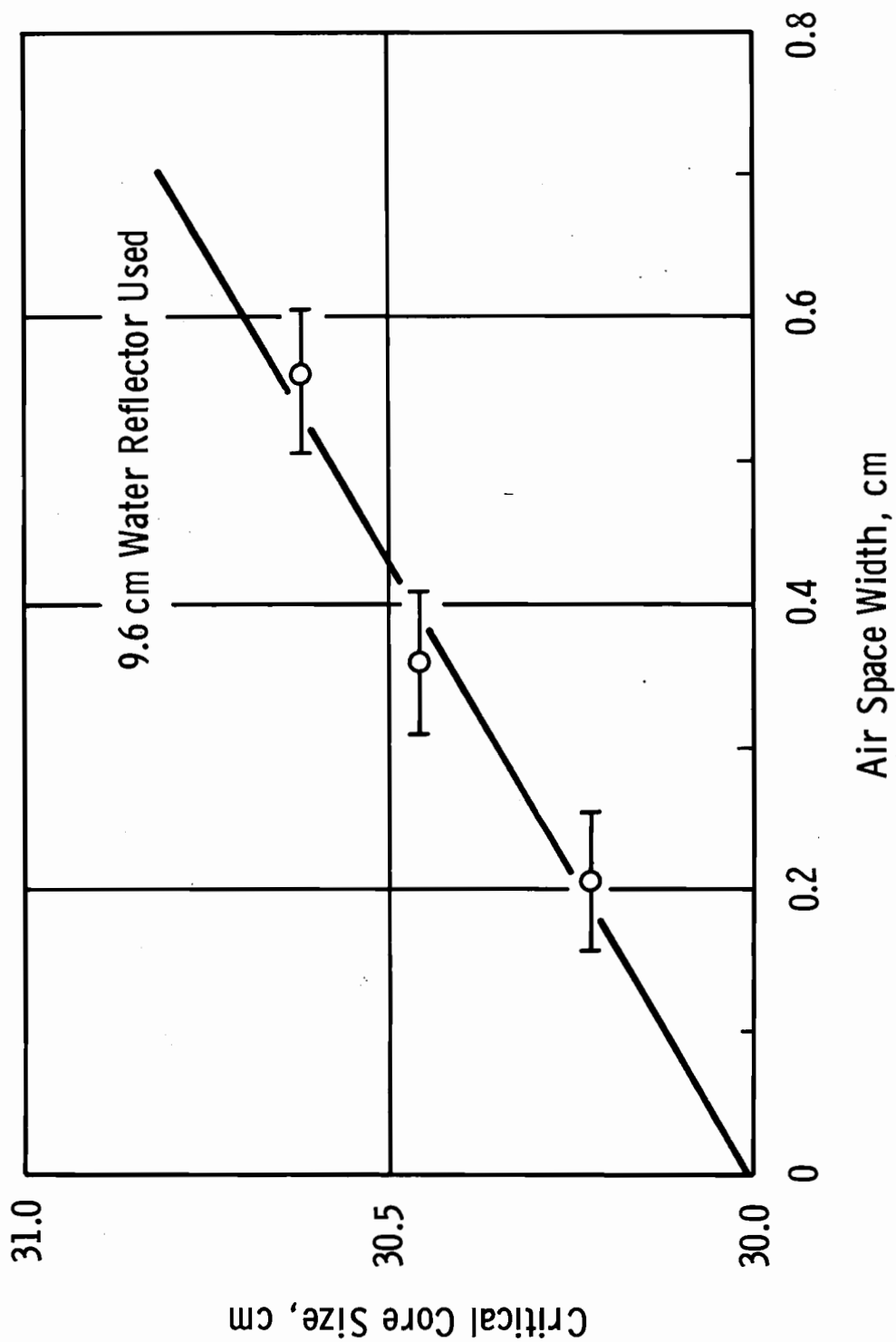


FIGURE 4
Air Space Width at Core-Reflector Interface Plotted Against Length of Critical Core

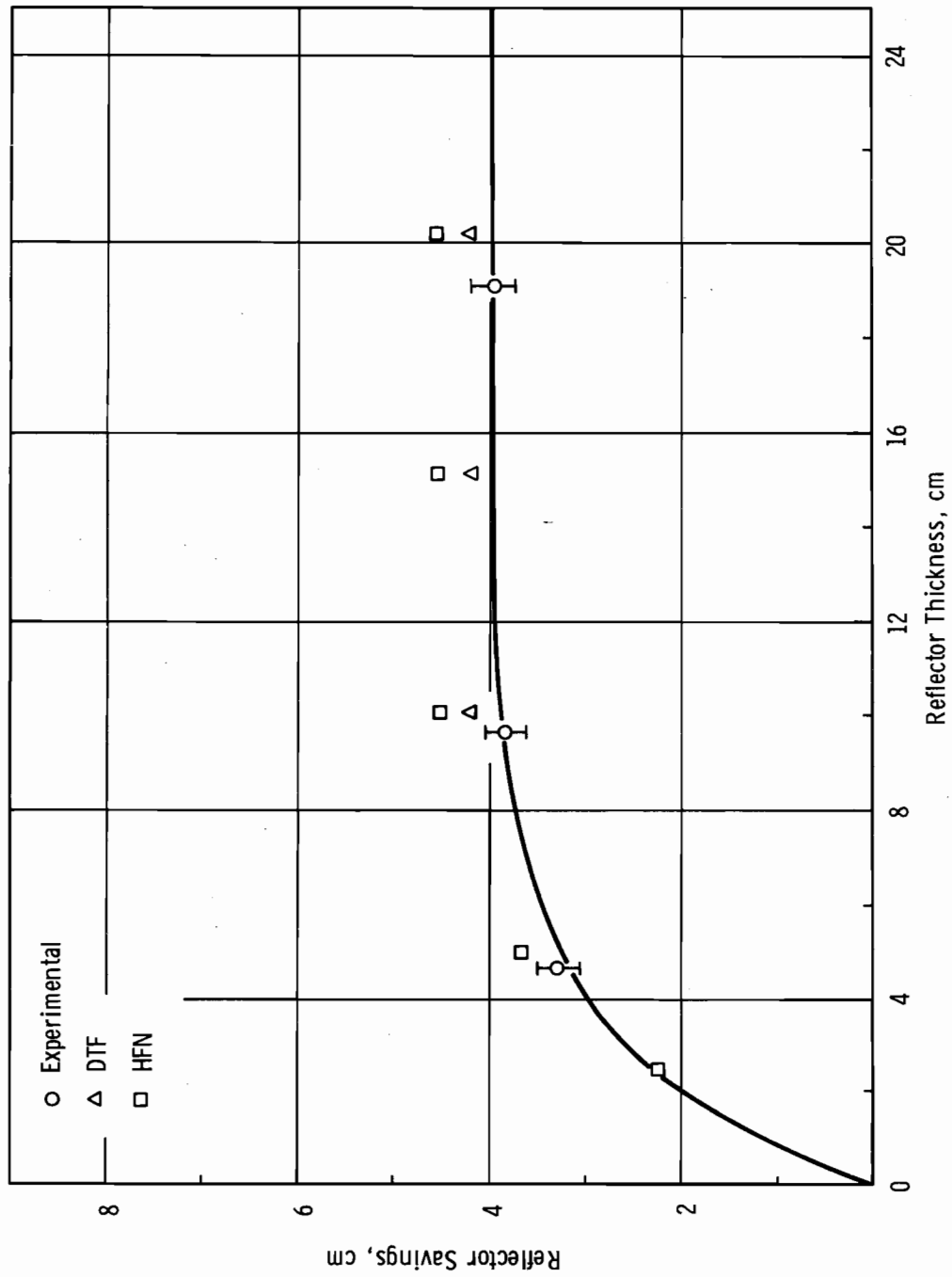


FIGURE 5
Reflector Savings Curve for Water Reflected Cuboid of PuO_2 -Polystyrene

