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THOREX THORIUM NITRATE

PRODUCT SPECIFICATIONS

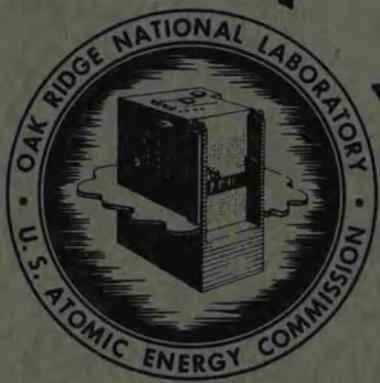
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THOREX THORIUM NITRATE PRODUCT SPECIFICATIONS

E. D. Arnold
R. P. Wischow

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0.0 ABSTRACT

Activity and ionic impurity specifications are presented for Thorex thorium nitrate products. Two sets of specifications are given, one set for direct handling during refabrication of production reactor thorium metal slugs and the second for refabrication of future power reactor thorium metal elements by semi-remote technics. Consideration was given to the health hazard problems associated with each process step between the Thorex process and final refabricated source material in order to arrive at these specifications.

1.0 SUMMARY

An analysis of the health hazards involved in the handling of thorium after it leaves the Thorex plant indicates that the most critical operations are the machining of the metal and those involving the handling of dry solids. The permissible activity in recycled thorium metal that is to be handled directly is taken as 1.4 times that of natural thorium.* This specification prevents overexposure of personnel to direct radiation and ensures that radiation from inhaled dust will be less than tolerance if a maximum of 150 μg of thorium per cubic meter of air is permitted in working areas.

For Thorex process thorium nitrate product that is to be converted to metallic thorium fuel elements, recommended levels of fission product activity, assuming a ruthenium decontamination factor of 10 in the oxalate precipitation, are:

* Natural thorium activity as used in this report means the activity of Th^{232} in complete equilibrium with its daughters.

	<u>For Direct Refabrication</u>	
	<u>Gamma*</u>	<u>Beta**</u>
	<u>(c/m/g Th)</u>	<u>(c/m/g Th)</u>
Ru	4×10^4	1.1×10^5
Zr + Nb	4.5×10^4	2.0×10^4
Pa	3×10^4	10^5
Rare earths	10^4	4.0×10^4
Total	1.3×10^5	2.7×10^5

Shipments of Thorex product meeting the direct refabrication specifications in 55-gal drums will read about half present ICC permissible levels. The direct refabrication specification for ruthenium activity ensures that the biological hazard of the waste from the oxalate precipitation step will be less than a factor of 2 over that of natural thorium and that ruthenium will not become a significant hazard unless during the remelt operation it concentrates in the vapor phase by a factor of more than 250.

Thorium product specifications for processing future power reactor fuel elements may exceed the above specifications by a factor of 4, since semi-remote refabrication of fuel elements will be necessary in this case. Thorium used in power reactors after several cycles will be at least 4 times as radioactive as natural thorium, since long irradiation periods which build up the Th^{228} content are mandatory in a power reactor economy.

2.0 INTRODUCTION

Production of U^{233} involves irradiation of thorium metal slugs

* Determined by scintillation counting through a 5-mg/cm^2 lead absorber.

** Based on 10% counting geometry.

or rods in a heterogeneous reactor, recovery of the U^{233} and thorium by the Thorex process after a suitable decay period, reconversion of the thorium to metal, and reirradiation in the reactors. After being separated from impurities in the Thorex plant, the thorium would be transferred, as a concentrated nitrate solution, to a metal refabrication plant. The thorium would there be precipitated as the oxalate, fired to the oxide, fluorinated to ThF_4 with HF, reduced to the metal in a bomb, remelted and cast into an ingot, and finally refabricated into slugs or rods for reirradiation. The Thorex process must adequately decontaminate the irradiated thorium to the point that all these operations can be carried out safely without expensive precautions.

This report summarizes our understanding of the radiation hazards involved in the anticipated handling of the thorium after it leaves the Thorex plant and recommends radiation and ionic specifications for the Thorex thorium product based on these hazards. The contribution of Th^{228} and Th^{234} with their decay chains is considered and permissible irradiation and decay times to meet specifications are given.

Recommended Thorex product specifications are also included for the special case of processing power reactor fuel elements containing metallic thorium. In these reactors long irradiation periods are mandatory for economy.

The authors gratefully acknowledge the valuable assistance given by H. K. Jackson and D. E. Ferguson in reviewing the original manuscript and making many valuable suggestions and by members of the ORNL Chemical Technology Division Thorex Committee, especially E. M. Shank, R. H. Rainey, and B. F. Bottenfield, in supplying data and suggestions.

3.0 THORIUM SPECIFICATIONS

Thorium specifications, whether for the salt or the metal, are based on the total external gamma radiation and on the alpha, beta, and gamma internal radiological hazards associated with all steps between chemical separation and final slug fabrication. There should be no significant biological hazard, either internal or external, for shipping Thorex thorium product, oxalate precipitation, calcination and hydrofluorination, reduction, remelting, or final slug fabrication. Handling and operating technics must be carefully controlled in order to limit dust contamination, waste stream activity levels, fume evolution from metal furnaces, and unnecessary handling time.

Thorex thorium product and metal activity specifications are set by the maximum gamma dosage that can be tolerated by personnel under present working conditions, the maximum permissible waste stream activity levels, and other health physics standards. To prevent overexposure of personnel machining and handling thorium slugs, a maximum total activity approximately 1.4 times that of natural thorium, i.e., ~ 1.4 times the activity of Th^{232} in complete equilibrium with its daughters, must be maintained. This corresponds to an activity level of 0.332 r/hr at the surface of a thorium slug. This ensures that the radiation from inhaled dust will be less than tolerance provided that not more than 150 μg of thorium per cubic meter of air is permitted in working areas.

Ionic specifications are based on the limit of impurities that allows the production of acceptable thorium metal for both slug fabrication and neutron economy.

3.1 Thorex Product Thorium Nitrate Specifications for Direct Refabrication of Thorium Metal Slugs

The Thorex thorium nitrate product, before the oxalate precipitation,

is allowed a factor of 10 greater ruthenium activity than final specifications permit because a ruthenium decontamination factor of at least 10 will be obtained in this step. Laboratory data¹ have shown that the oxalate precipitation may yield a ruthenium decontamination factor of 85. Since scaling up to plant size and recycle of chemicals may decrease the decontamination, a factor of 10 was used in setting the specifications. As shown in Table 1, the total beta activity contributed by the fission products and protactinium is 2.70×10^9 d/m/kg Th whereas the total average gamma activity is 1.25×10^5 c/m/g Th.* Figure 1 permits translation of the gamma activity specifications based on 300 days' total decay to any decay times less than 300 days. This figure can be utilized with any feed activity level in calculation of decontamination factors to meet these specifications. In the case of the UX₁ and UX₂ activity, only the UX₂ need be considered on a gamma activity basis.

3.2 Thorium Metal Specifications

The basis for the metal specifications is that the total radiation dose at the surface of a thorium slug (1.36 in. x 6 in.) shall not exceed 140% of the Th²³² equilibrium activity of 0.240 r/hr, i.e., 0.332 r/hr. Table 2 indicates the activity contribution of the major

* The scintillation counts per gamma disintegration were converted to counts per minute per gram of thorium by the conversion factors

Zr ⁹⁵	0.214	Ru ¹⁰³	0.182	Pa ²³³	0.042
Nb ⁹⁵	0.224	Ru ¹⁰⁶	0.022		

Since the Ru¹⁰³ and Ru¹⁰⁶ half-lives, fission yields, and efficiencies are appreciably different, the total efficiency or counting conversion factor is a function of irradiation time, reactor flux, and decay time. Therefore the assumption was made that at 300 days' decay, the Ru¹⁰³ beta activity would be 20% of the total ruthenium activity, the Ru¹⁰⁶ contributing the remaining 80%. The factors were supplied by S. A. Reynolds of the Analytical Chemistry Division; the scintillation counts were made through a 5-g/cm² lead absorber.

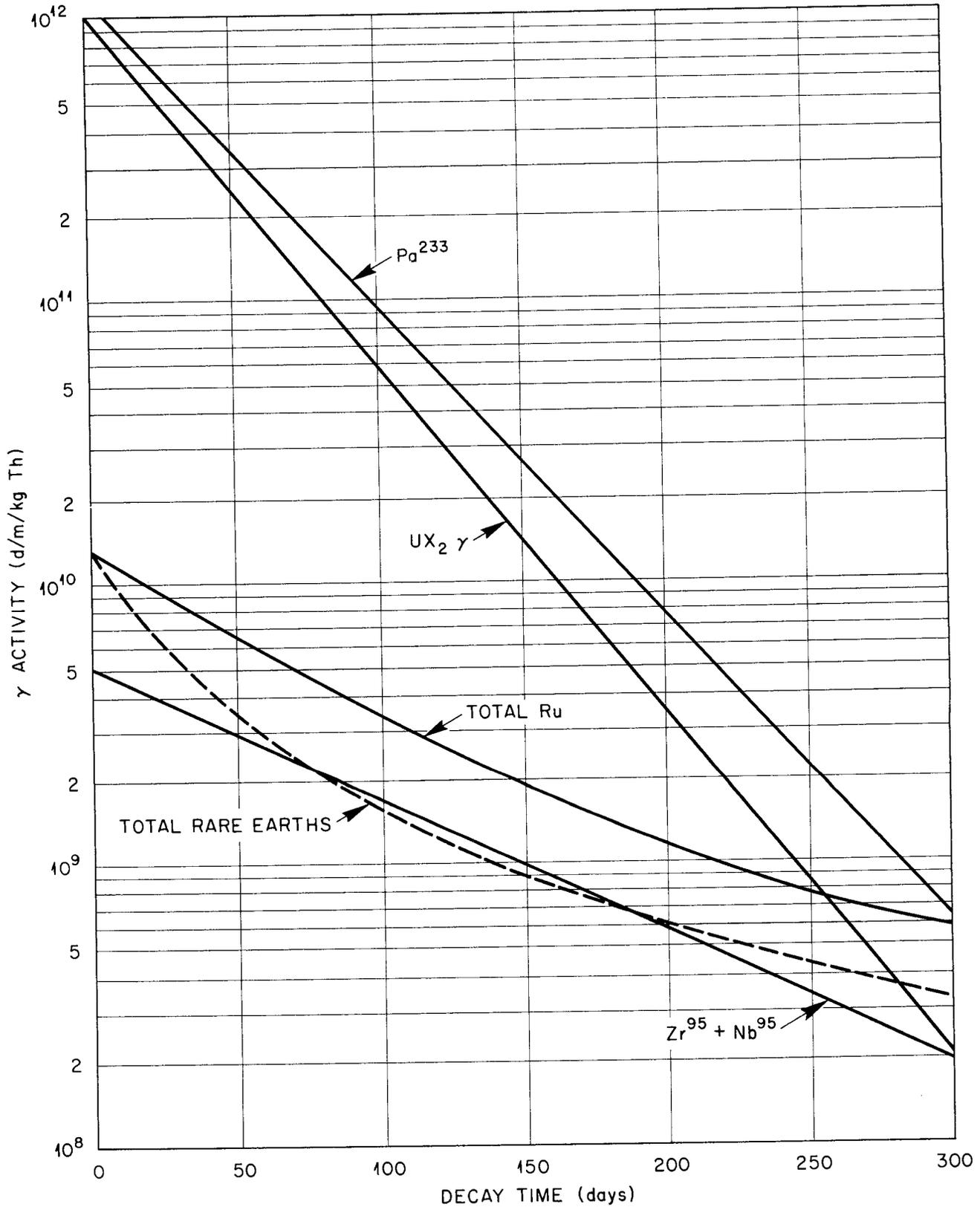


Fig. 1. Gamma Activity Associated with Thorex Process Thorium Nitrate Product as a Function of Total Decay Time, Assuming Thorex Product Specifications Are Met after 300 days' Decay.

