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E. D. Arnold

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CHEMICAL TECHNOLOGY DIVISION

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E. D. Arnold

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0.0 ABSTRACT

The importance of the U^{232} daughter activities that will build up in the Thorex process thorium and U^{233} products was analyzed. The high γ activities result mainly from Bi^{212} and Tl^{208} and are great enough to affect U^{233} handling, thorium metallurgy and refabrication techniques, and the pre-Thorex cooling period.

From thorium containing 2000 g of U^{233} per ton, the recovered U^{233} will have a γ activity two years after the Thorex separation equivalent to 185 r/hr at the surface of a sphere containing 1 kg of U^{233}. This activity is much higher than has ever been considered for direct metallurgical operations.

The Bi^{212} and Tl^{208} in the thorium product are formed from the Th^{228} produced by U^{232} decay during the irradiation and pre-Thorex cooling periods. The γ activity of recycled thorium metal would be 0.083 r/hr at the surface of a 1.36- by 6-in. slug when a 60-day pre-Thorex cooling period is used. This activity is less than that of natural thorium, but recycled thorium metal that has been irradiated to higher U^{233} concentrations and cooled for longer periods before Thorex separation would have a γ activity much higher than that of natural thorium. The calculated activity for a similar natural thorium slug would be 0.237 r/hr at the surface.

Results indicate that as short a cooling period as is possible (60 days or less) would be mandatory in order to limit thorium activity.

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1.0 SUMMARY AND DISCUSSION

The results of the study of the U^{232} decay chain indicate that possibly restrictive gamma activity will be present in both the U^{233} and thorium products of the Thorex process. The surface activity of a 1-kg sphere of this U^{233} will be 185 r/hr after 2 yr storage when the metal is fabricated from the product of 2000-gt[a] material produced at a $\sigma_{02}(n,2n)$ cross-section of 3 mb. The activity of U^{233} metal for other conditions will be proportional to the gt level of the thorium, the $\sigma_{02}(n,2n)$ cross-section, and the Th^{228} (half-life of 1.9 yr) buildup factor (which depends on storage time). The radiation intensity from this sphere at a distance of 1 ft will be approximately 0.75 r/hr. The effect the U^{233} concentration would have on both surface activity and radiation intensity at 1 ft is shown in Table 1.

The radiation intensities from the thorium product, which results from Th^{228} decay, would be lower than these by a factor of about 100. However, the activity of the recycled thorium metal could be somewhat greater than that attributable to natural thorium. The effects the gt level and the pre-Thorex cooling time will have on the maximum (25 days after separation) surface activity of a recycled thorium slug are shown in Table 2. The effects the gt level and the pre-Thorex cooling time will have on the activities attributable to such a slug at a distance of 5 cm and 1 ft are shown in Table 3. The surface activity of a thorium slug made from material containing 2000 g of U^{233} per ton, produced at a $\sigma_{02}(n,2n)$ cross-section of 3 mb and cooled for 60 days, would be 83 mr/hr, and the activities at 5 cm and 1 ft would be 11.5 and 0.58 mr/hr, respectively. This is approximately half the total activity of natural thorium. The surface activities of a 10- by 60-cm cyclinder would be 1.46 times those of the slug for corresponding conditions.

Infinite recycle of thorium metal through metallurgy and irradiation would increase the activity by a factor of 2-3. The exact factor depends upon the combination of cycle times used (see Sec. 5.2).

Calculations similar to these have been made by KAPL.[1]

[a] 2000 g of U^{233} per ton of thorium.

Table 1

Radiation Intensity from a 1-kg Sphere of U^{233} at the Surface and at a Distance of 1 ft

$\sigma_{02}(n,2n)$ = 3 mb; storage time = 2 yr; sphere diameter = 4.72 cm

gt Level (g of U^{233} per ton of Th)	Intensity (r/hr)	
	At Surface	At 1 ft
1000	92	0.37
2000	185	0.75
3000	267	1.12
4000	370	1.49