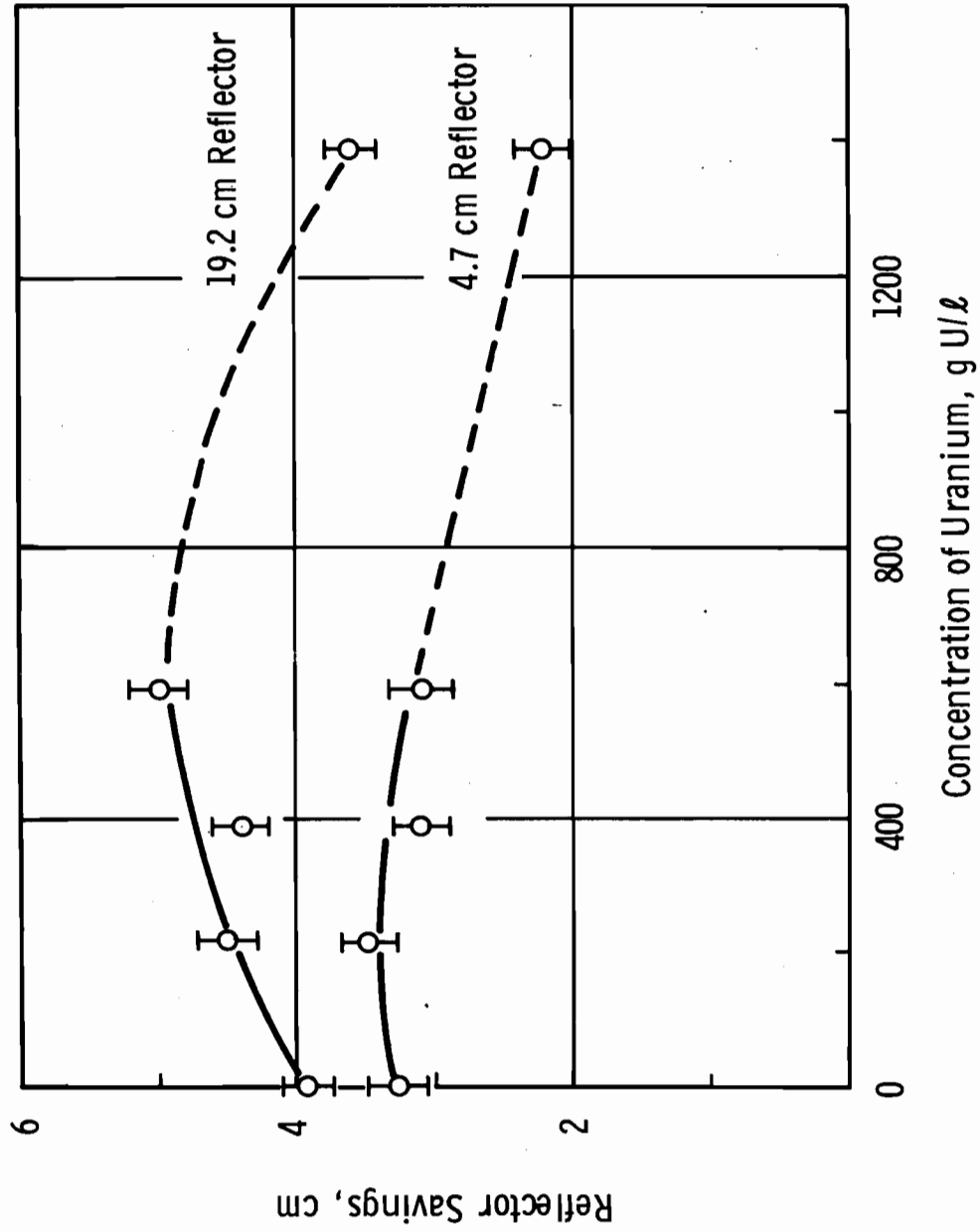


FIGURE 6
Reflector Savings vs Uranium Concentration for 2.14 wt% ^{235}U Enriched Uranyl
Nitrate Reflected Cuboid of PuO_2 -Polystyrene