Table 1. Thorex Thorium Nitrate Product Activity Specifications

Basis: ruthenium decontamination factor of 10 is obtained
in oxalate precipitation

Isotope	β Activity (d/m/kg Th)	Average γ Activity	
		(d/m/kg Th)	(c/m/g Th)
Ru ¹⁰³ + Ru ¹⁰⁶	1.1 x 10 ⁹	5.8 x 10 ⁸	4.0 x 10 ⁴
Zr ⁹⁵ + Nb ⁹⁵	2.0 x 10 ⁸	2.0 x 10 ⁸	4.5 x 10 ⁴
Pa ²³³	1.0 x 10 ⁹	7.0 x 10 ⁸	3.0 x 10 ⁴
Total rare earths	4.0 x 10 ⁸	3.3 x 10 ⁸	1.0 x 10 ⁴
Total	2.70 x 10 ⁹	1.81 x 10 ⁹	1.25 x 10 ⁵

Table 2. Thorium Metal Activity Specifications

Basis: total radiation dose at slug surface equal to 140% of
that of equilibrium Th²³²; metal slugs to be
directly refabricated

Isotope	β Activity (d/m/kg Th)	Average γ Activity		Radiation Dose at Slug Surface (r/hr)
		(d/m/kg Th)	(c/m/g Th)	
Ru ¹⁰³ + Ru ¹⁰⁶	1.1 x 10 ⁸	5.8 x 10 ⁷	4.0 x 10 ³	0.006
Zr ⁹⁵ + Nb ⁹⁵	2.0 x 10 ⁸	2.0 x 10 ⁸	4.5 x 10 ⁴	0.035
Pa ²³³	1.0 x 10 ⁹	7.0 x 10 ⁸	3.0 x 10 ⁴	0.015
Total rare earths	4.0 x 10 ⁸	3.3 x 10 ⁸	1.0 x 10 ⁴	0.005
Total	1.71 x 10 ⁹	1.29 x 10 ⁹	8.9 x 10 ⁴	0.062

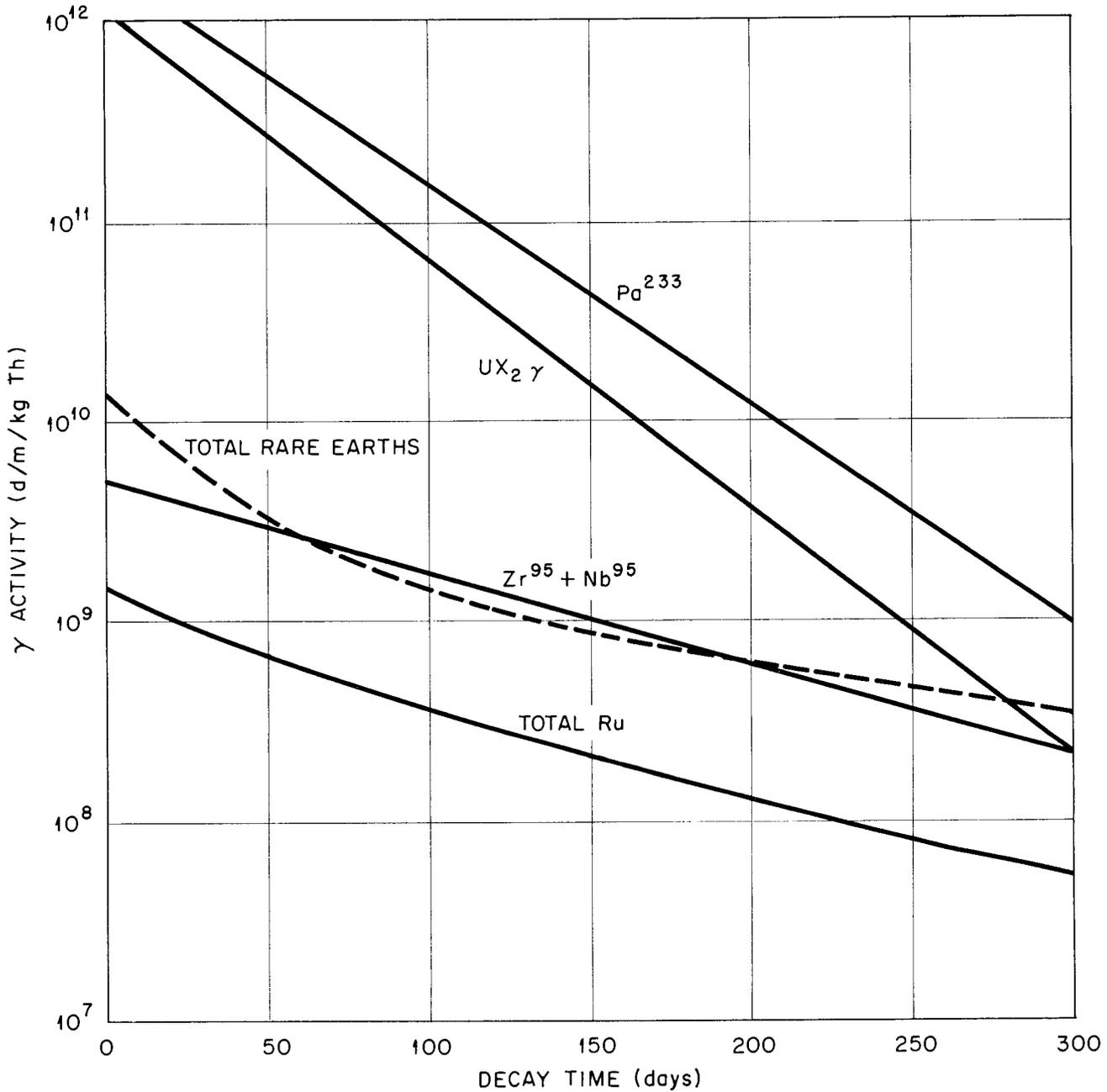


Fig. 2. Gamma Activity Associated with Thorium Metal as a Function of Total Decay Time, Assuming Metal Specifications Are Met with a Ruthenium Decontamination Factor of 10 in Oxalate Precipitation and a Total Decay Time of 300 days.

fission product nuclides and Pa²³³ after a 300-day total decay time; Fig. 2 illustrates the associated gamma activity specifications at any decay time less than 300 days. Table 3 lists the residual and fertile daughter activities in the thorium product or metal which meets the above specifications after infinite recycle. For several combinations of irradiation, pre-Thorex decay, post-Thorex decay, and refabrication times the thorium metal activity due to the Th²²⁸ chain after an infinite number of cycles is calculated to be three times that after one cycle. The minimum total cycle time which will establish this condition would be 400 days between the nth loading of a reactor and the "n + 1"th loading. A nomograph (Fig. 18) which may be used to determine these combinations is given in the appendix. Particular note must be taken of the activity contribution of the Th²²⁸ decay chain after one cycle as compared to the activity after an infinite recycle. That is, if only one cycle was used, the Th²²⁸ chain activity would be a factor of 3 less than for the infinite recycle system; thus, on a one-cycle basis, the difference of 0.106 r/hr dosage at a slug surface could be applied to an increase in the fission product and Pa²³³ activity levels with the resultant permissible decrease in the required decontamination factors.

3.3 Maximum Permissible Irradiation Levels and Decay Times

The irradiation level and/or the decay time² must be fixed in order to have thorium metal which meets handling specifications after the fission product, Pa²³³, and UX₁ + UX₂ limits are set. Figure 3 indicates the permissible irradiation level for infinitely recycled thorium, as a function of thermal flux and decay time, which meets total activity specifications of 1.4 times that of Th²³² in equilibrium with its natural decay daughters. This level is the maximum that may be directly handled under present operating conditions. The curves are based on an apparent (n, 2n) cross-section of 3 mb for Th²³². The

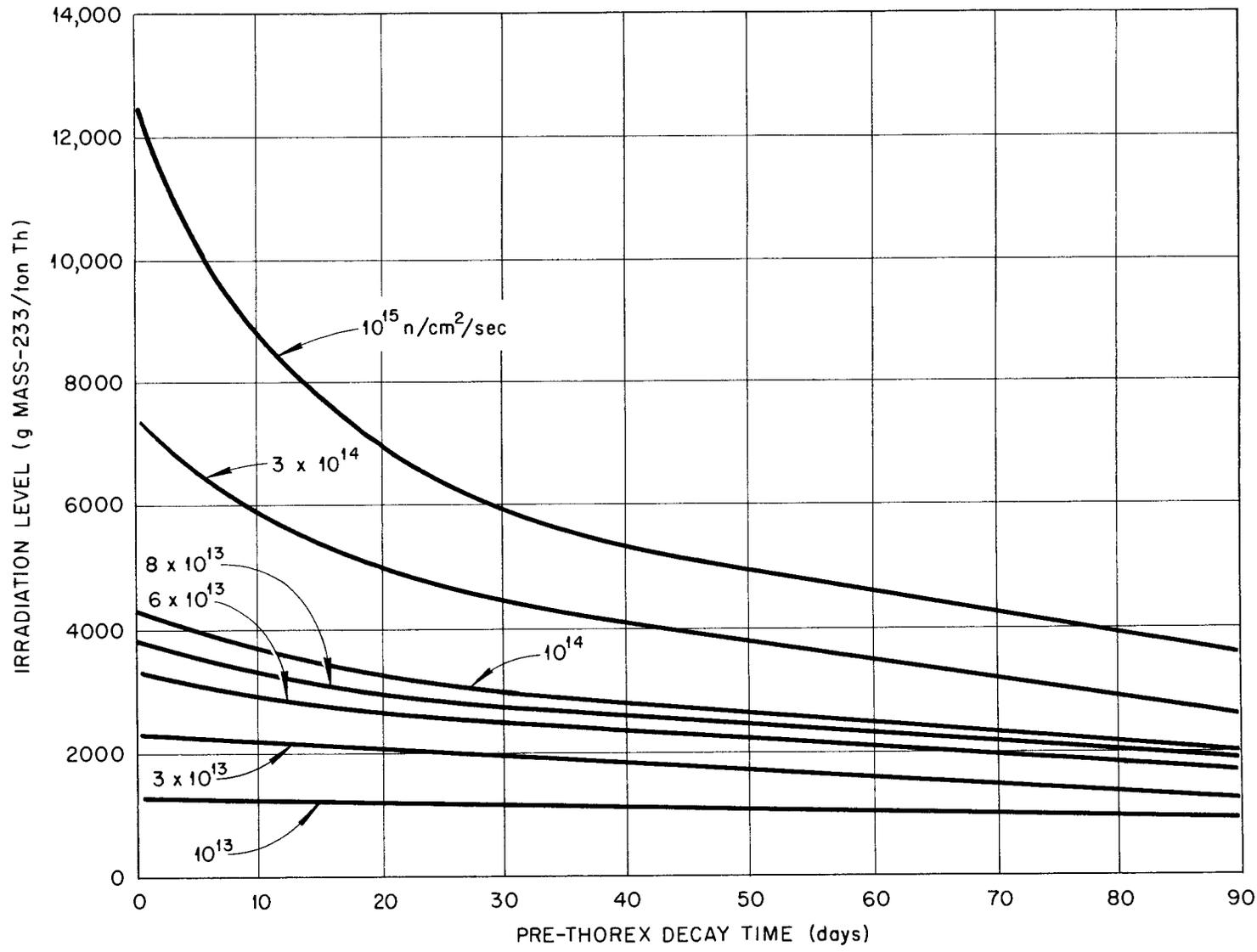


Fig. 3. Maximum Permissible Irradiation Level as a Function of Flux and Total Decay Time. Based on specification that after infinite recycle the maximum activity of thorium that is to be reused in a reactor is 1.4 times that of natural thorium; $\text{Th}^{232} \sigma_{n,2n} = 3 \text{ mb}$.

permissible g/t level* for any other cross-section may be found by multiplying the plotted value by $\sqrt{3/\sigma_{02}(n, 2n)}$.

Table 3. Residual and Fertile Daughter Activities in Thorium Metal Meeting Specifications

Basis: total irradiation dosage at slug surface equivalent to 140% of that of equilibrium Th^{232} after infinite recycle; metal specifications for direct slug refabrication considered

Isotope	β Activity (d/m/kg Th)	Average γ Activity		Radiation Dose at Slug Surface (r/hr)
		(d/m/kg Th)	(c/m/g Th)	
$\text{UX}_1 + \text{UX}_2$	1.0×10^9	2.0×10^8 (UX_2)	4.5×10^4	0.030
Th^{228} chain After ∞ recycle	5.8×10^8	3.5×10^8	5.8×10^4	0.160
(After 1 cycle)	(2.0×10^8)	(1.2×10^8)	(2.0×10^4)	(0.054)
1/3 of activity of Th^{232}	1.7×10^8	1.8×10^8	3.0×10^4	0.080
Total after infinite recycle	1.75×10^9	7.3×10^8	1.33×10^5	0.270

3.4 Decontamination Factors Required to Meet Metal Specifications

In order to produce metal meeting the fission product and Pa^{233} specifications from 2000-g/t material, it is necessary to obtain the decontamination factors shown in Table 4. The individual fission product decontamination factors required vary as a function of the g/t levels (Fig. 4). The required decontamination values given in Table 4 are extended by Figs. 4 and 5 to cover a 1000- to 4000-g/t irradiation range and 0 to 300 days' decay time.

* g/t level = grams of mass-233 per ton of thorium.