Table 2

Surface Activity of Thorium Slugs as a Function of gt Level and Cooling Period

Thermal flux = 3×10^{13} n/cm^2/sec; $\sigma_{02}(n,2n) = 3$ mb; slug 1.36 in. dia., 6 in. long; 1680 g of Th; one cycle

gt Level (g of U^{233} per ton of Th)	Slug[a] Surface Activity from U^{232} Daughters (r/hr)			
	60 days Pre-Thorex Cooling	130 days Pre-Thorex Cooling	200 days Pre-Thorex Cooling	300 days Pre-Thorex Cooling
1000	0.017	0.031	0.043	0.059
2000	0.083	0.135	0.181	0.244
3000	0.228	0.343	0.443	0.590
4000	0.475	0.676	0.846	1.088

[a] Similar slug of natural thorium with no removal of Ra^{228} and Ac^{228} would read 0.237 r/hr at surface (neglecting $Ac^{228}\gamma$); similar slug of natural thorium processed by Ames solvent (TBP) extraction method to remove Ra^{228} and Ac^{228} with 1.9 yr decay after processing would read 0.12 r/hr at surface.

Table 3

Activity of Thorium Slugs at Distances of 5 cm and 1 ft as a Function of gt Level and Cooling Period

Thermal flux = 3×10^{13} n/cm^2/sec; $\sigma_{02}(n,2n) = 3$ mb; slug 1.36 in. dia., 6 in. long; 1680 g of Th; one cycle

gt Level (g of U^{233} per ton of Th)	Slug[a] Activity from U^{232} Daughters (mr/hr)							
	At 5 cm				At 1 ft			
	Pre-Thorex Cooling 60 days	Pre-Thorex Cooling 130 days	Pre-Thorex Cooling 200 days	Pre-Thorex Cooling 300 days	Pre-Thorex Cooling 60 days	Pre-Thorex Cooling 130 days	Pre-Thorex Cooling 200 days	Pre-Thorex Cooling 300 days
1000	2.4	4.4	6.0	8.2	0.12	0.22	0.31	0.42
2000	11.5	19.1	25.5	34.4	0.58	0.93	1.3	1.75
3000	32	48.3	62.0	83	1.62	2.4	3.1	4.1
4000	67	95	118	152	3.3	4.8	6.0	7.7

[a] Similar slug of natural thorium with no removal of Ra^{228} and Ac^{228} would read ~33 mr at 5 cm, ~1.6 mr at 1 ft; similar slug of natural thorium processed by Ames solvent (TBP) extraction method to remove Ra^{228} and Ac^{228} with 1.9 yr decay after processing would read ~17 mr at 5 cm and ~0.8 mr at 1 ft; a 2.56- by 15.5-cm slug at ORNL, Chemical Technology Division, read 4.5 mr at 5 cm and 0.35 mr at 1 ft (age since reduction unknown).

The fast neutron cross-section for the (n,2n) reaction on thorium was estimated and found to agree reasonably well with experimental results. The (n,2n) cross-section for thorium within a Hanford receptacle slug was estimated to be 9 mb based on the thermal flux. Loading thorium in a separate process tube should reduce this cross-section to 1 mb. The cross-sections for other types of loadings were also estimated, and indications are that the expected (n,2n) cross-section in the currently designed Savannah River loading would be of the order of 5-10 mb. The effect of buildup of U^{233} within a thorium slug was also interpreted. Indications of this effect are that the apparent (n,2n) cross-section would increase linearly with U^{233} concentration.

The contribution of β particles in the 1-kg sphere was estimated to be approximately 30% of the total surface γ contribution. This β contribution was estimated by assuming that one-half of the β particles produced in the outside skin of 1.7×10^{-2} cm (estimated mean range[2]) would penetrate the sphere and deliver an energy contribution of 0.33 of the maximum β which is 2.0 Mev (avg). The effect of β particles would be negligible at distances from the sphere.

2.0 CONCLUSIONS

In order to maintain low γ activities in both the U^{233} and thorium that would be within physical reason, the following conclusions may be drawn:

1. Irradiation, if possible, should be undertaken in regions of the reactor in which the ratio of fast to thermal flux is a minimum.
2. As low a gt-level material as is economically feasible should be produced.
3. A short cooling period before Thorex processing would minimize the activity of thorium metal. A 60-day cooling period is suggested to accomplish this aim. A longer post-Thorex cooling period would then be needed for Th^{234} decay.[3] Handling of thorium in this way would produce thorium metal that is only slightly more radioactive than natural thorium.
4. Weapons-grade U^{233} may have to be reprocessed periodically in order to maintain a low U^{232} daughter activity.
5. The short pre-Thorex cooling period necessary to reduce Th^{228} chain activities in the thorium product will necessitate a higher Pa^{233} concentration in the Thorex extraction column feed stream and thus a high β activity in this stream and in the extraction column.