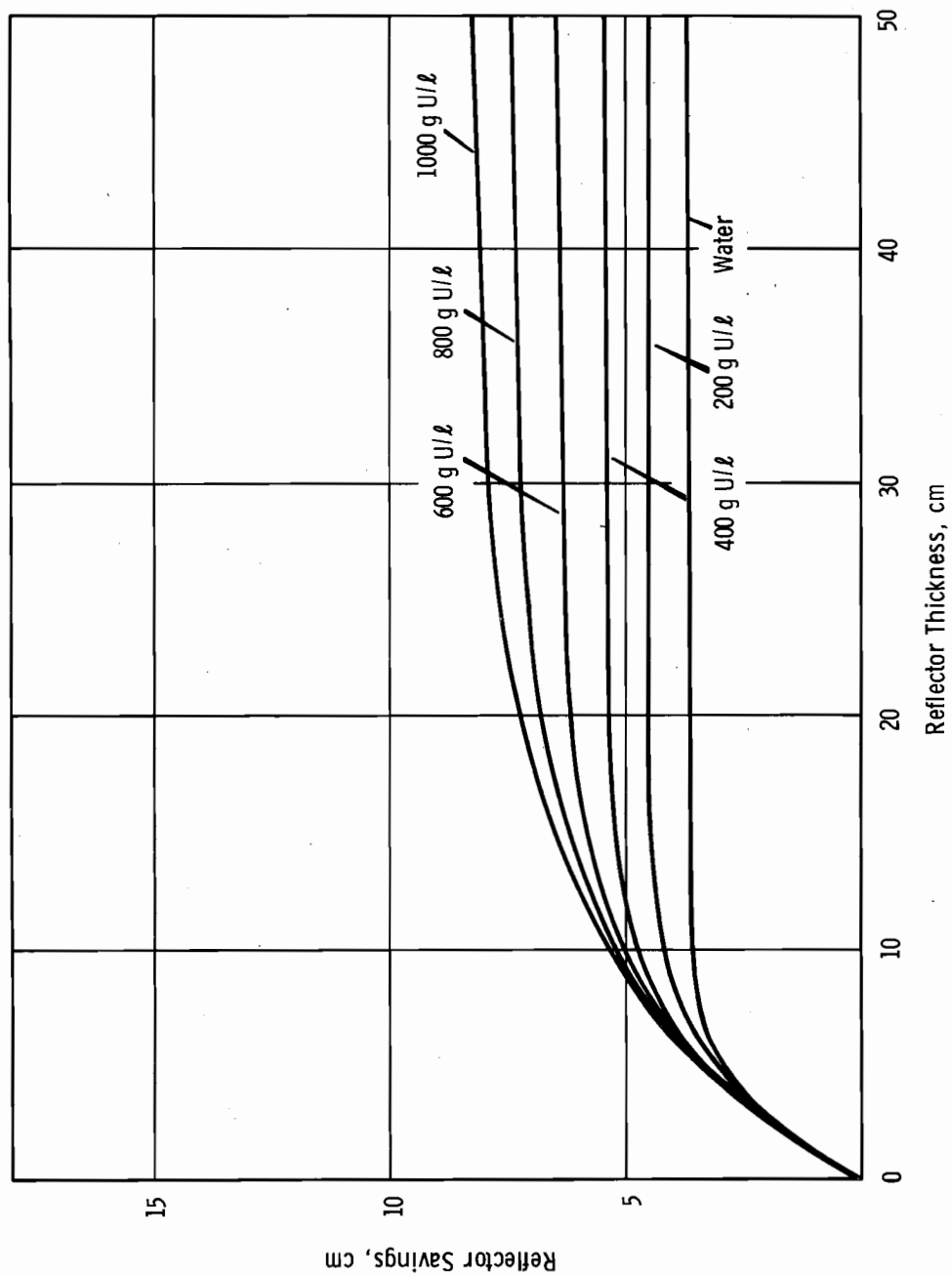


FIGURE 7

Reflector Savings for 2.0 wt% ^{235}U Enriched Uranyl Nitrate Reflecting a Spherical Lattice of 2.0 wt% ^{235}U Enriched Uranium Rods in Water

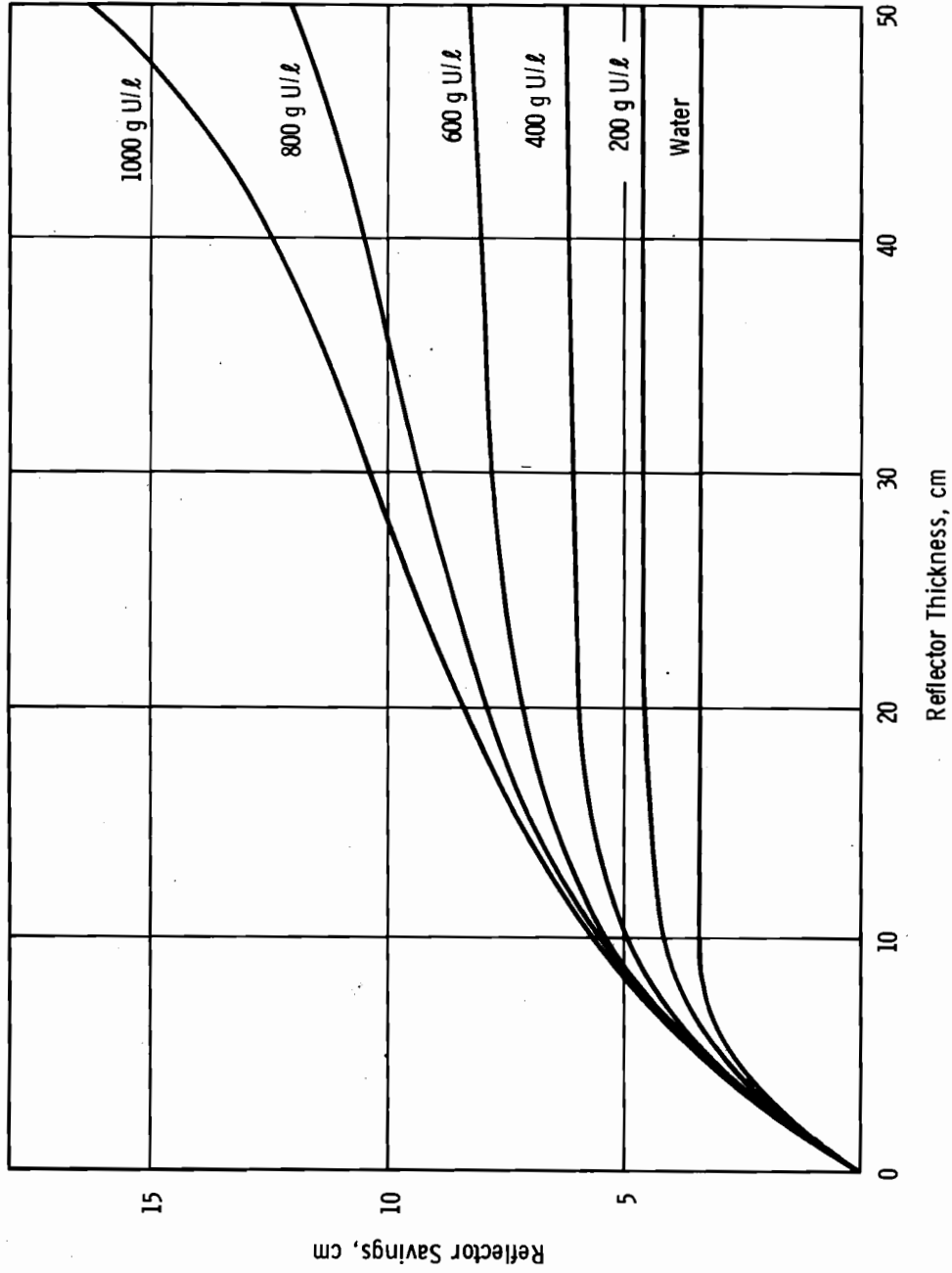


FIGURE 8

Reflector Savings for 3.0 wt% ^{235}U Enriched Uranyl Nitrate Reflecting a Spherical Lattice of 3.0 wt% ^{235}U Enriched Uranium Rods in Water