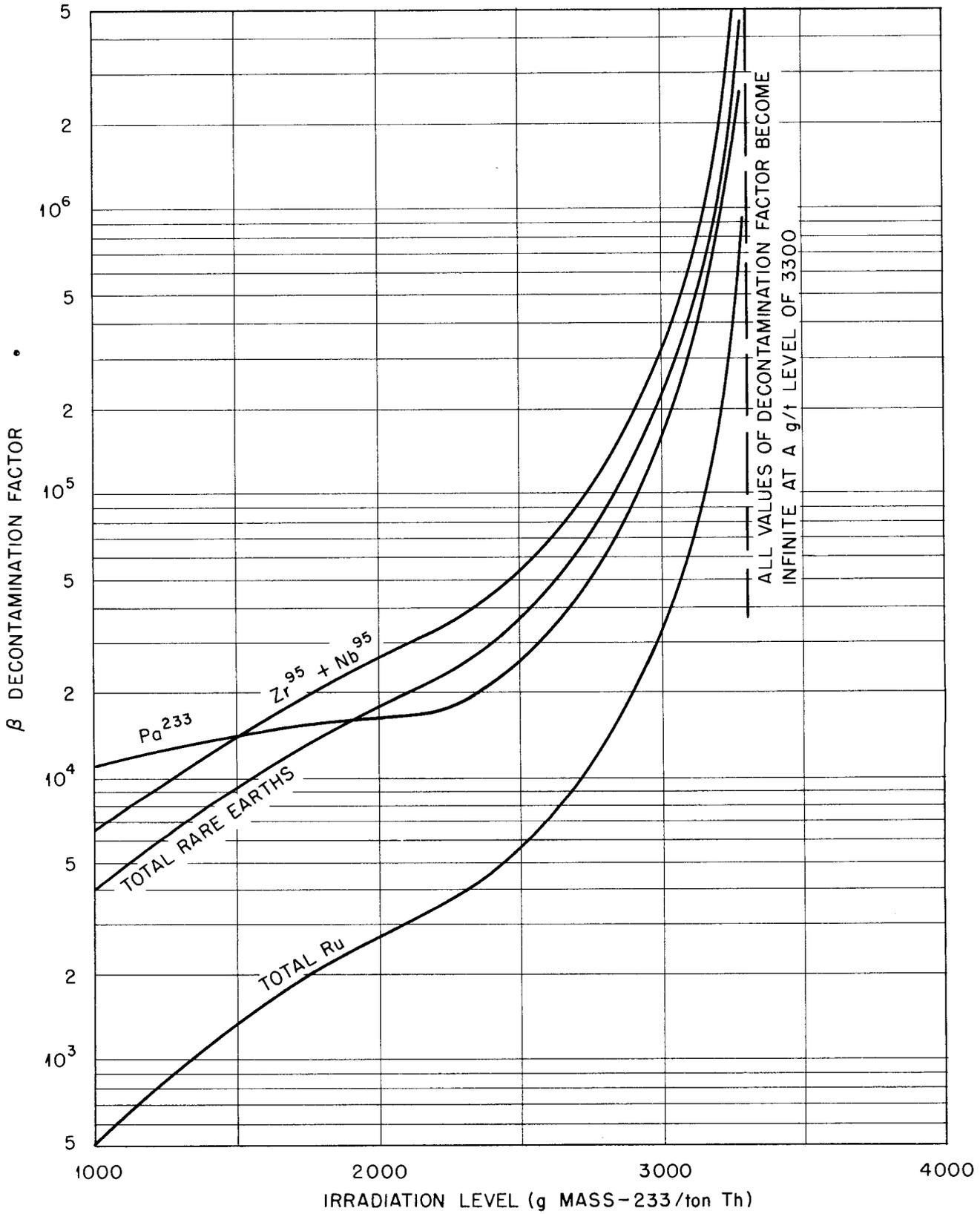


Fig. 4. Beta Decontamination Factor Required as a Function of g/t Level. Flux, 3×10^{13} n/cm²/sec; total decay time, 300 days; Th²³² $\sigma_{n,2n} = 3$ mb.

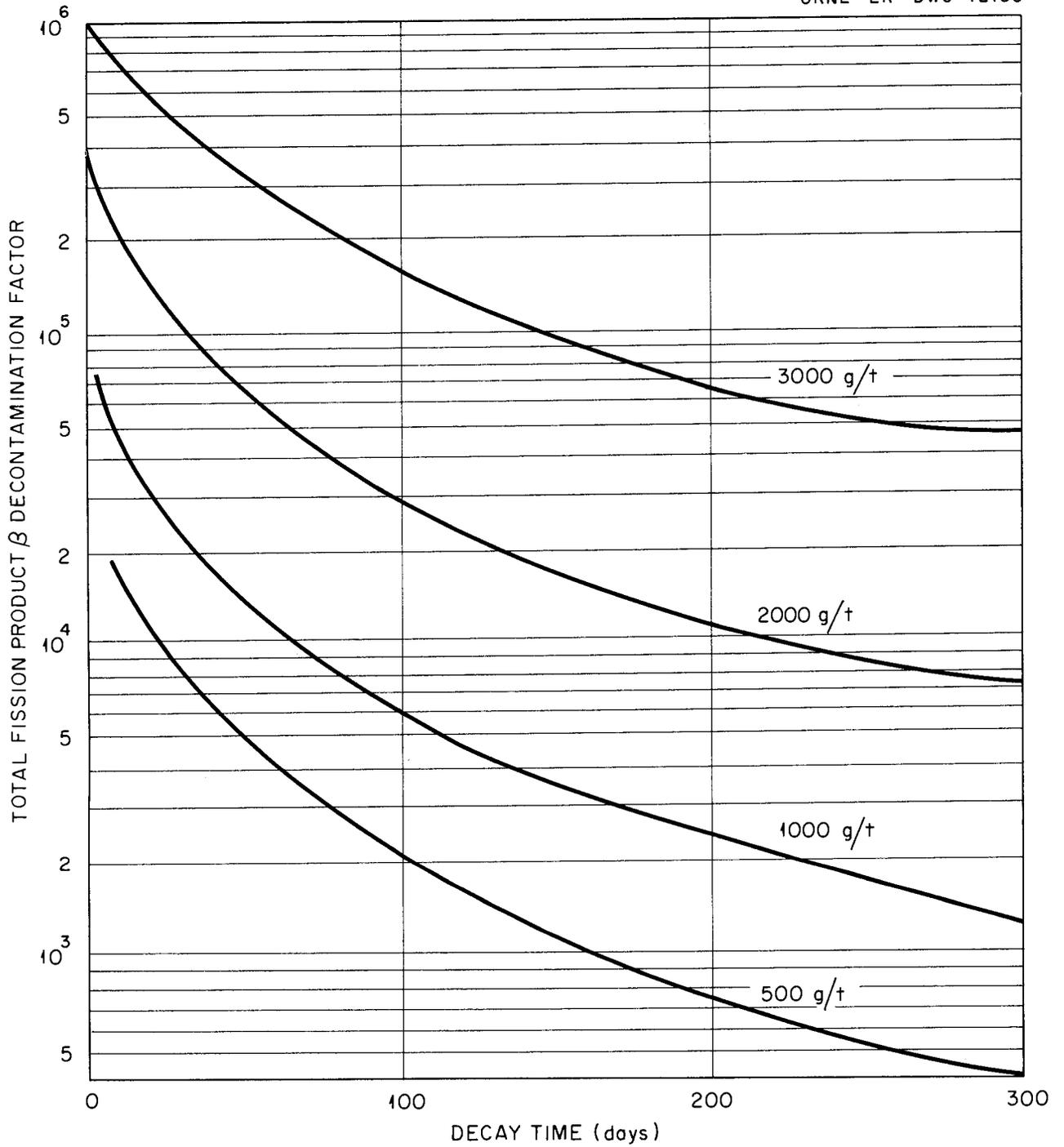


Fig. 5. Total Fission Product β Decontamination Factor Required to Meet Metal Specifications as a Function of Total Decay Time. Flux: 3×10^{13} n/cm²/sec; Th²³² $\sigma_{n,2n} = 3$ mb.

Table 4. Nuclide Decontamination Factors Required to Meet Metal Specifications from 2000-g/t Material

Basis: total radiation at slug surface equivalent to 140% of equilibrium Th²³² activity; irradiation at 3×10^{13} n/cm²/sec; decay time set by Th²²⁸ chain

Nuclide	Decontamination Factor
Ru ¹⁰³ + Ru ¹⁰⁶	2.7×10^3
Zr ⁹⁵	2.6×10^4
Nb ⁹⁵	2.6×10^4
Pa ²³³	1.4×10^4
Total rare earths	1.7×10^4

Thorium irradiated to 3300-4000 g/t at a thermal flux of 3×10^{13} n/cm²/sec cannot be decontaminated to meet specifications since infinite recycle of such thorium will build up Th²²⁸ chain activities that are much greater than the permissible 1.4 times that of natural thorium. Higher fluxes would be necessary to allow the production of 3300-4000 g/t material. Thus 4000-g/t material can be produced only at fluxes in excess of 8×10^{13} n/cm²/sec in order to meet the Th²²⁸ chain activity specifications.

3.5 Thorex Thorium Nitrate Product Ionic Specifications

The ionic impurities (Table 5) allowable in the Thorex thorium product are calculated from considerations of neutron economy, uranium losses, and probable corrosion product concentrations, i.e., chromium, iron, and nickel. The loss of neutrons (< 0.3% of total U²³³ production) due to rare earth capture is not critical in this case because, if the activity specifications for rare earths are met, the ionic concentrations will be much less than the ionic specifications.

Table 5. Thorex Process Thorium Nitrate Product Ionic Specifications³

Impurity	Allowable Conc. (g/g Th)	Impurity	Allowable Conc. (g/g Th)
U	10^{-6}	Cd	10^{-7}
B	5×10^{-8}	Dy	10^{-7}
Al	10^{-4}	Ce, La, Nd, Pr, Y	5×10^{-7} each
Fe	3×10^{-4}	Other impurities	10^{-5} each
Sm, Gd, or Eu	5×10^{-8} each		

4.0 JUSTIFICATION

In none of the steps of the complete thorium refabrication operation may the biological dose rate be substantially increased over that now encountered with unirradiated thorium as either an internal or external radiological hazard. The thorium nitrate product must meet ICC specifications in order for interstate carriers to be used in shipping. Slightly higher activity levels of a shipment are permissible if the carrier is owned and operated by the processing plant. The activity of the oxalate precipitation waste stream must be compatible with existing disposal procedures. The activity of the oxalate precipitation product must be within the limits imposed by the dust problems associated with the calcination and hydrofluorination steps. The total gamma activity of the reduced metal as well as activity levels associated with dusting impose restrictions on the procedures used in the reduction step. The level of volatile radioisotopes must be minimized in order to reduce any hazard associated with the remelting step. The gamma activity of the metal must be minimized in order to maintain permissible radiation levels in the machining room.

4.1 Considerations for Shipping Thorex Thorium Nitrate Product

Specifications based on ICC shipping regulations are that the activity at the surface of a 55-gal drum must be less than 200 mr/hr and that the activity 1 meter from the drum must be less than 10 mr/hr. Thorium nitrate solutions containing 400 g of thorium per liter, which have an activity 5.0 times that of equilibrium Th^{232} (Th^{232} + equilibrium daughters), will almost meet the surface activity specification but will not meet that at 1 meter. A 55-gal drum of this solution will read 190-250 mr/hr at the surface and 12-15 mr/hr at 1 meter from the drum. Processing or refabrication plants that operate their own trucks could ship this material. Thorium with an activity three times that of natural thorium would meet ICC shipping specifications. A solution of thorium which has an activity only 1.4 times that of equilibrium Th^{232} would give readings of 70 mr/hr at the surface and ~ 4 mr/hr at 1 meter. Thus it may be possible to ship solutions or slurries containing as much as 1000 g of thorium per liter which meet these specifications.

4.2 Contribution of Ruthenium to the Biological Hazard of the Oxalate Precipitation Waste

The permissible ruthenium activity of the thorium nitrate before oxalate precipitation was set at 1.1×10^9 β d/m/kg Th, or 10 times that permitted in the metallic thorium product. In laboratory experiments¹ more than 95% of this ruthenium, along with some zirconium and niobium, rare earths, and Pa^{233} , remained in the supernatant from the oxalate precipitation step. When unirradiated thorium is processed, the controlling activity in the oxalate precipitation waste is 6.7-year Ra^{228} , which is in equilibrium with Th^{232} . Other removable activities are of very little importance since their half-lives are short. The average activity of unirradiated thorium now being processed is approximately one-third that of Th^{232} in

equilibrium with its daughters. This condition indicates an activity of removable radioisotopes [Ra^{228} , Th^{228} which will build up in the waste stream, Ra^{224} , Rn^{220} , Po^{216} , Bi^{212} (34%), and Po^{212} (66%)] of $\sim 2.7 \times 10^8 \alpha$ d/m/kg Th two years after precipitation. The additional alpha activity or the biological equivalent of the beta activity of all radioisotopes other than the residual Th^{232} that can be added to the present waste streams must be less than $2.7 \times 10^8 \alpha$ (or equivalent β) d/m/kg Th. Since the alpha particle has a relative biological effectiveness 5 times that of a beta particle, the maximum permissible activity of beta emitters that can be added to the present waste streams is $1.3 \times 10^9 \beta$ d/m/kg Th to maintain the total biological hazard at less than 2 times the present alpha hazard.

4.3 Dust Hazards from Calcination and Hydrofluorination

The calcination and hydrofluorination steps create dust hazards which limit the dust contamination permissible. The total air contamination by Th^{232} must be kept below 3×10^{-11} curie per cubic meter of air* if there is no other activity source, external or internal, present.^{4,5} The external radiation level in the calcination and hydrofluorination areas should be less than that in the machining area. If it is assumed that the hazard is half internal and half external and that the external radiation level in these areas should be half that in the machining area, the maximum permissible Th^{232} activity per cubic meter of air is 1.5×10^{-11} curie (the corresponding activity of Th^{232} in equilibrium with all its daughters is $1.5 \times 10^{-10} \alpha + \beta$ curie per cubic meter of air), which is 150 μg of Th^{232} with a total $\alpha + \beta$ activity of 2.0×10^9 d/m/kg Th. The air contamination must be reduced

* This value may be lowered in the future by as much as a factor of 15, but no official value other than this has been published and the new value is still in dispute by several health physicists. (Personal communication from K. Z. Morgan, ORNL Health Physics Division.)

proportionally if the thorium product has a total activity greater than 2.0×10^9 d/m/kg or if the average external radiation in the operating area is greater than 3.7 mr/hr. Thus with thorium that has an activity 5.0 times that of equilibrium Th^{232} , only 30 μg per cubic meter of air could be tolerated.