3.0 INTRODUCTION

The natural radioactive series of thorium, 4n, was the second decay scheme studied. The unique feature of this decay scheme is that there is a short cut produced by 70-yr U^{232}. Whereas Th^{232} must decay through Ra^{228} (6.7 yr) and Ac^{228} (6.13 hr) in order to form Th^{228} (1.9 yr), it is possible to produce U^{232} by a series of neutron reactions in a thorium-U^{233} production reactor. This U^{232} then decays directly by α emission to form Th^{228}. It is thus possible to produce much larger concentrations of Th^{228} and its daughters than would be present in natural thorium. Also, the U^{232} will be present in the U^{233} product from the Thorex process, and thus much larger concentrations of these daughters will build up in the uranium than would be present in the thorium.

The U^{232} chain has been discussed in previous works,[3] but mainly from the standpoint of high-energy α emission in the U^{233} product. These high energy α's, which also result from the daughters, have sufficient energy to cause (α,n) reactions on light metals, which may contaminate the weapons-grade U^{233}. The high-energy α's must therefore be kept at a minimum, or the light metal contaminants must be minimized in order to make satisfactory weapons-grade U^{233}. At the time that the α energies were under study, very little attention was given to the high-energy γ's also present in the decay chain. It was therefore the purpose of this report to study the U^{232} decay chain from the aspect of radiation hazards.

It is of interest to compare U^{232} effects in the U^{233} production scheme to naturally occurring and capture-chain-produced activities in uranium and plutonium.

The U^{232} chain, which will be present in the Thorex U^{233} product, is much more important than activities which will be present in either U^{238}, U^{235}, or Pu^{239}. Protactinium^{231}, which is the high-cross-section (250 b) intermediate of the U^{232} production scheme, will be in equilibrium with U^{235} present in natural uranium. However, the total U^{232} and daughter

activities produced from the Pa^{231} in uranium irradiated to 600 g of plutonium per ton will be a factor of approximately 10-20 below the activity level that will build up in thorium irradiated to 4000 g of U^{233} per ton. The Pa^{231} may be separated from uranium by the gaseous diffusion process . Uranium^{237} is produced during an irradiation of U^{235}, but this material decays in about 135 days[a] to 2.25 x 10^6-yr Np^{237}, which thus creates a large block in the decay scheme.

Very little activity will be produced in Pu^{239} except its own γ activities of 0.42 and 0.2 Mev, since Pu^{239} decays to U^{235} and any Pu^{240} which may be present will decay to long-lived U^{236}. Both these decay schemes thus have large blocks which stop rapid buildup of decay products.

Therefore, from the standpoints of the natural radioactive decay series and/or the short cuts that may be created, the activity produced in the thorium-U^{233} production scheme will be much more important than that produced in either U^{235} irradiation or Pu^{239} production.

(a) Time to cool below 10^9 d/m/kg U^{235}.

4.0 URANIUM^{233} STUDY

4.1 Activity in Thorex U^{233} Product

The source of the high γ activity in the Thorex U^{233} product is the associated U^{232}. The U^{232} is produced predominantly (>95%) by the reactions

$$Th^{232}(n,2n)Th^{231} \xrightarrow[24.6h]{\beta^-} Pa^{231}(n,\gamma)Pa^{232} \xrightarrow[1.32d]{\beta^-} U^{232}$$

and both the amount produced and the U^{232}/U^{233} α activity ratio increase as the irradiation time and flux increase (see Figs. 1 and 2). The (n,2n) reaction involves neutrons of energies greater than 6.37 Mev,[a] and the effective thermal cross-sections are therefore considered to be in the range 2-13 mb. This range of values was used in calculating[3] the product concentrations shown in the curves in Figs. 1 and 2. However, there have been some indications[4] that the lower limit is ~1 mb. Other cross-section values used in this report to obtain product concentrations are

$\sigma_{02}(n,\gamma) = 7.0$ b $\qquad \sigma_{03}(n,\gamma) = 1350$ b $\qquad \sigma_{23}(n,f) = 505$ b