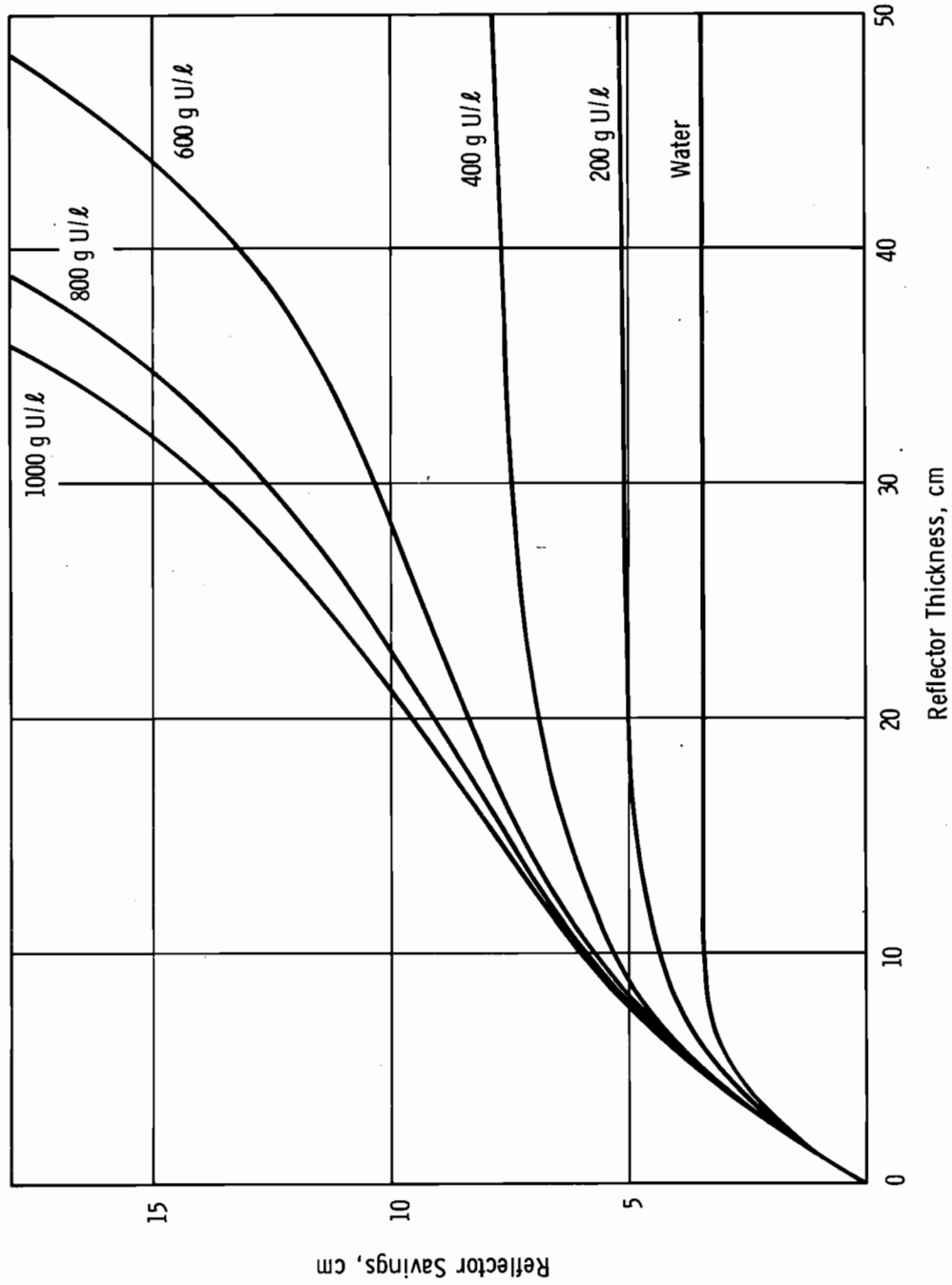


FIGURE 9

Reflector Savings for 4.0 wt% ^{235}U Enriched Uranyl Nitrate Reflecting a Spherical Lattice of 4.0 wt% ^{235}U Enriched Uranium Rods in Water

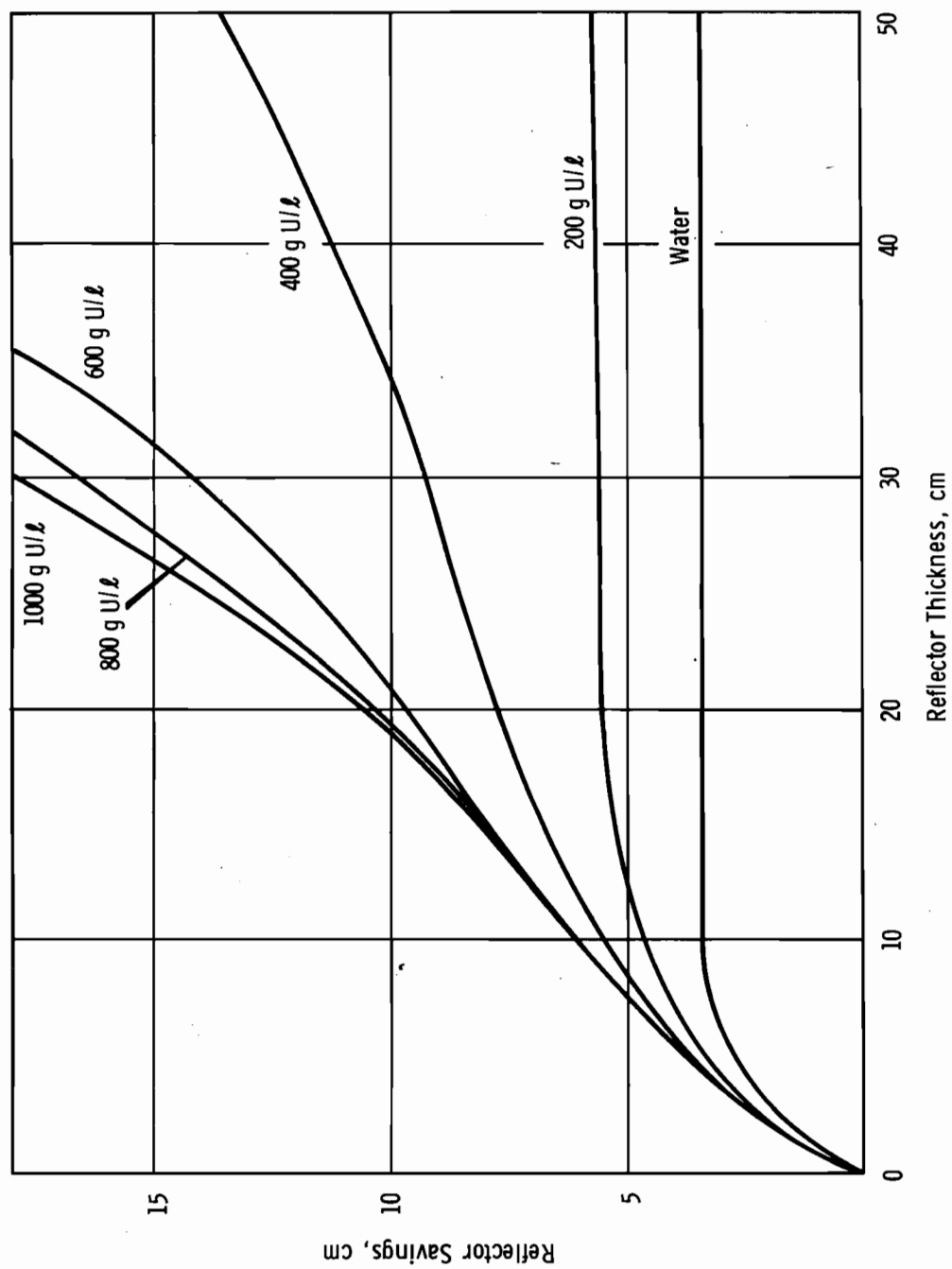


FIGURE 10

Reflector Savings for 5.0 wt% ^{235}U Enriched Uranyl Nitrate Reflecting a Spherical Lattice of 5.0 wt% ^{235}U Enriched Uranium Rods in Water