4.4 Limitations Imposed by the Reduction Step

The reduction step will have the same dust problem as the calcination and hydrofluorination steps. For safe operation during the reduction step, the gamma activity of the reduced metal must be the same as that of the final metal slug.

4.5 Limitations Imposed by the Remelting Step

The limitations imposed by the remelting step require that the total gamma activity of the thorium be no greater than that of the final slug. However, the limiting problem may be ruthenium boil-off during the remelting. The allowable Ru^{106} activity (when by itself) in air is 2×10^{-8} curie per cubic meter of air. If a ruthenium air contamination of 10% of the maximum permissible ruthenium activity is allowed to keep the total activity within limits, then a limit of 4.4×10^3 d/m per cubic meter of air must be set for any ruthenium boiled off or carried over.^{4,5} This activity is 260 times that of the ruthenium associated with a thorium dust concentration of 150 μg per cubic meter of air. Unless there is a large ruthenium concentration in the vapor phase during remelting, the total dust problem would be much more important than ruthenium boiled off during the remelting step.

4.6 Considerations for Handling Thorium Slugs and Other Metal Shapes

The final metal specifications were based on the handling operation involved in the fabrication of thorium metal slugs. The limit of activity at the surface of a slug was set at 1.4 times that of equilibrium Th^{232} . The present machining operations involve metal which has an activity of approximately 2/3 that of equilibrium Th^{232} , and the personnel involved in these operations have been exposed to a total radiation of 2-6 mr/hr.* The integrated average of 3.6 mr/hr may be used as the radiation to be expected from thorium which has an activity level of 2/3 that of equilibrium Th^{232} provided normal operating and metal handling practices are used. Increasing thorium activities to 1.4 times equilibrium Th^{232} would increase the average external radiation level to 7.4 mr/hr or just slightly less than the maximum permissible radiation level. An external radiation level of 7.4 mr/hr will allow only 12 μg of Th^{232} per cubic meter of air in the machining room and thus maintain a 300 mr/week external plus internal total radiation exposure. Unless the thorium dust level can be reduced from 150 μg per cubic meter of air, which is now maintained and which should be acceptable in other areas, to 12 μg per cubic meter of air in the machining area, a slight modification in operating distances, working periods, or methods is necessary.

5.0 RELAXATION OF THOREX SPECIFICATIONS

These Thorex specifications are based on activity contribution from fission products of only 19% of the total activity. Modifications

* Letter, T. C. Runion (FMPC) to H. K. Jackson (ORNL) which states, in part: "Presently, the machinists are exposed to a radiation level of 2-6 mr/hr when working with virgin thorium which is at about 2/3 of its equilibrium with daughter decay products."

in present metal refabrication technics may allow a large increase in the allowable activity in thorium metal. This increase in allowable activity could then allow greater fission product and/or Th²²⁸ contamination of the product. For example: An increase of 50% in the allowable activity could allow a 2.5-fold increase in fission product contamination, a 20% increase in irradiation level, or some combination of the two. The exact magnitude of the change in any operation will depend on the overall economics of reactor irradiation, chemical processing, and metal refabrication.

The thorium nitrate specification was set low in order that most of the ruthenium would be removed in the Thorex plant, where the waste disposal system is equipped to handle this fission product, which is not the case in the metal refabrication plant. More ruthenium could be permitted in the thorium nitrate product if the oxalate precipitation was carried out in the Thorex plant, where oxalate waste, containing large amounts of ruthenium, could be handled in existing conventional facilities.

6.0 REFERENCES

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3. L. L. Marsh, Jr., J. R. Keeler, "The Technology of Thorium," BMI-76 (July 18, 1951); C. A. Burkart et al., "Final Report on Purification of Thorium Nitrate by Solvent Extraction with Tributyl Phosphate. II. Mixer-Settler Pilot Plant Investigations," BMI-263 (July 31, 1952).
4. E. D. Arnold, A. T. Gresky, "Relative Biological Hazards of Radiations Expected in Homogeneous Reactors TBR and HPR," ORNL-1982 (Nov. 15, 1955).
5. "Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Permissible Concentrations in Air and Water," Natl. Bur. Standards Handbook 52, U. S. Gov't Printing Office, Washington, 1953 (Report of the Subcommittee on Permissible Dose, of the National Committee on Radiation Protection).
6. A. T. Gresky, E. D. Arnold, "Products Produced in Batch Neutron Irradiation of Thorium," ORNL-1818 (Dec. 5, 1955).
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7.0 APPENDIX

7.1 Power Reactor Specifications

Fission product specifications may be relaxed somewhat for power reactor thorium blankets since the total activity of the thorium nitrate produced in processing them will necessarily be 4-5 times that of equilibrium Th^{232} . Fission product limits for metallic thorium may be increased approximately fourfold, with fission products contributing only 17% of the total activity (Table 6). The bulk of the total activity associated with infinitely recycled thorium metal will be due to the Th^{228} chain formed during irradiation and the processing decay period (Table 7). This chain will have a beta activity after one cycle of 1.1×10^9 β d/m/kg Th; after infinite recycle the beta activity will be 3.3×10^9 d/m/kg Th, or approximately 74% of the total activity.

The future of power reactors that use metallic thorium blankets depends on either a higher permissible metal activity or a 5-7 year holdup of recovered thorium. In order for thorium to be recycled through power reactors economically, irradiation levels of 3000-4000 g/t or higher must be used and the refabrication process must be designed to handle thorium which is approximately 5 times as radioactive as equilibrium Th^{232} .² Dust levels must be reduced to 30-40 μg per cubic meter of air (instead of 150 μg of unirradiated thorium per cubic meter of air now permitted) and direct handling of thorium metal reduced to 2-5 hr/week. For the remainder of the operating week an average distance of 8 in. must be maintained between the work and the hands and 18 in. between the work and the body. Thorium metal with an activity 10-20 times that of equilibrium Th^{232} would have to be handled entirely by semi-remote technics.*

* In semi-remote handling, tongs or manipulators are used, which place the hands 1 ft or more and the head and body more than 2 ft from the work.

Table 6. Future Power Reactor Thorium Metal Specifications

Basis: total radiation at slug surface equal to 5.0 times that of equilibrium Th²³²

Isotope	β Activity (d/m/kg Th)	Average γ Activity		Radiation Dose at Slug Surface (r/hr)
		(d/m/kg Th)	(c/m/g Th)	
Ru ¹⁰³ + Ru ¹⁰⁶ a	4.5 x 10 ⁸	2.15 x 10 ⁸	1.5 x 10 ⁴	0.025 ^b
Zr ⁹⁵ + Nb ⁹⁵	6.0 x 10 ⁸	6.0 x 10 ⁸	1.3 x 10 ⁵	0.100
Pa ²³³	2.9 x 10 ⁹	2.0 x 10 ⁹	8.5 x 10 ⁴	0.040
Total rare earths	1.6 x 10 ⁹	1.3 x 10 ⁹	4.0 x 10 ⁴	0.040

- a. Ru¹⁰³ activity will be much higher after processing, but a 300-day total decay time would reduce it to less than 20% of the Ru¹⁰⁶ activity.
 b. Radiation due mainly to Rh¹⁰⁶ γ + Ru¹⁰³ γ with Ru¹⁰³ activity = 20% of Ru¹⁰⁶ activity.

Table 7. Residual and Fertile Daughter Activities in Thorium Metal Meeting Future Power Reactor Metal Specifications

Basis: total radiation at slug surface equal to 5.0 times that of equilibrium Th²³² after infinite recycle

Isotope	β Activity (d/m/kg Th)	Average γ Activity		Radiation Dose at Slug Surface (r/hr)
		(d/m/kg Th)	(c/m/g Th)	
UX ₁ + UX ₂ Th ²²⁸ chain	1.0 x 10 ⁹	2.0 x 10 ⁸	4.5 x 10 ⁴	0.030
After ∞ recycle	3.3 x 10 ⁹	2.0 x 10 ⁹	3.4 x 10 ⁵	0.912
(After 1 cycle)	(1.1 x 10 ⁹)	(6.6 x 10 ⁸)	(1.1 x 10 ⁵)	(0.304)
1/3 of activity of Th ²³²	1.7 x 10 ⁸	1.8 x 10 ⁸	3.0 x 10 ⁴	0.080
Total after Infi- nite Recycle	4.47 x 10 ⁹	2.38 x 10 ⁹	4.15 x 10 ⁵	1.022

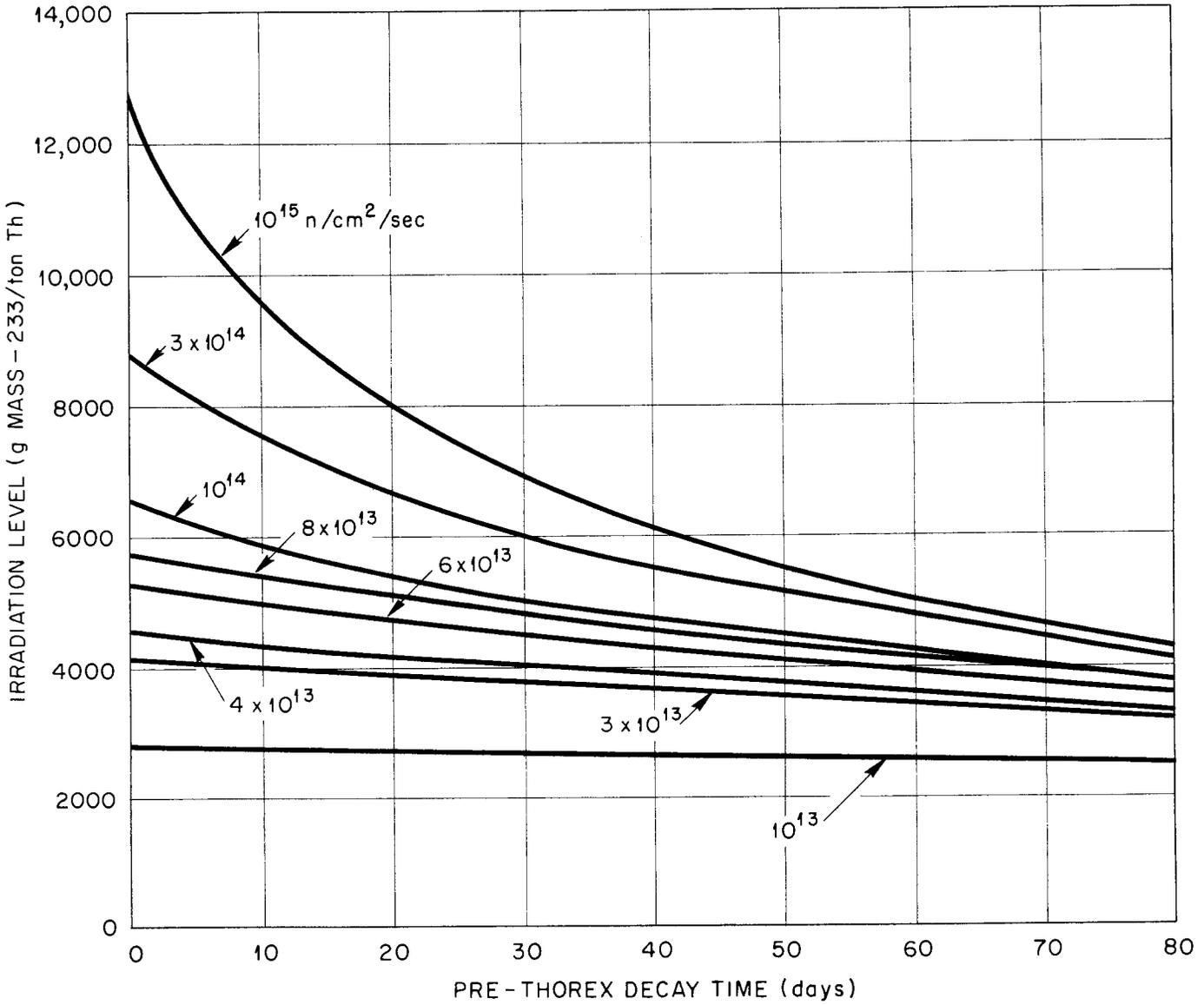


Fig. 6. Maximum Permissible Irradiation Level as a Function of Flux and Total Decay Time. Based on the specification that, after infinite recycle, the maximum activity of thorium that is to be reused in a power reactor shall not be more than 5 times that of Th^{232} at equilibrium. $\text{Th}^{232} \sigma_{\gamma,2.7} = 3\text{mb}$.

Figure 6 indicates the irradiation level, as a function of thermal flux and decay time, of thorium which meets an activity specification 5.0 times that of Th^{232} in equilibrium with its natural decay daughters. The curves are based on an apparent $(n, 2n)$ cross section of 3 mb. The permissible g/t level for any other cross section may be found by multiplying the plotted value by $\sqrt{3/\sigma_{02}(n, 2n)}$.