$\sigma_{13}(n,\gamma) = 150$ b $\qquad \sigma_{11}(n,\gamma) = 250$ b $\qquad \sigma_{23}(n,\gamma) = 55$ b

The differential equations involved in the formation of U^{232} are:

$$\frac{dN_{22}}{dt} = N_{11}\phi\sigma_{11} \tag{1}$$

$$\frac{dN_{11}}{dt} = \phi N_{02}\sigma_{02}(n,2n) - N_{11}\phi\sigma_{11} \tag{2}$$

Integration of these equations gives

$$N_{11} = \frac{\phi N_{02}\sigma_{02}(n,2n)}{\phi\sigma_{11}}\left(1 - e^{-\phi\sigma_{11}t}\right) \tag{3}$$

$$N_{22} = N_{02}\sigma_{02}(n,2n)\left[\phi t - \frac{1}{\sigma_{11}}\left(1 - e^{-\phi\sigma_{11}t}\right)\right] \tag{4}$$

[a] The neutron binding energy.

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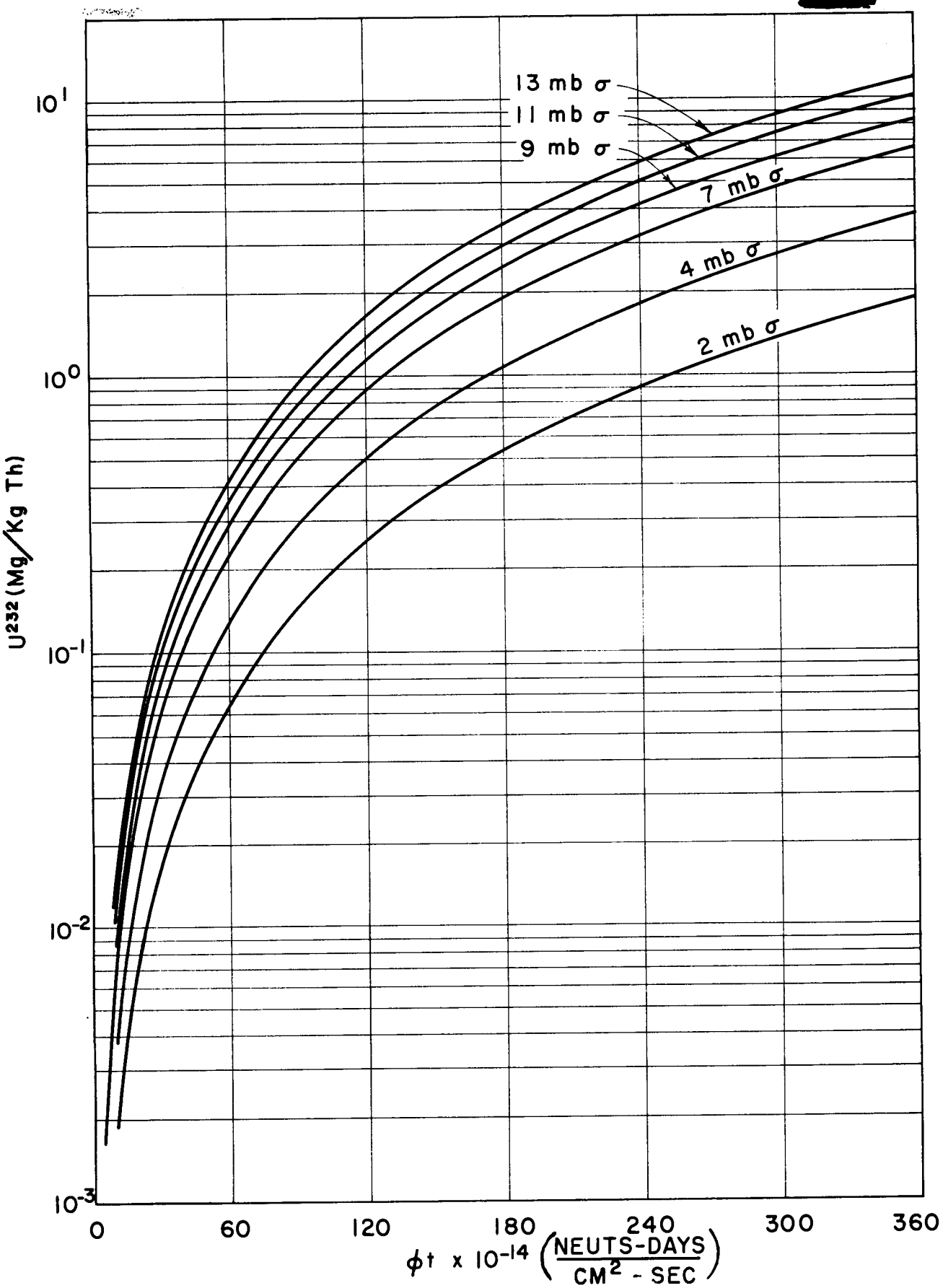