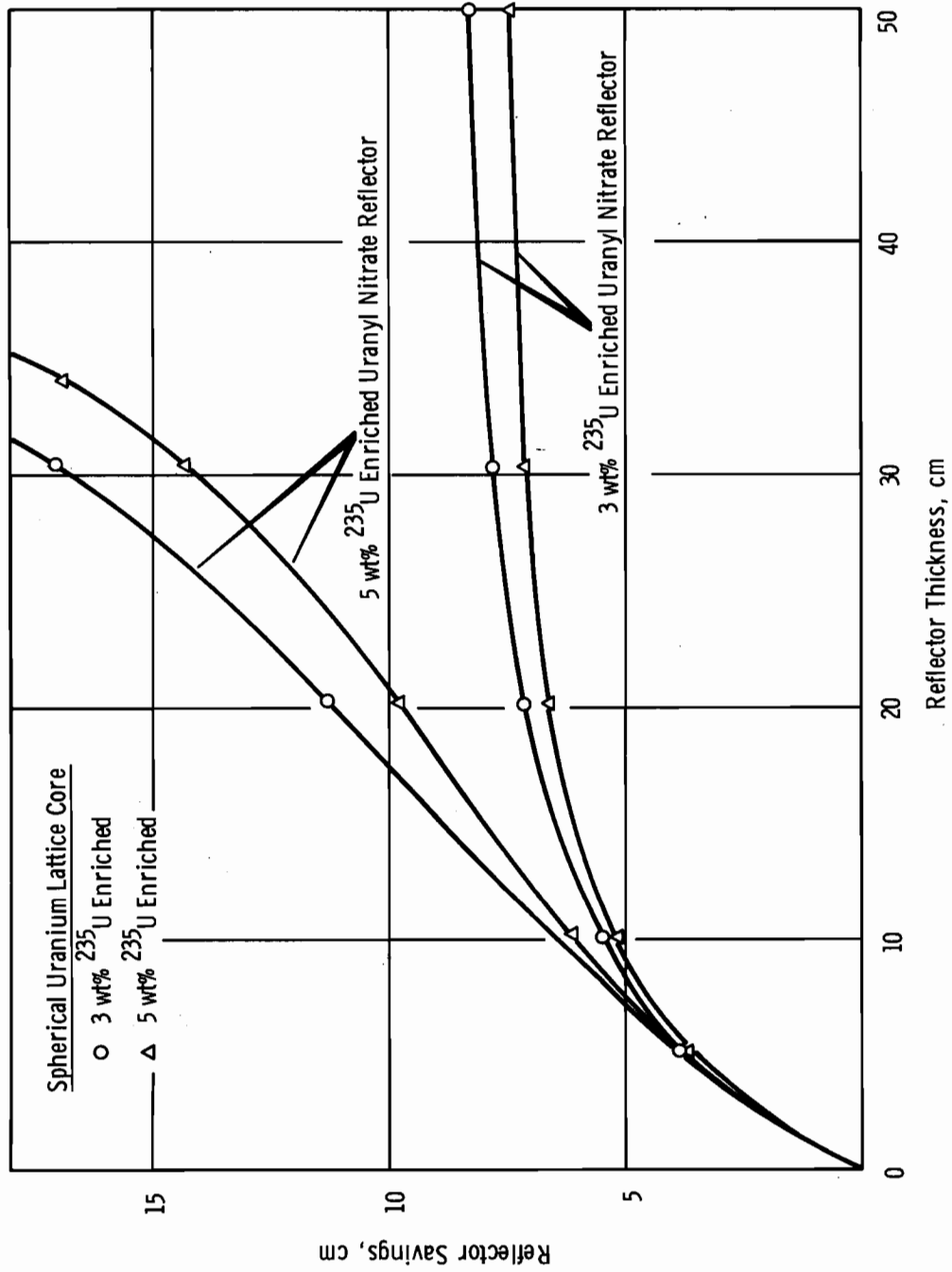


FIGURE 11
Reflector Savings for Uranyl Nitrate at 600 g U/l

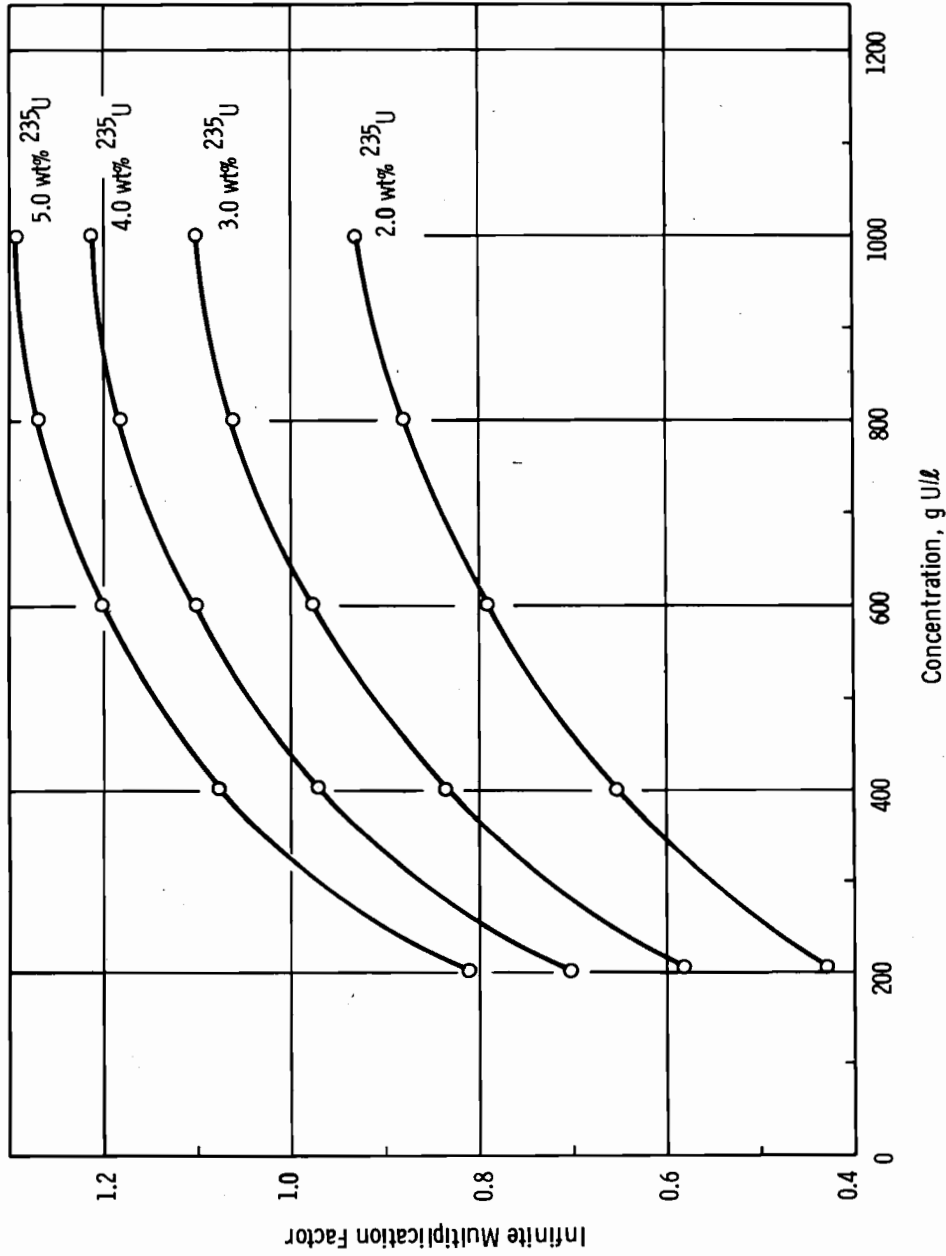


FIGURE 12
 k_{∞} of Homogeneous Uranyl Nitrate as a Function of Uranium Concentration and ^{235}U Enrichment