7.2 Activity Graphs

The following graphs are used for calculation of decontamination factors and activities of individual isotopes or chains (many of the data were obtained from references 6 and 7). Figures 7-10 are plots of important fission product and Pa^{233} beta activities as functions of irradiation level and decay time for a constant flux of 3×10^{13} n/cm²/sec. Figure 11 is a plot of nuclide beta activity specifications as a function of decay time. Figure 12 is a plot of total rare earth beta activity as a function of irradiation level and decay time. Figure 13 is a plot of Th^{228} chain activities in thorium metal as a function of cooling time before Thorex. Figure 14 is a plot of $\text{UX}_1 + \text{UX}_2$ activities⁶ in thorium metal as a function of irradiation level and decay time. Figure 15 is a plot indicating the ratio of the activity level to one-third the activity of Th^{232} as a function of decay time before processing. Figure 16 is a plot of important fission product activities at time of discharge as a function of irradiation levels. Figure 17 is a plot of Ba^{140} - La^{140} activity as a function of irradiation level and decay time.

7.3 Nomenclature and Definitions

g/t = grams of $\text{U}^{233} + \text{Pa}^{233}$ per ton of thorium

ϕ = thermal flux, neutrons per square centimeter per second

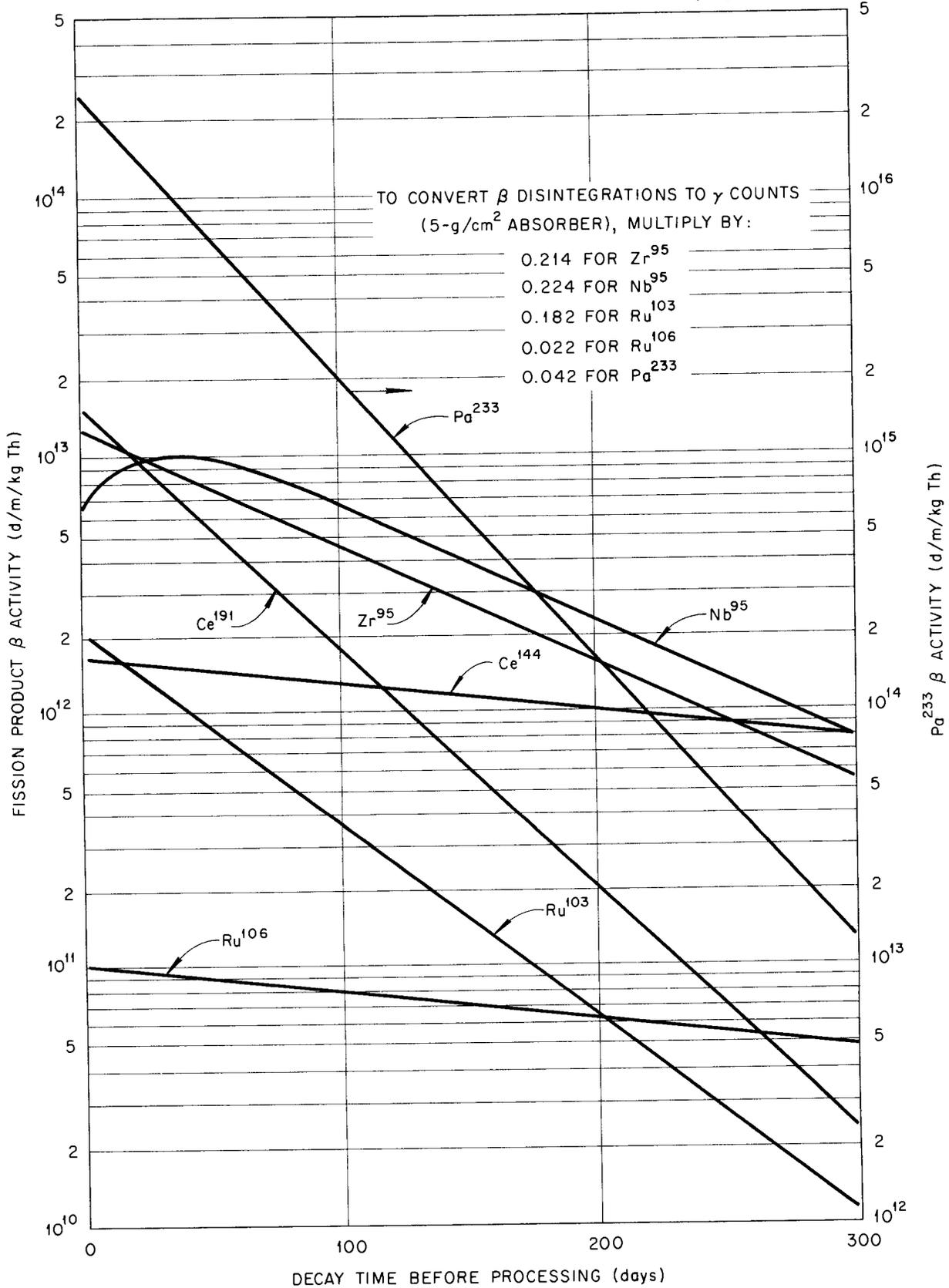


Fig. 7. Nuclide β Activity as a Function of Decay Time before Processing. Irradiation level, 1000 g of mass-233 per ton of thorium; flux, 3×10^{13} n/cm²/sec.

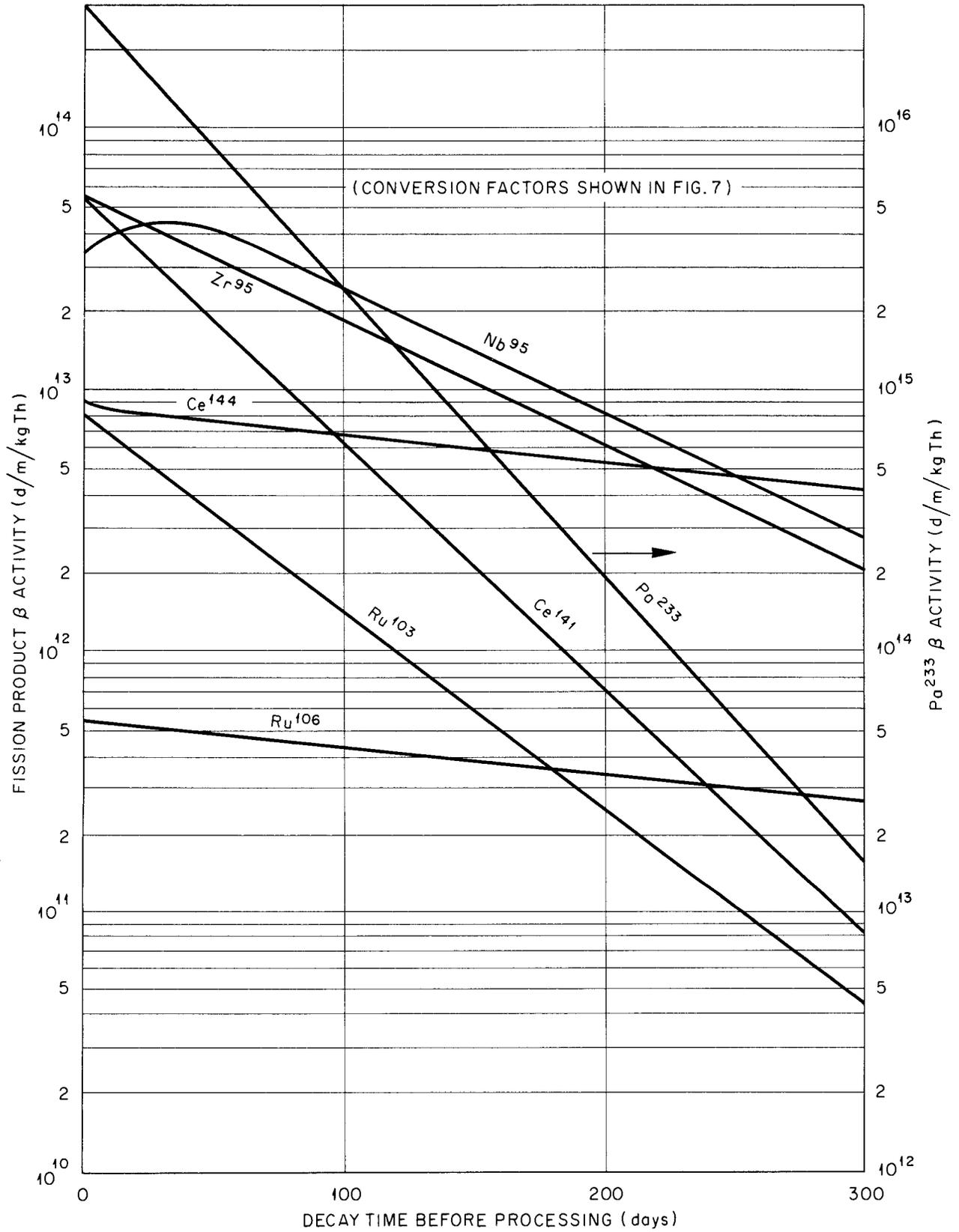


Fig. 8. Nuclide β Activity as a Function of Decay Time before Processing. Irradiation Level, 2000 g of mass-233 per ton of thorium; flux, 3×10^{13} n/cm²/sec.

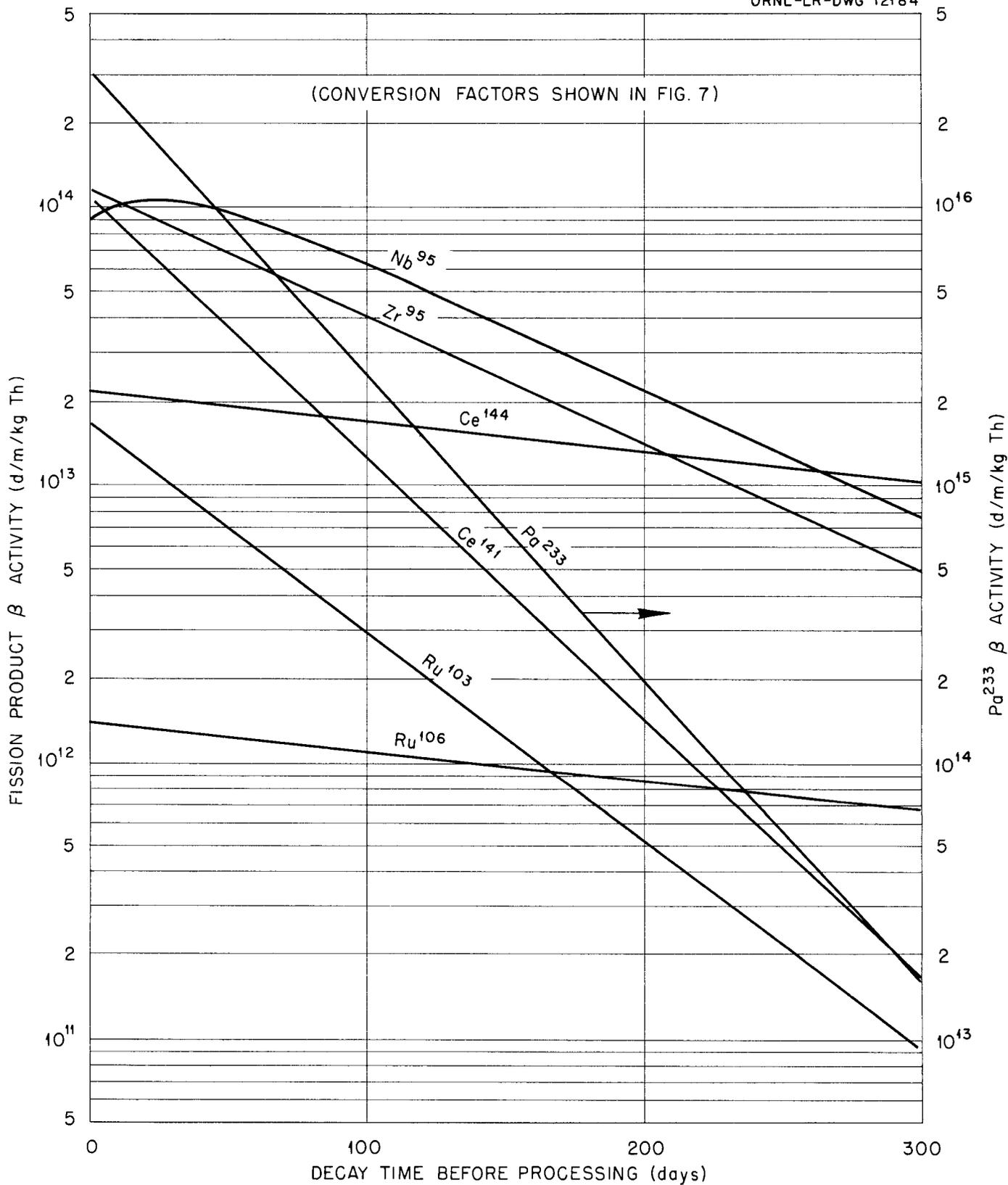


Fig. 9. Nuclide β Activity as a Function of Decay Time before Processing. Irradiation level, 3000 g of mass-233 per ton of thorium; flux, 3×10^{13} n/cm²/sec.