FIGURE 1

EFFECT OF IRRADIATION TIME AND FLUX ON U^{232} FORMATION

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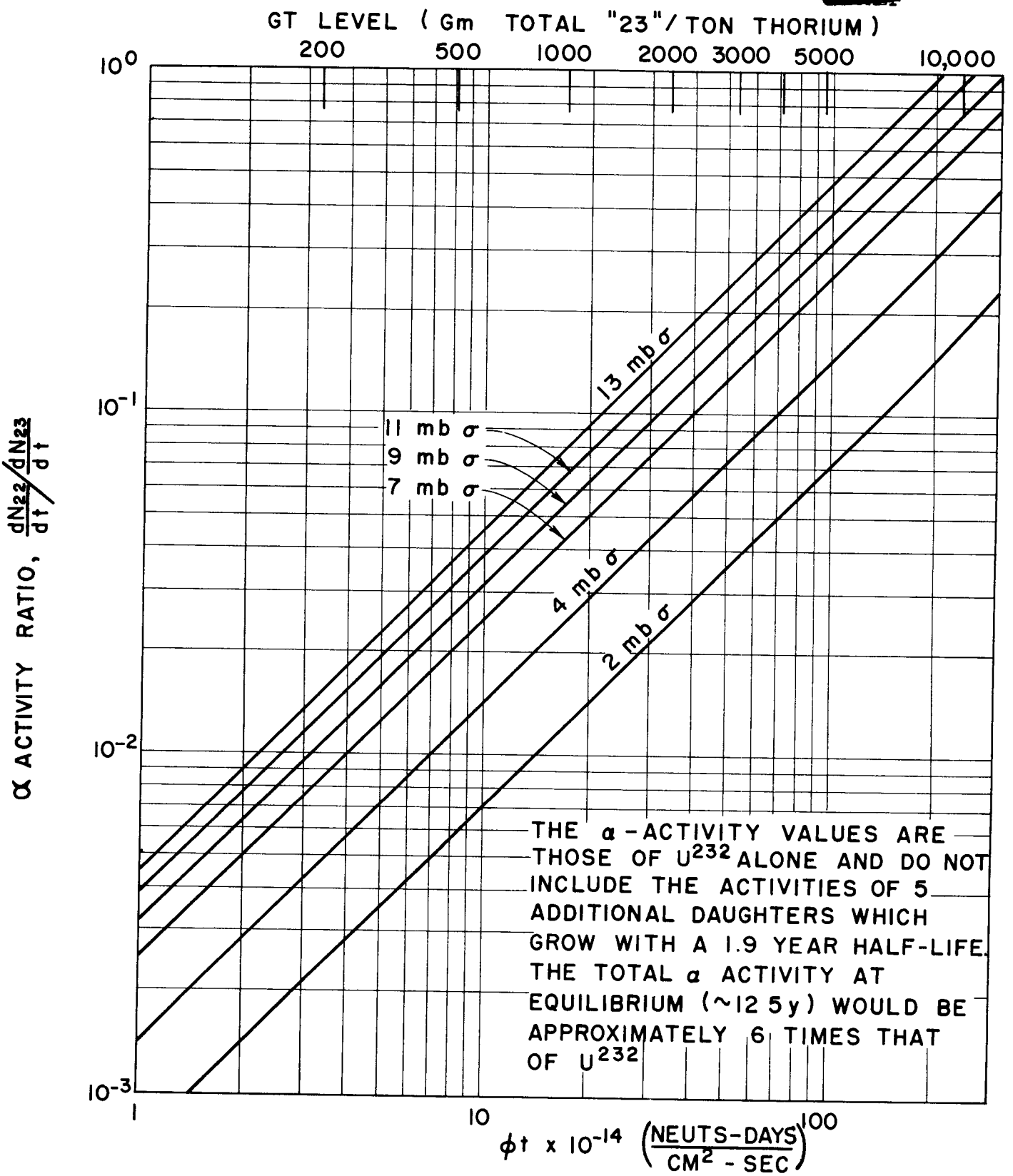