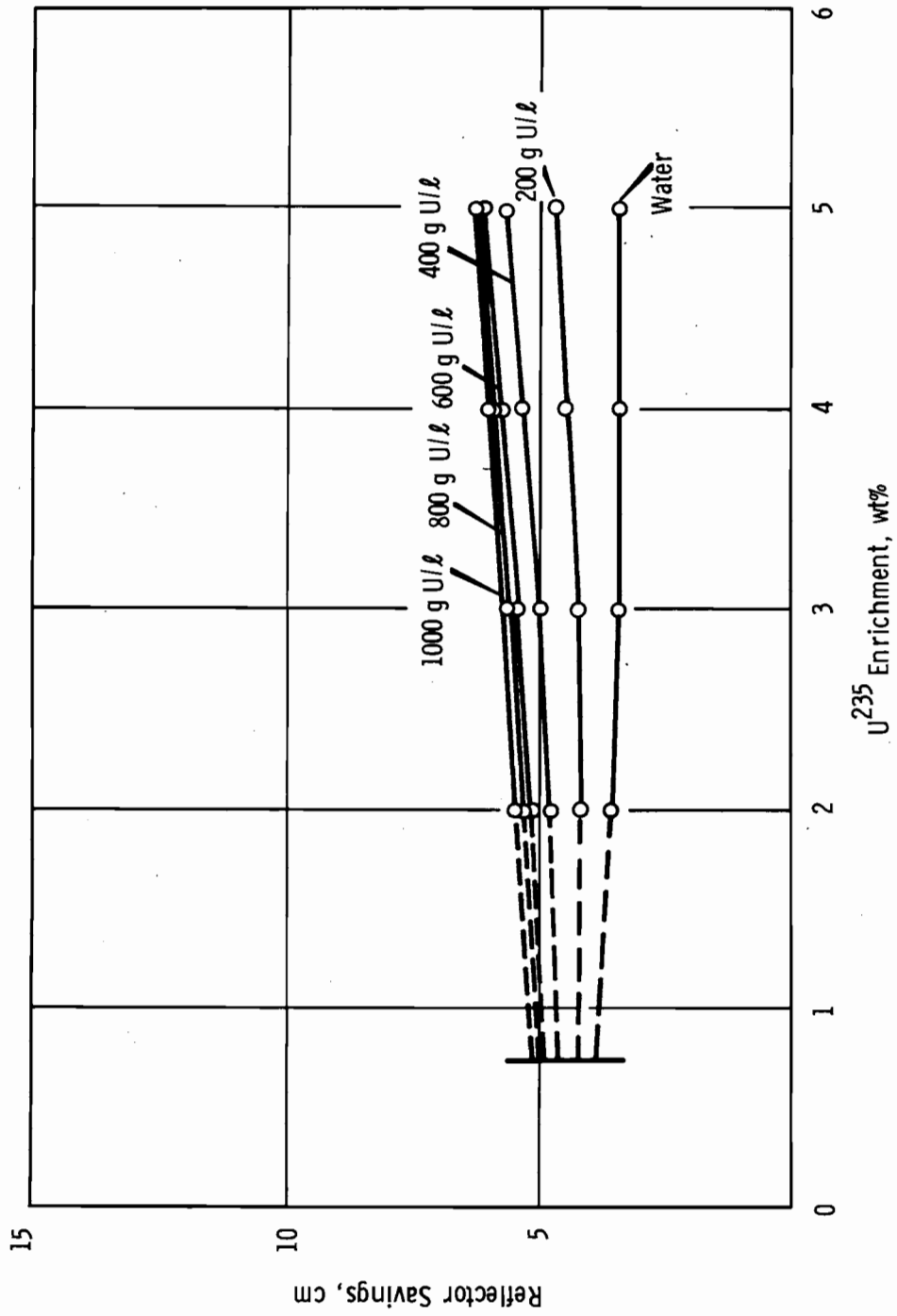


FIGURE 13
Reflector Savings as a Function of Enrichment for a 10.16 cm Thick Uranyl Nitrate Reflector Surrounding a Spherical Uranium Lattice