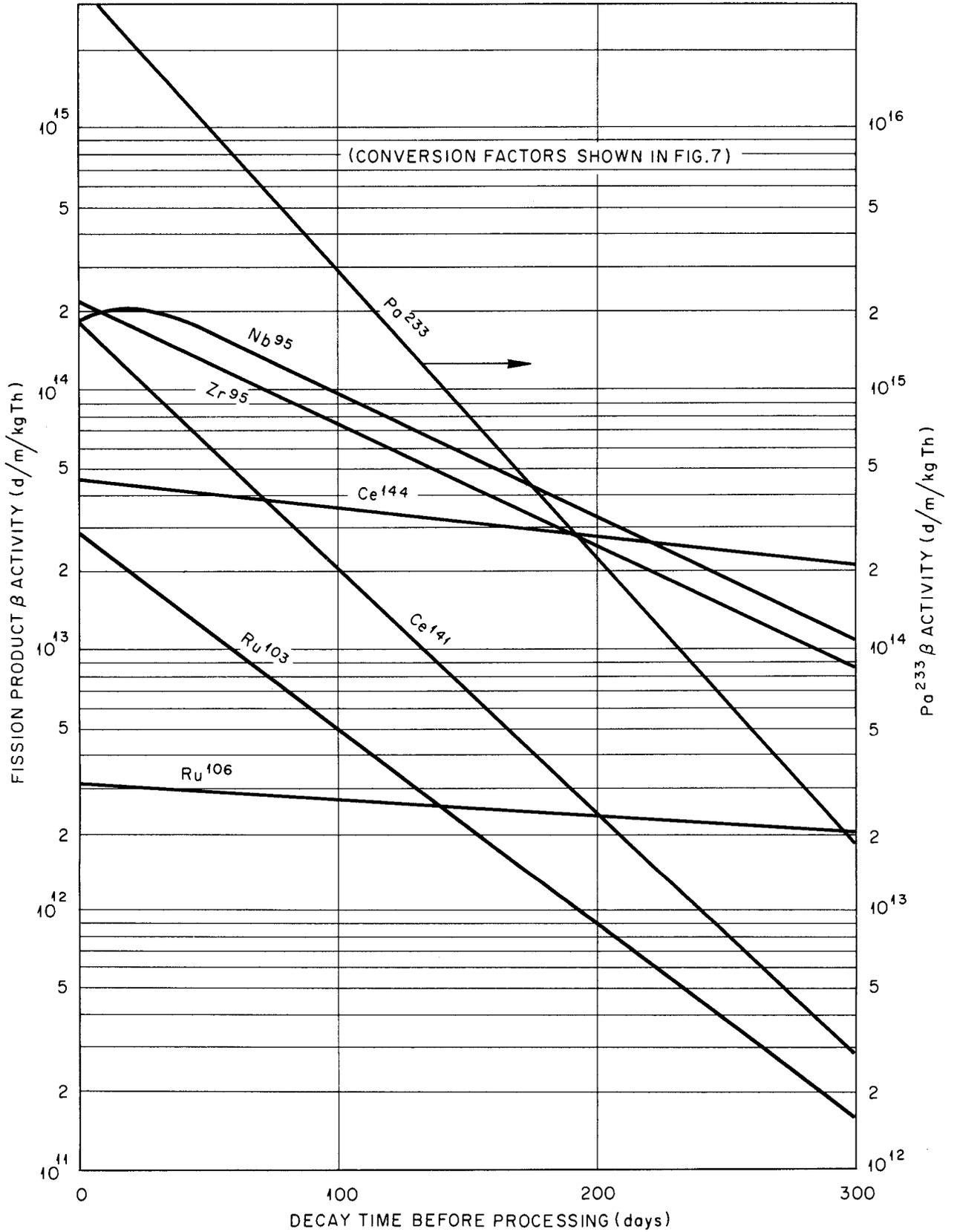


Fig. 10. Nuclide β Activity as a Function of Decay Time before Processing.
Irradiation Level, 4000 g of mass - 233 per ton of thorium;
flux, 3×10^{13} n/cm²/sec.

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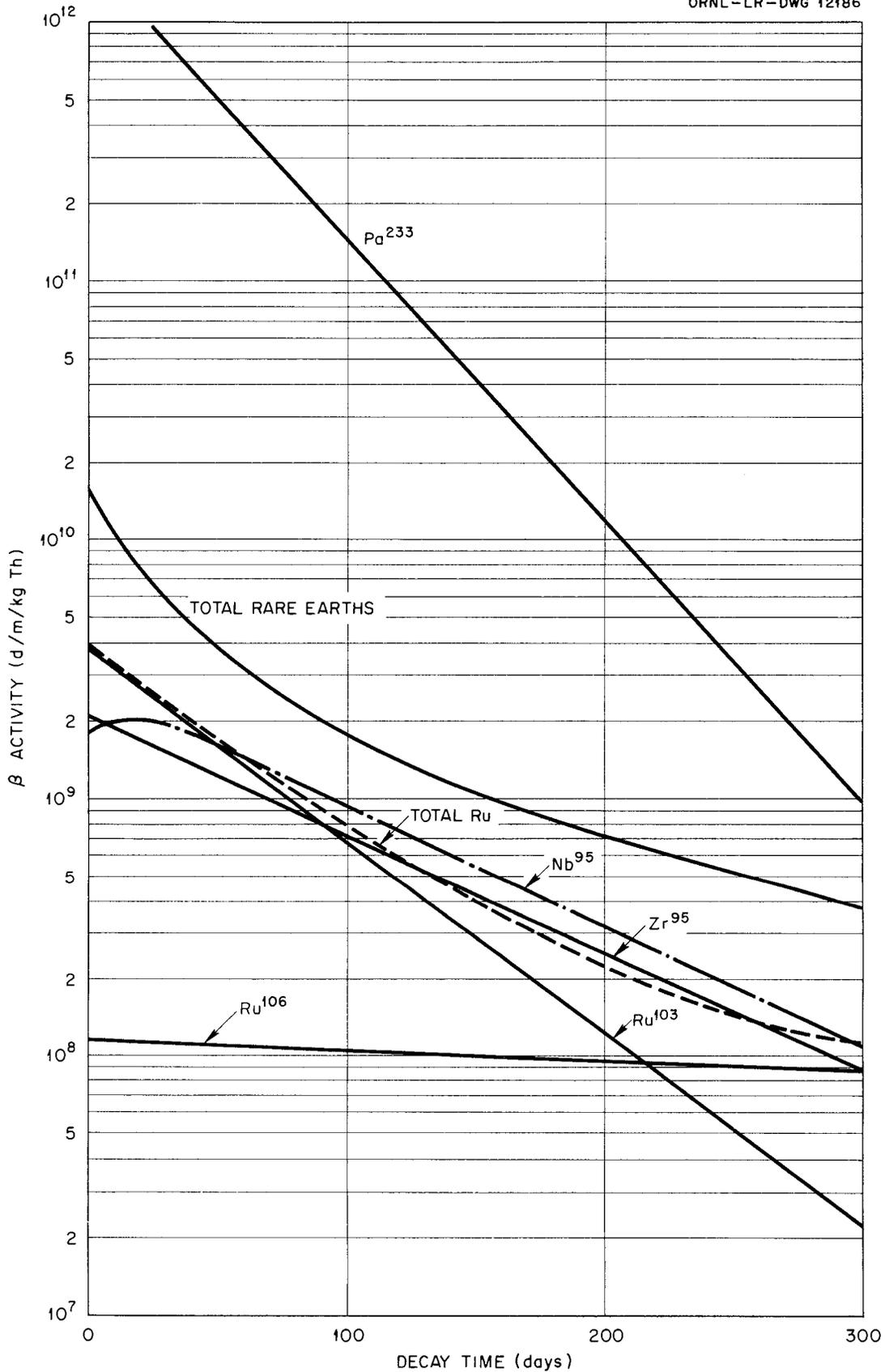


Fig. 11. Beta Activity Associated with Thorium Metal as a Function of Total Decay Time, Assuming Metal Specifications Are Met with a Ruthenium Decontamination Factor of 10 in Oxalate Precipitation and a Total Decay Time of 300 days.

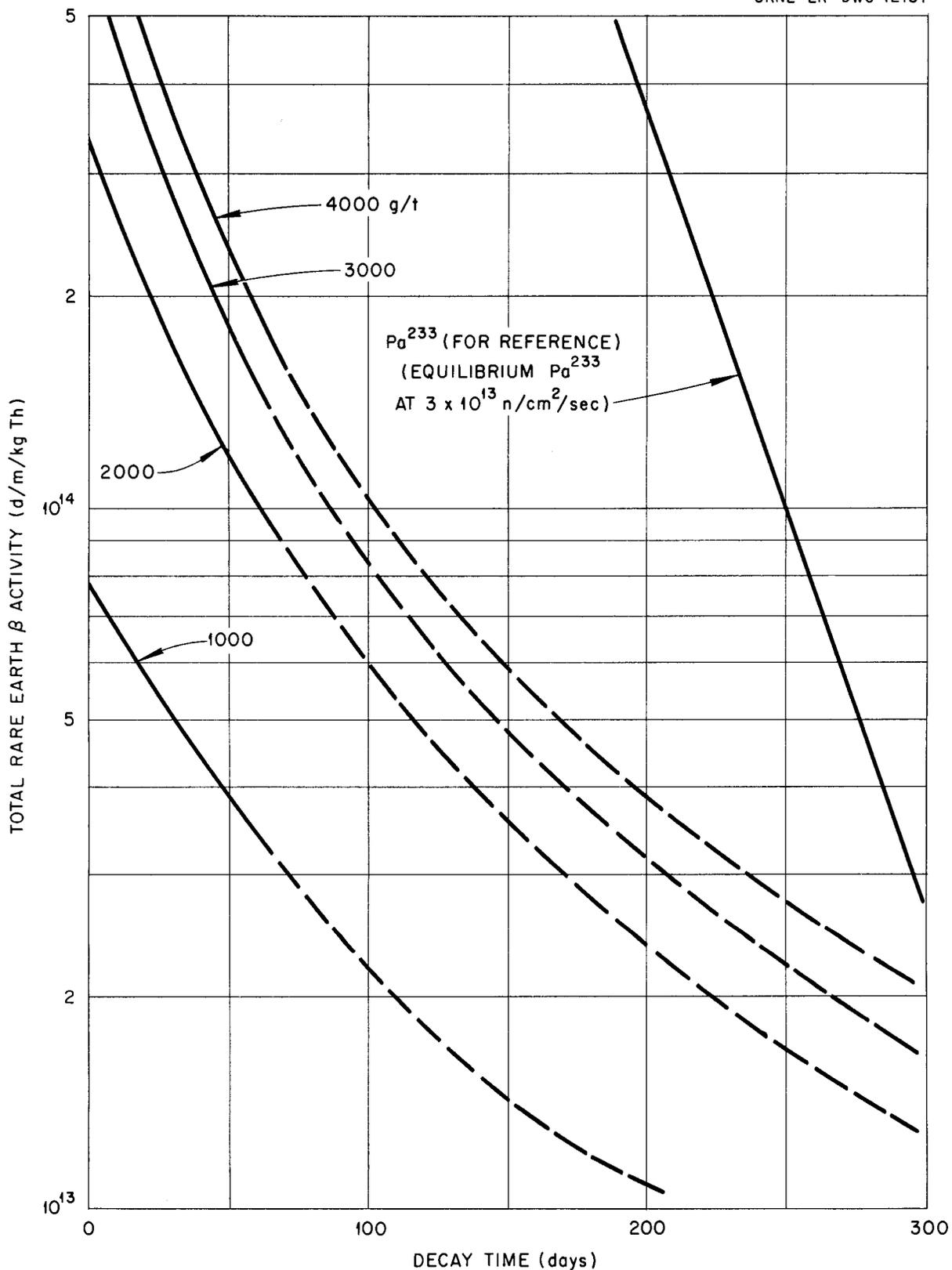


Fig. 12. Decrease in Total Rare Earth β Activity as a Function of Irradiation Level and Total Decay Time. Flux, $3 \times 10^{13} \text{ n/cm}^2/\text{sec}$.