FIGURE 2

EFFECT OF IRRADIATION TIME AND FLUX ON U^{232} TO U^{233} ALPHA ACTIVITY RATIO, ATD

where the symbols have the usual significance, i.e., N = concentration, t = time, σ = cross-section, ϕ = flux, and the subscripts 22, 11, and 02 refer to U^{232}, Pa^{231}, and Th^{232}, respectively.

Uranium^{232} decays by the chain

$$U^{232} \xrightarrow[70y]{\alpha} Th^{228} \xrightarrow[1.9y]{\alpha} Ra^{224} \xrightarrow[3.64d]{\alpha} Rn^{220} \xrightarrow[55s]{\alpha} Po^{216}$$

$$\xrightarrow[0.16s]{\alpha} Pb^{212} \xrightarrow[10.6h]{\beta^-} Bi^{212} \xrightarrow{60.5m} \begin{cases} \xrightarrow[34\%]{\alpha} Tl^{208} \xrightarrow[3.1m]{\beta^-} Pb^{208} \\ \xrightarrow[66\%]{\beta^-} Po^{212} \xrightarrow[0.3\,\mu s]{\alpha} Pb^{208} \end{cases} \quad (s)$$

that is, to the series of daughters which build in with a half-life of 1.9 yr (that of Th^{228}). The remaining daughters may be assumed to be in equilibrium with Th^{228} since their half-lives are much shorter. The concentration of Th^{228} as a function of time is

$$N_{08} = \frac{\lambda_{22}}{\lambda_{08} - \lambda_{22}} N^o_{22}(e^{-\lambda_{22}t} - e^{-\lambda_{08}t}) + N^o_{08}\, e^{-\lambda_{08}t} \tag{5}$$

and the activity of Th^{228} as a function of time, assuming $N^o_{08} = 0$, is

$$A_{08} = \frac{\lambda_{08}}{\lambda_{08} - \lambda_{22}} A^o_{22}\, (e^{-\lambda_{22}t} - e^{-\lambda_{08}t}) \tag{6}$$

where A^o_{22} = U^{232} activity at time of reactor discharge
λ_{08} = decay constant of Th^{228} = 3.647×10^{-1} yr^{-1}
λ_{22} = decay constant of U^{232} = 9.9×10^{-3} yr^{-1}

The burnup of Th^{228} due to neutron capture is neglected.

In order to compare Thorex U^{233} products, it was necessary to determine the α activity of each product due to U^{232} and then to determine the α, β, or γ activity due to the daughters. The decay and energy data for the U^{232} decay scheme given above are shown in Table 4.

Table 4

Decay Data for Daughters of U^{232} (5)

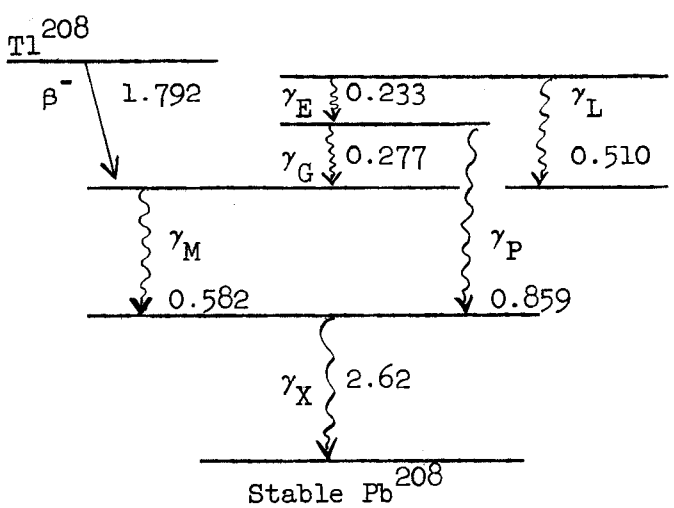
Chain Member	Half-life	Decay Method	Energy (Mev)	%
Th^{228}	1.9y	α	5.423	72
		α	5.338	28
		γ	0.083, 0.087	---
Ra^{224}	3.64d	α	5.66	95
		α	5.448	4.6
			5.194	0.4
		γ	~0.250	~5
Rn^{220}	55s	α	6.282	100
Po^{216}	0.16s	α	6.774	100
		β^-	---	0.014
Pb^{212}	10.6h	β^-	0.331	88
		β^-	0.569	12
		γ	0.238	90
		γ	0.115, 0.300	10
Po^{212}	0.3μs	α	8.776	~100
		α	9.5-10.5	Few ppm
Bi^{212}	60.5m	α	(See next page)	33.7
		β^-	Avg 2.25 (See next page)	66.3
Tl^{208}	3.1m	β^-	1.792	(See next page)