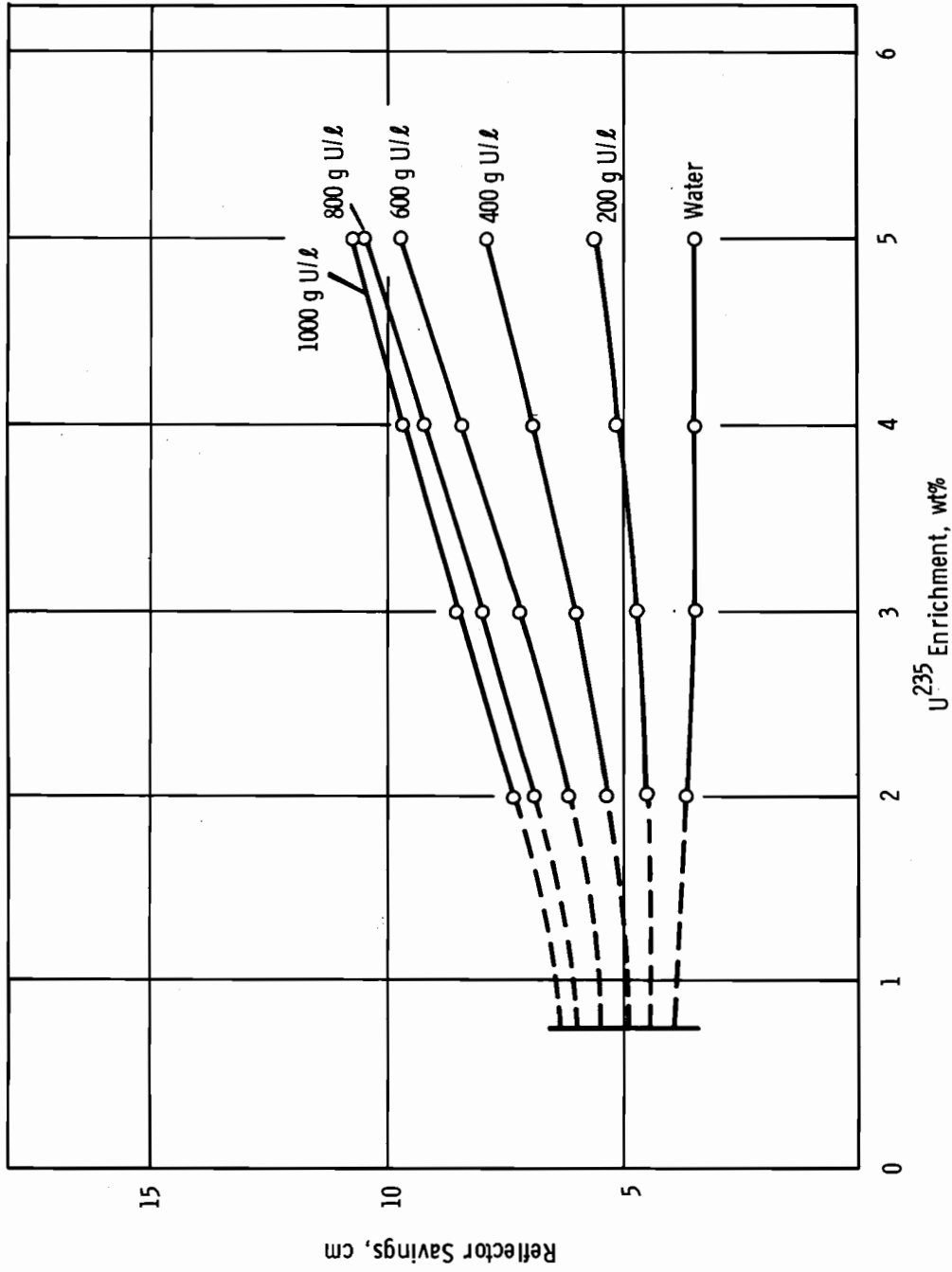


FIGURE 14
 Reflector Savings as a Function of Enrichment for a 20.32 cm Uranyl Nitrate
 Reflector Surrounding a Spherical Uranium Lattice

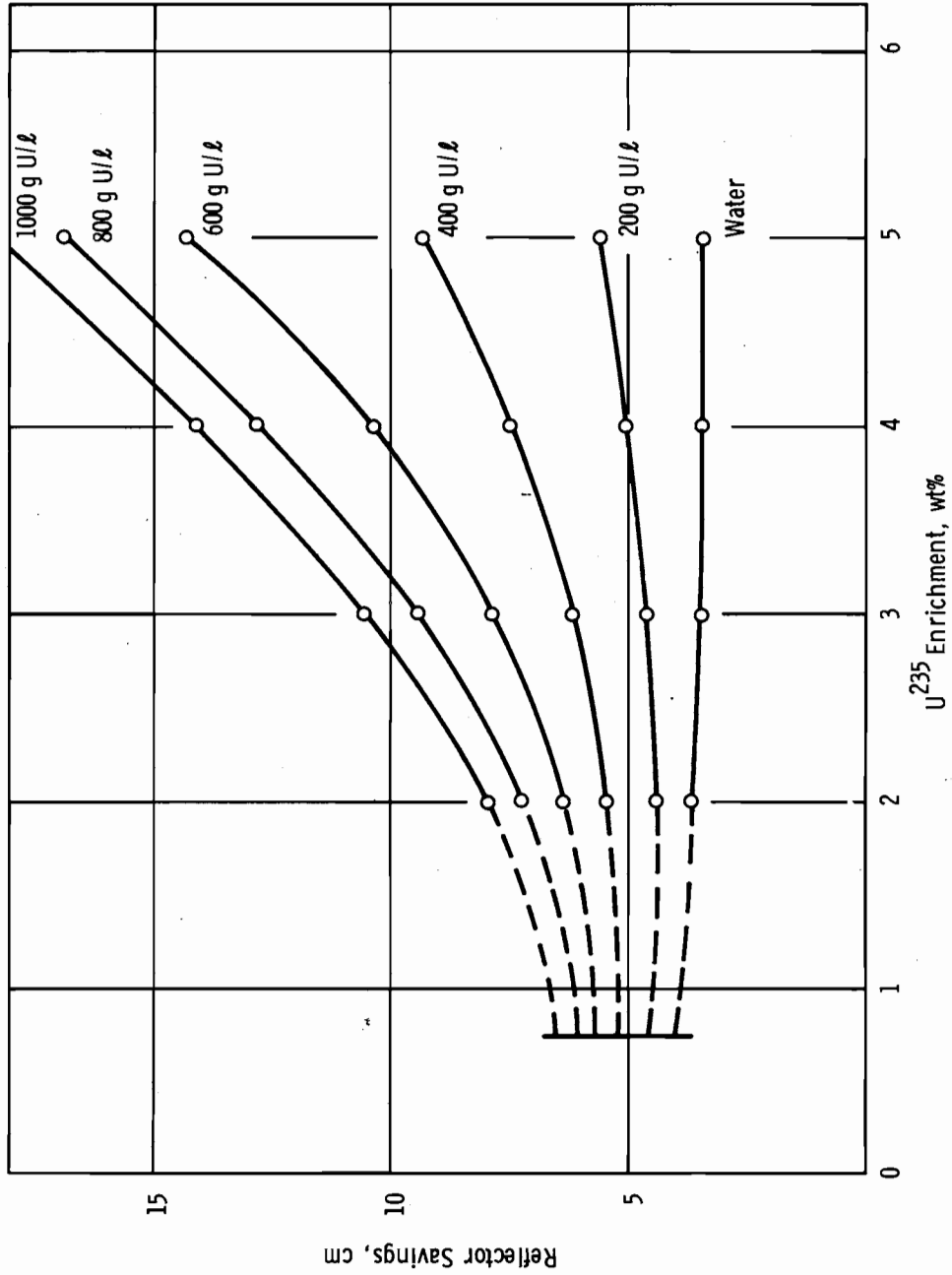


FIGURE 15
Reflector Savings as a Function of Enrichment for a 30.48 cm Thick Uranyl
Nitrate Reflector Surrounding a Spherical Uranium Lattice