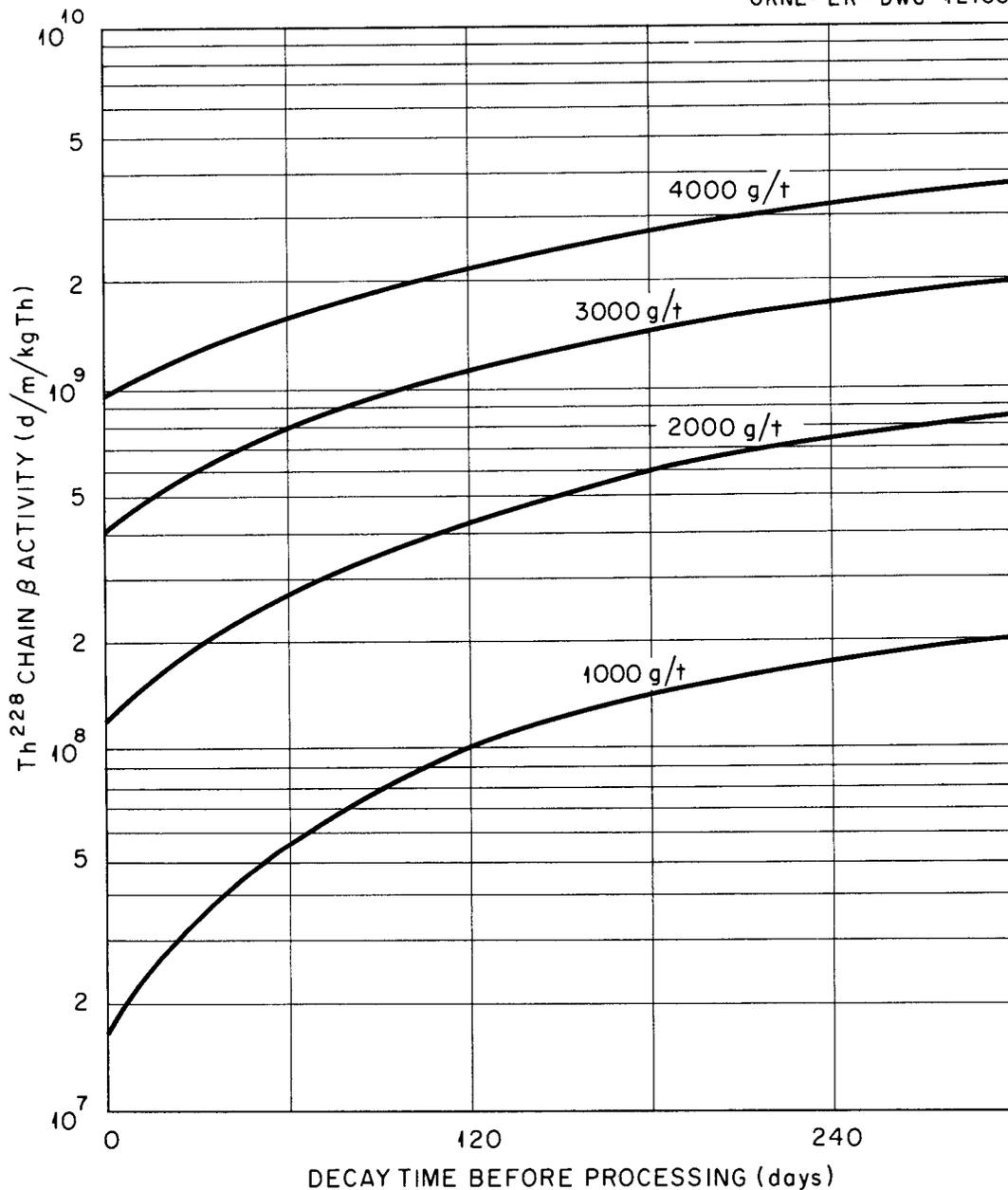


Fig. 13. Th²²⁸ Chain Activities in Thorium Metal as a Function of Decay Time before Processing. Flux, 3×10^{13} n/cm²/sec; (See Table 8). Th²³² $\sigma_{n,2n} = 3$ mb.

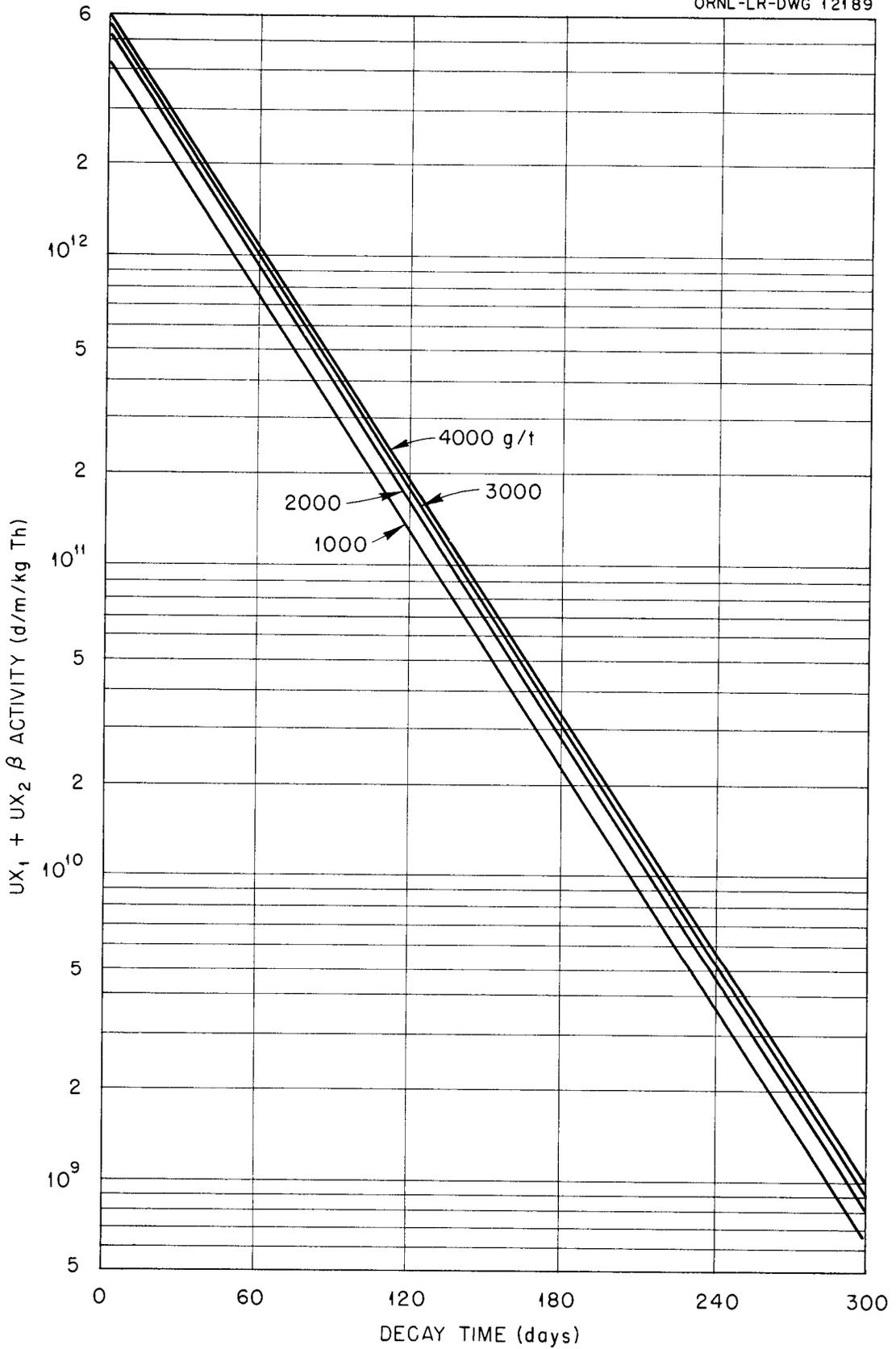


Fig. 14. $UX_1 + UX_2$ Activities in Thorium Metal as a Function of Total Decay Time. Flux, 3×10^{13} n/cm²/sec.

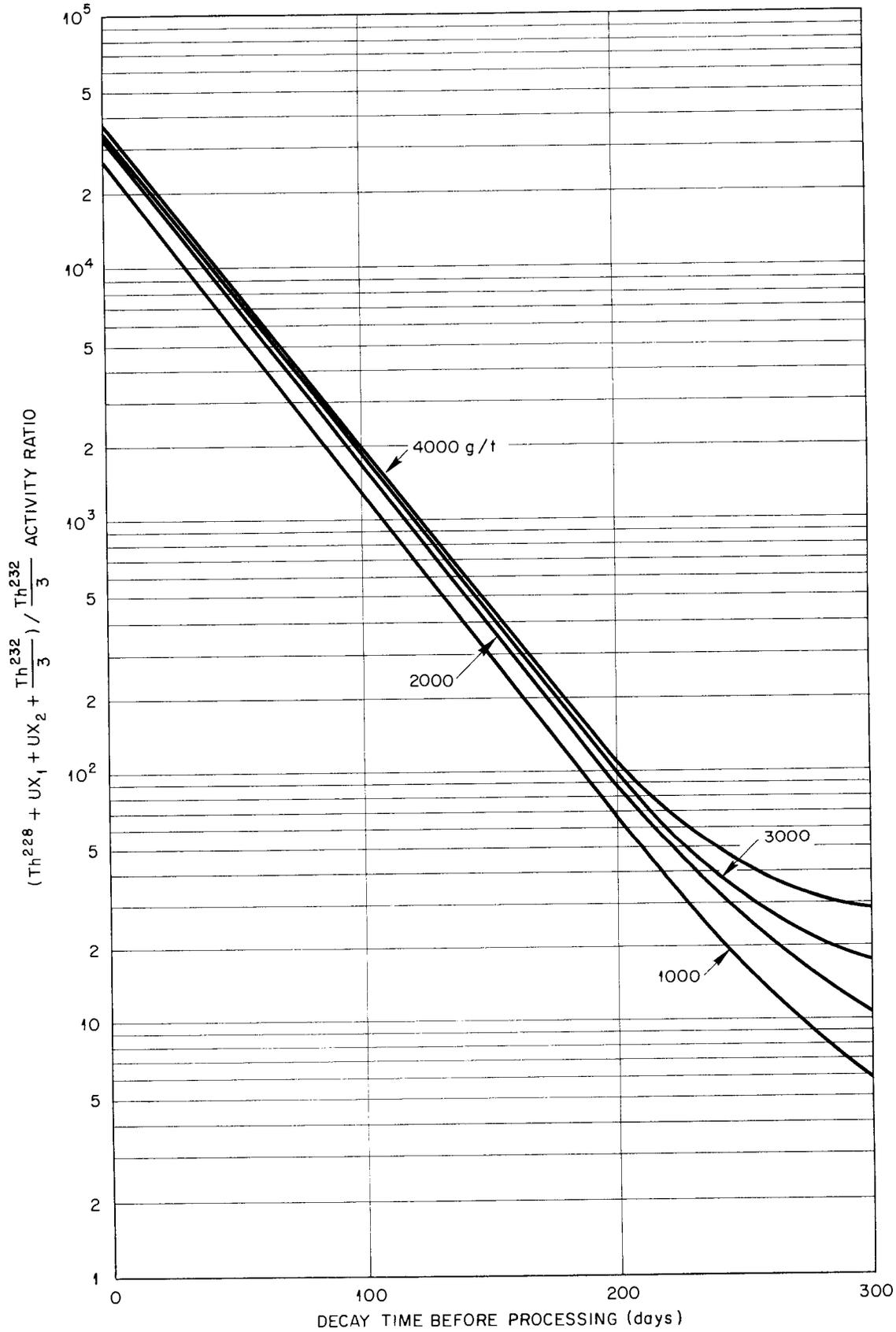


Fig. 15. Ratio of Activity Level of All Thorium Isotopes to One-third the Activity of Th^{232} as a Function of Decay Time before Processing. Flux, 3×10^{13} n/cm²/sec. $\sigma_{n,2n} = 3$ mb.

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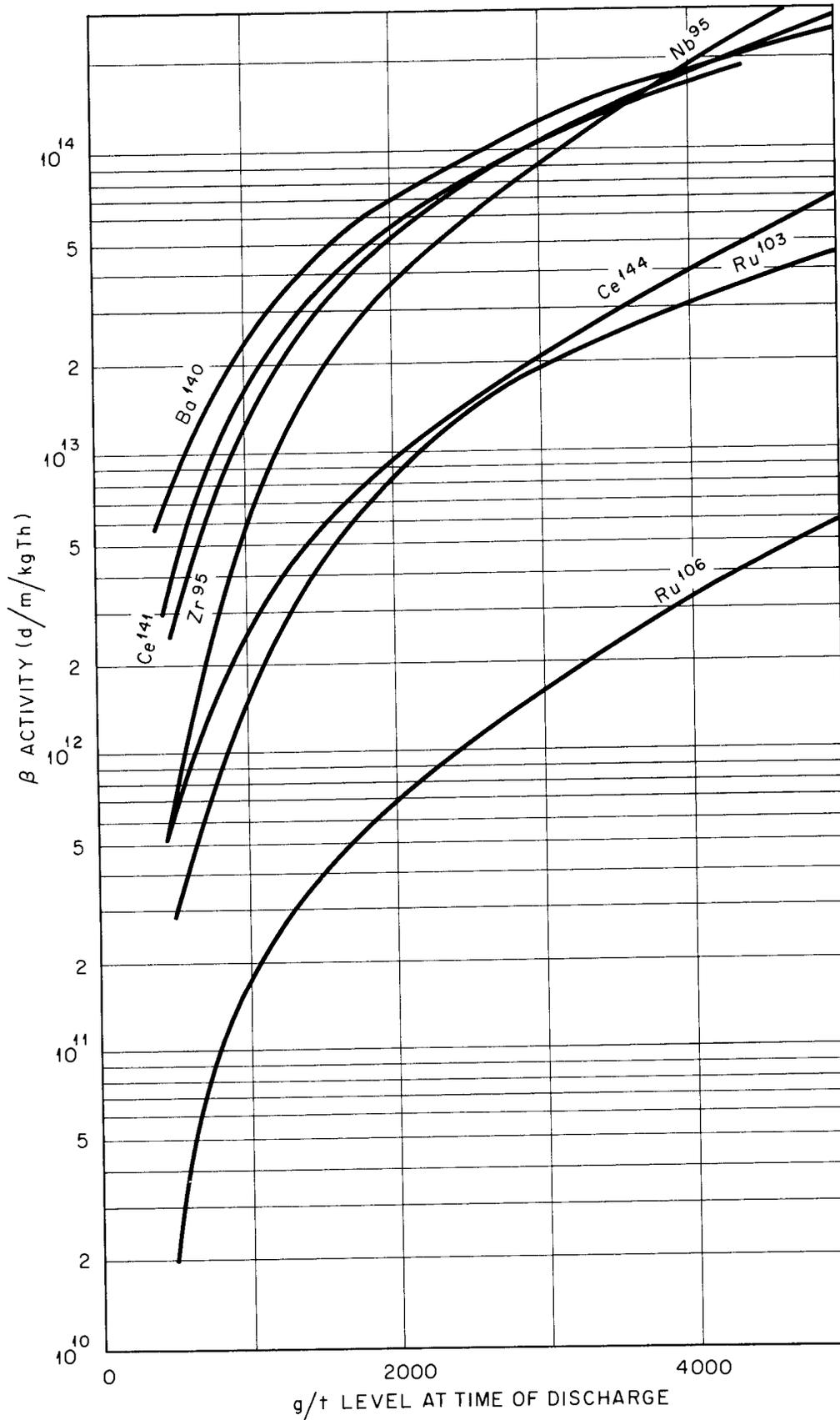


Fig. 16. Activities of Important Fission Products at Time of Reactor Discharge as a Function of g/t Level. Flux, 3×10^{13} n/cm²/sec.