Table 4, Continued

Bi^{212}

	α, 33.7%			β⁻, 66.3%		
	Energy (Mev)	%	Associated γ Energy (Mev)	Associated γ	%(a)	Energy (Mev)
α_1	5.601	1.10	0.451, 0.491	γ_1	18.5	0.72
α_2	5.620	0.16	0.431, 0.471	γ_2	16	0.83
α_3	5.762	1.80	0.287, 0.327	γ_3	5.5	1.03
α_4	6.044	69.8	0.040	γ_4	4.5	1.34
	6.074	~77	---		1.8	1.35
α_5	6.083	27.2	---	γ_5	7.0	1.61
	6.113	~23	---		5.0	1.60
				γ_6	7.0	1.81
					3.0	1.80
				γ_7	2.5	2.20

(a) Total percentage used in calculations which will place probable γ activity approximately 10% high.



The buildup of the remaining daughters was calculated in an analogous manner (by equations similar to Eqs. 5 and 6) by using the appropriate branching ratios and percentage disintegrations. The activity fractions of U^{232}, Th^{228}, and the important daughters as a function of time and the γ source strengths of the important daughters as a function of both time and α-activity ratio[a] are listed in Table 5. Figure 3 shows the effect of storage time on the β-γ source strength in the U^{233} product.

4.2 Effect of U^{232} Daughter Activities on U^{233} Product Activity

The effect of U^{232} daughter activities on the activity of the U^{233} product is shown below by calculations based on a sphere of 1 kg of U^{233}. The activity is expressed both as surface activity and the radiation intensity at 1 ft. The following assumptions are made:

Product contains 4000 g of Mass-233 per ton

$\sigma_{02}(n,2n) = 0.003$ barn

$\alpha_{22}/\alpha_{23} = 0.07$ from Fig. 1

Time of storage = 2.0 yr (approximately the Th^{228} half-life)

The expected source strengths of the important daughters are:

$$Bi^{212}\ \gamma = 8.40 \times 10^{10} \times 0.07 = 5.88 \times 10^{9}\ d/s/kg\ U^{233} \tag{7}$$

$$Tl^{208}\ \gamma\ (each) = 6.11 \times 10^{10} \times 0.07 = 4.28 \times 10^{9}\ d/s/kg\ U^{233} \tag{8}$$

The radiation intensities from a unit volume of a solid source, I_o^* (see Sec. 7), are

$$I_o^* = AN_o/4\pi X^3 \tag{9}$$

where X^3 = volume of 1 kg. Then

$$I_o^* = \frac{5.88 \times 10^9}{3.7 \times 10^{10}} \times \frac{1.18}{169} \times \frac{1.18 \times 10^7}{4\pi \times 55.5} = 18.8\ r/hr/cc \text{ for } Bi^{212} \text{ for 1 steradian} \tag{10}$$

[a] The term "α-activity ratio" is defined as $N_{22}\lambda_{22}/N_{23}\lambda_{23}$, where λ_{23} is the decay constant for $U^{233} = 4.28 \times 10^{-6}\ yr^{-1}$.