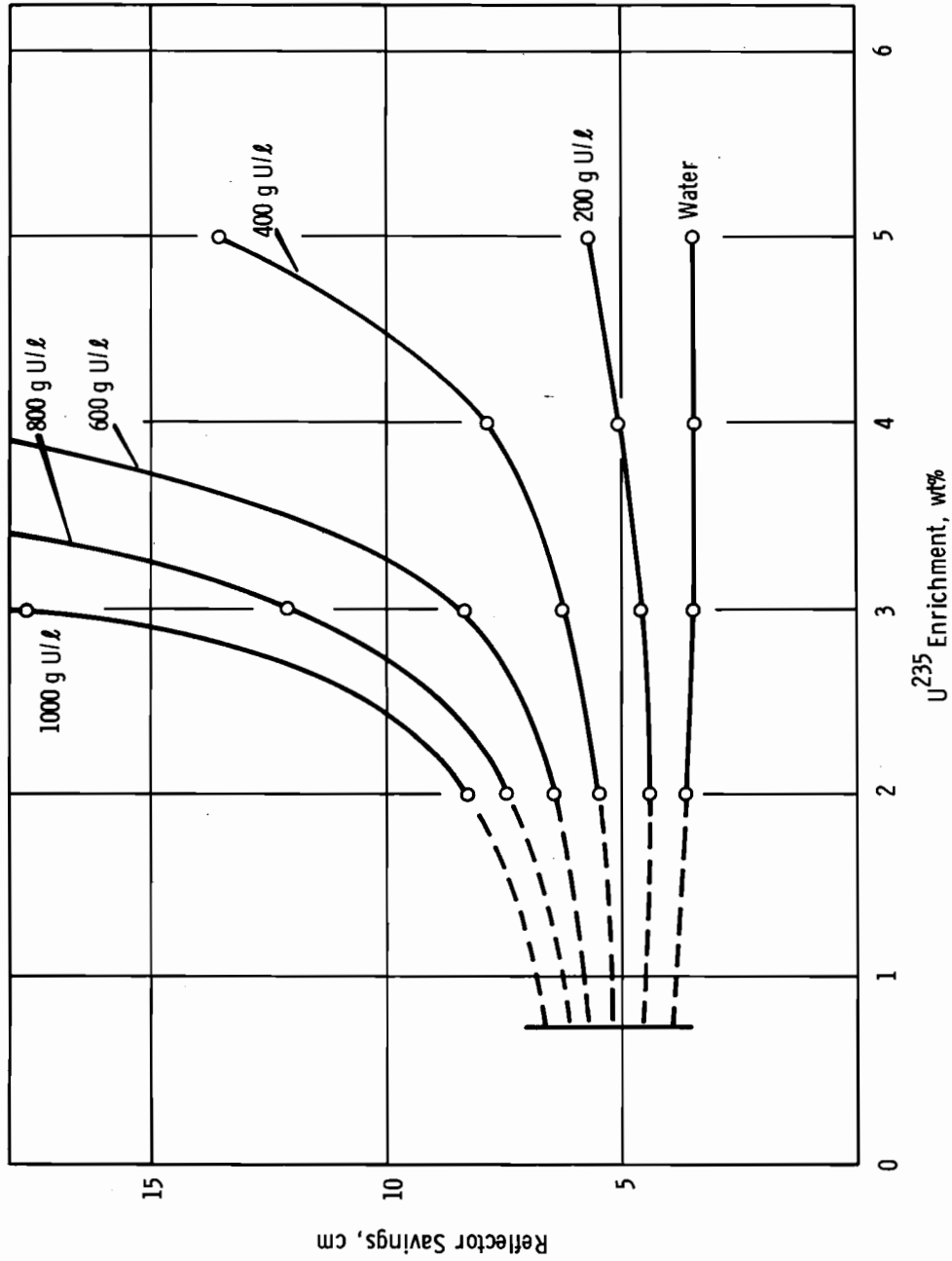


FIGURE 16

Reflector Savings as a Function of Enrichment for a 50.80 cm Thick Uranyl Nitrate Reflector Surrounding a Spherical Uranium Lattice

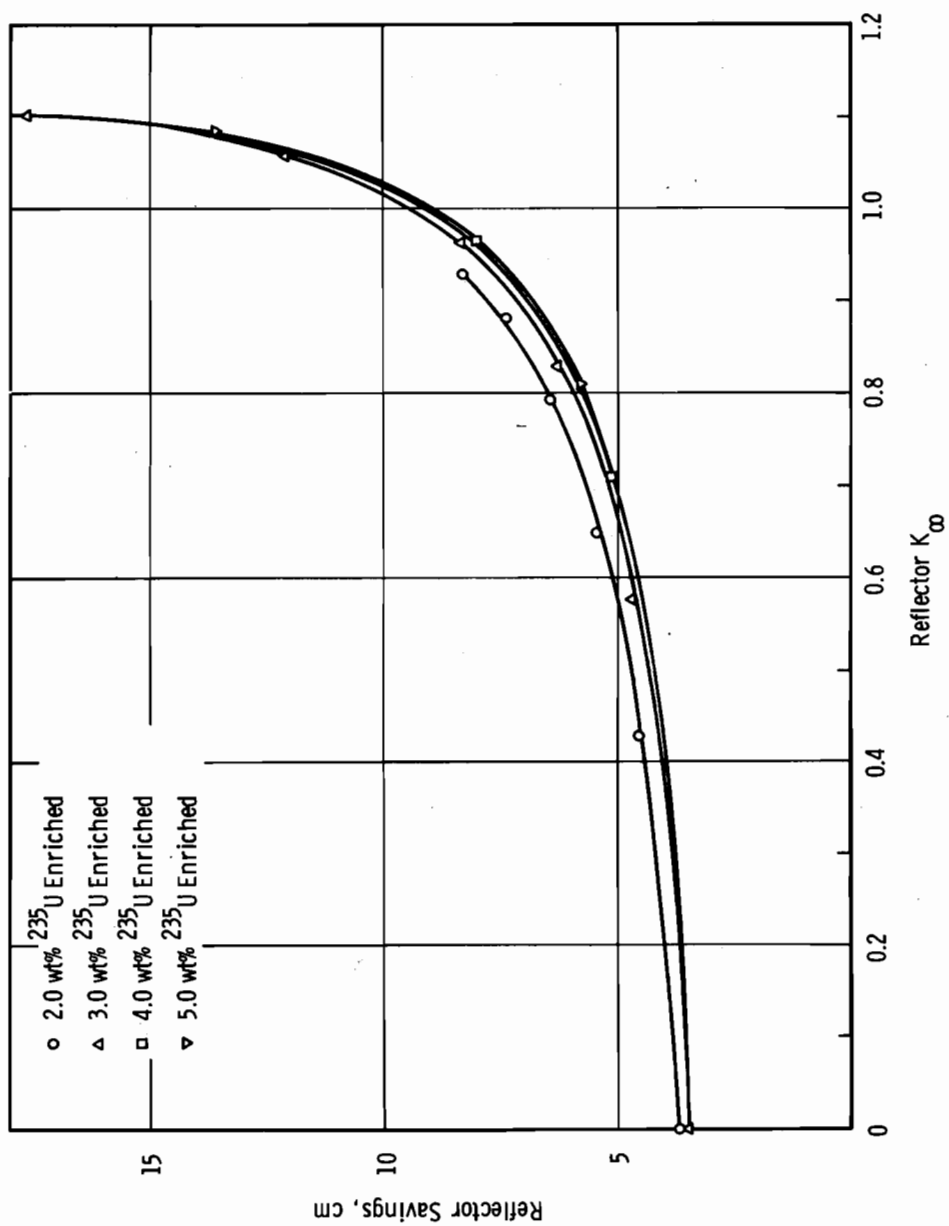


FIGURE 17
Reflector Savings as a Function of Reflector K_{∞} for a 50.80 cm Thick Uranyl Nitrate Reflector Surrounding a Spherical Uranium Lattice

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