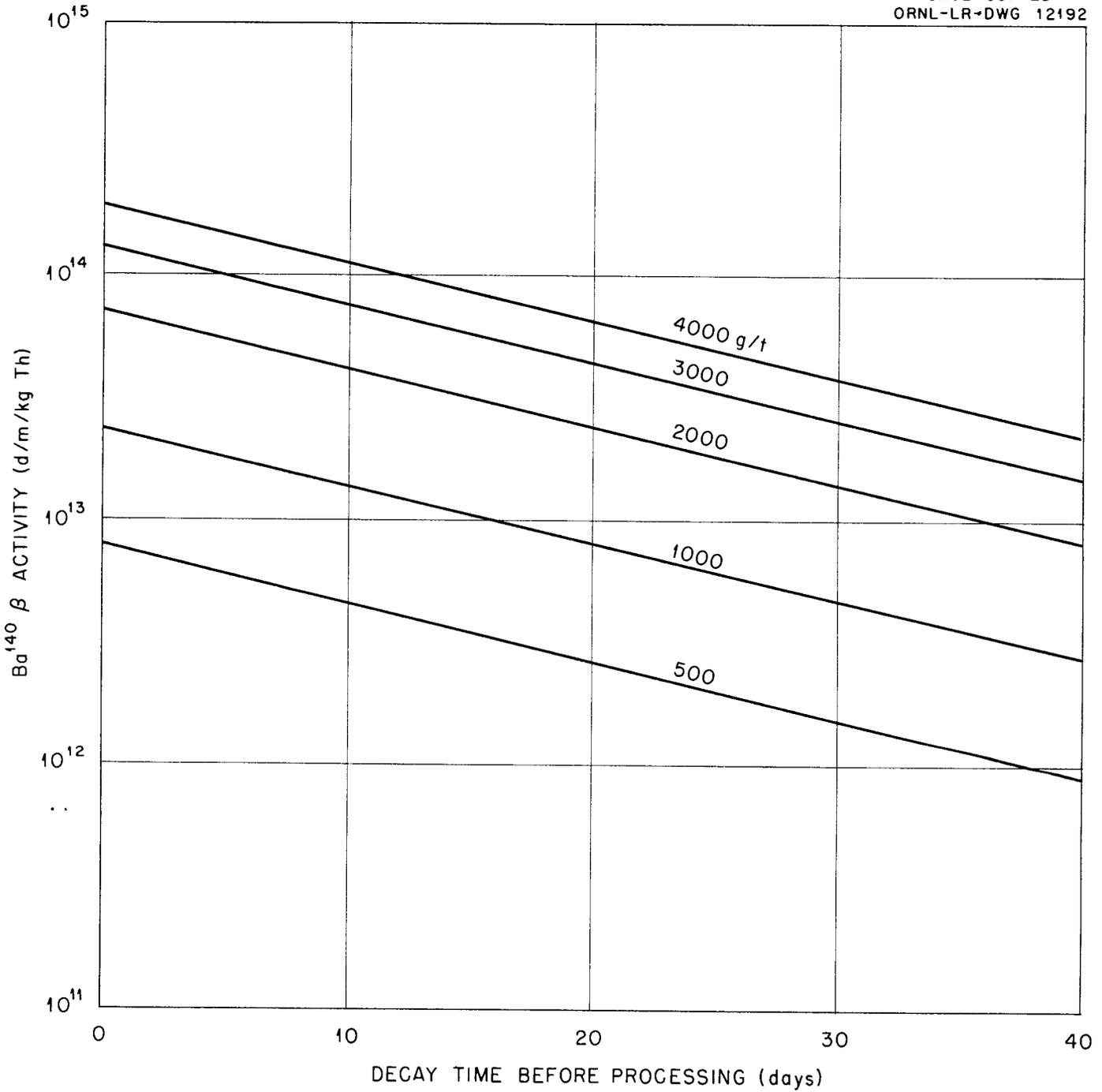


Fig. 17. Decay of Ba^{140} Activity as a Function of Decay Time. Flux, 3×10^{13} n/cm²/sec.

$$T_B = T_C + \frac{1}{3} T_R; \text{ WHERE}$$
$$T_D = T_{PT} + \frac{2}{3} T_R$$

T_C = PRE-THOREX DECAY PERIOD
 T_R = REACTOR PERIOD
 T_{PT} = POST-THOREX PROCESS PERIOD

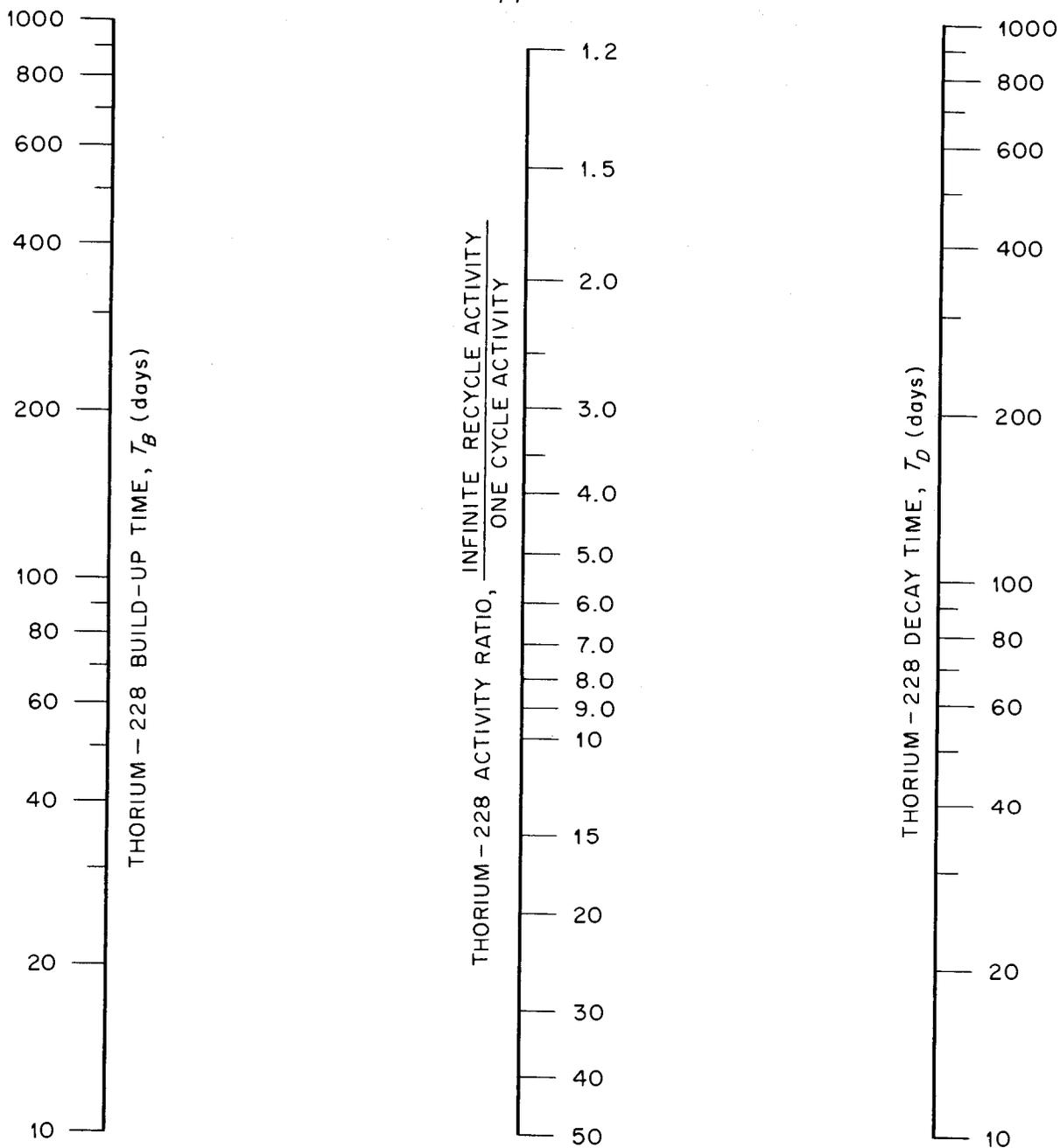


Fig.18. Nomograph for Computing Infinite Recycle Thorium-228 Activity in Thorium-232.

t = irradiation time, days or seconds

Equilibrium $\text{Th}^{232} = \text{Th}^{232}$ in equilibrium with all its decay chain daughters, e.g., Th^{232} in thorium ore (1/3 of the activity of equilibrium Th^{232} activity = activity equivalent to 1/3 of that associated with equilibrium Th^{232}); residual activity assumed to be 1/3 equilibrium Th^{232}

Figure 18 is a nomograph which may be used to calculate the activity after infinite recycle when the Th^{228} build-up time and the Th^{228} decay time are known. The value of the ratio of Th^{228} activity after infinite recycle to that after one cycle is given by the point at which a line connecting known values of T_B and T_D crosses the Th^{228} activity ratio scale.

$$T_B = T_C + 1/3 T_R = \text{Th}^{228} \text{ build-up time}$$

$$T_D = T_{PT} + 2/3 T_R = \text{Th}^{228} \text{ decay time}$$

T_C = pre-Thorex decay time

T_R = reactor irradiation time

T_{PT} = post-Thorex decay time

Table 8. Activities in Thorium Metal

Component	g/t Level	Beta Activity in Thorium Metal (d/m/kg Th)			
		60 days Pre-Thorex Decay	130 days Pre-Thorex Decay	200 days Pre-Thorex Decay	300 days Pre-Thorex Decay
Th ²²⁸ chain ^a	1000	5.8 x 10 ⁷	1.1 x 10 ⁸	1.5 x 10 ⁸	2.0 x 10 ⁸
Th ²³⁴ + Pa ²³⁴		<u>7.2 x 10¹¹</u>	<u>8.6 x 10¹⁰</u>	<u>1.14 x 10¹⁰</u>	<u>6.4 x 10⁸</u>
Total		7.2 x 10 ¹¹	8.6 x 10 ¹⁰	1.16 x 10 ⁹	8.4 x 10 ⁸
Th ²²⁸ chain ^a	2000	2.8 x 10 ⁸	4.6 x 10 ⁸	6.2 x 10 ⁸	8.3 x 10 ⁸
Th ²³⁴ + Pa ²³⁴		<u>9.0 x 10¹¹</u>	<u>1.08 x 10¹¹</u>	<u>1.44 x 10¹⁰</u>	<u>8.0 x 10⁸</u>
Total		9.0 x 10 ¹¹	1.08 x 10 ¹¹	1.5 x 10 ¹⁰	1.63 x 10 ⁹
Th ²²⁸ chain ^a	3000	7.8 x 10 ⁸	1.17 x 10 ⁹	1.51 x 10 ⁹	2.0 x 10 ⁹
Th ²³⁴ + Pa ²³⁴		<u>9.6 x 10¹¹</u>	<u>1.16 x 10¹¹</u>	<u>1.54 x 10¹⁰</u>	<u>8.8 x 10⁸</u>
Total		9.6 x 10 ¹¹	1.17 x 10 ¹¹	1.69 x 10 ¹⁰	2.88 x 10 ⁹
Th ²²⁸ chain ^a	4000	1.6 x 10 ⁹	2.3 x 10 ⁹	2.9 x 10 ⁹	3.7 x 10 ⁹
Th ²³⁴ + Pa ²³⁴		<u>1.0 x 10¹²</u>	<u>1.2 x 10¹¹</u>	<u>1.6 x 10¹⁰</u>	<u>9.0 x 10⁸</u>
Total		1.0 x 10 ¹²	1.22 x 10 ¹¹	1.9 x 10 ¹⁰	4.6 x 10 ⁹

a. Includes β activities of Pb²¹², Bi²¹², and Tl²⁰⁸.