Table 5

Effect of Storage Time upon Buildup of Activity in Thorex U^{233} Product

Storage Time (yr)	U^{232} α Activity Fraction, $N_{22}\lambda_{22}/N^{o}_{22}\lambda_{22}$	Th^{228} α Activity Fraction, $N_{08}\lambda_{08}/N^{o}_{22}\lambda_{22}$	Total Daughter α Activity ÷ $N_{22}\lambda_{22}$	Bi^{212} β Activity Fraction, 0.66 Total Bi^{212} Activity	Bi^{212} γ Activity Fraction, 0.708 Total Bi^{212} β Activity	Bi^{212} γ Activity Based on Initial $\alpha_{22}/\alpha_{23} = 1$ (d/sec/kg U^{233})	Tl^{208} γ Activity Fraction, 0.34 Total Bi^{212} Activity	Tl^{208} γ Activity Based on Initial $\alpha_{22}/\alpha_{23} = 1$ (d/sec/kg U^{233})
0.1	0.9990	0.0365	0.1824	0.0240	0.01698	5.96×10^{9}	0.0124	4.33×10^{9}
0.2	0.9980	0.0729	0.3648	0.0481	0.0340	1.19×10^{10}	0.0248	8.72×10^{9}
0.3	0.9970	0.1035	0.5181	0.0684	0.0484	1.70×10^{10}	0.0352	1.23×10^{10}
0.4	0.9960	0.1384	0.6770	0.0893	0.0632	2.22×10^{10}	0.0460	1.60×10^{10}
0.5	0.9950	0.1662	0.8313	0.1097	0.0777	2.73×10^{10}	0.0565	1.97×10^{10}
0.7	0.9930	0.2244	1.1226	0.1482	0.1049	3.68×10^{10}	0.0763	2.67×10^{10}
1.0	0.9901	0.3039	1.5200	0.2008	0.1424	4.99×10^{10}	0.1034	3.63×10^{10}
1.2	0.9880	0.3523	1.7620	0.2325	0.1646	5.77×10^{10}	0.1198	4.21×10^{10}
1.5	0.9852	0.4185	2.0930	0.2763	0.1956	6.87×10^{10}	0.1422	4.98×10^{10}
1.7	0.9832	0.4581	2.2904	0.3023	0.2140	7.51×10^{10}	0.1558	5.46×10^{10}
2.0	0.9802	0.5121	2.561	0.3380	0.2393	8.40×10^{10}	0.1742	6.11×10^{10}
2.5	0.9752	0.5895	2.948	0.3891	0.2755	9.67×10^{10}	0.2004	7.03×10^{10}
3.0	0.9703	0.6535	3.268	0.4313	0.3054	1.07×10^{11}	0.2222	7.80×10^{10}
3.5	0.9654	0.7059	3.530	0.4660	0.3299	1.15×10^{11}	0.2400	8.42×10^{10}
4.0	0.9604	0.7485	3.743	0.4940	0.3497	1.22×10^{11}	0.2545	8.93×10^{10}
4.5	0.9555	0.7836	3.918	0.5172	0.3661	1.28×10^{11}	0.2665	9.35×10^{10}
5.0	0.9505	0.8215	4.107	0.5421	0.3838	1.34×10^{11}	0.2792	9.79×10^{10}
6.0	0.9406	0.8519	4.259	0.5623	0.3980	1.39×10^{11}	0.2896	1.01×10^{11}
8.0	0.9238	0.8939	4.470	0.5900	0.4177	1.47×10^{11}	0.3040	1.06×10^{11}
10.0	0.9058	0.9044	4.522	0.5969	0.4227	1.48×10^{11}	0.3075	1.08×10^{11}
12.0	0.8879	0.8998	4.499	0.5939	0.4205	1.47×10^{11}	0.3060	1.08×10^{11}
14.0	0.8703	0.8884	4.442	0.5864	0.4152	1.46×10^{11}	0.3021	1.05×10^{11}
16.0	0.8534	0.8742	4.372	0.5770	0.4085	1.44×10^{11}	0.2972	1.04×10^{11}
20.0	0.8203	0.8432	4.216	0.5565	0.3940	1.38×10^{11}	0.2866	1.00×10^{11}
24.0	0.7886	0.8106	4.053	0.5351	0.3788	1.33×10^{11}	0.2756	9.67×10^{10}
28.0	0.7576	0.7787	3.894	0.5140	0.3639	1.28×10^{11}	0.2647	9.29×10^{10}
32.0	0.7283	0.7486	3.744	0.4941	0.3498	1.22×10^{11}	0.2545	8.93×10^{10}

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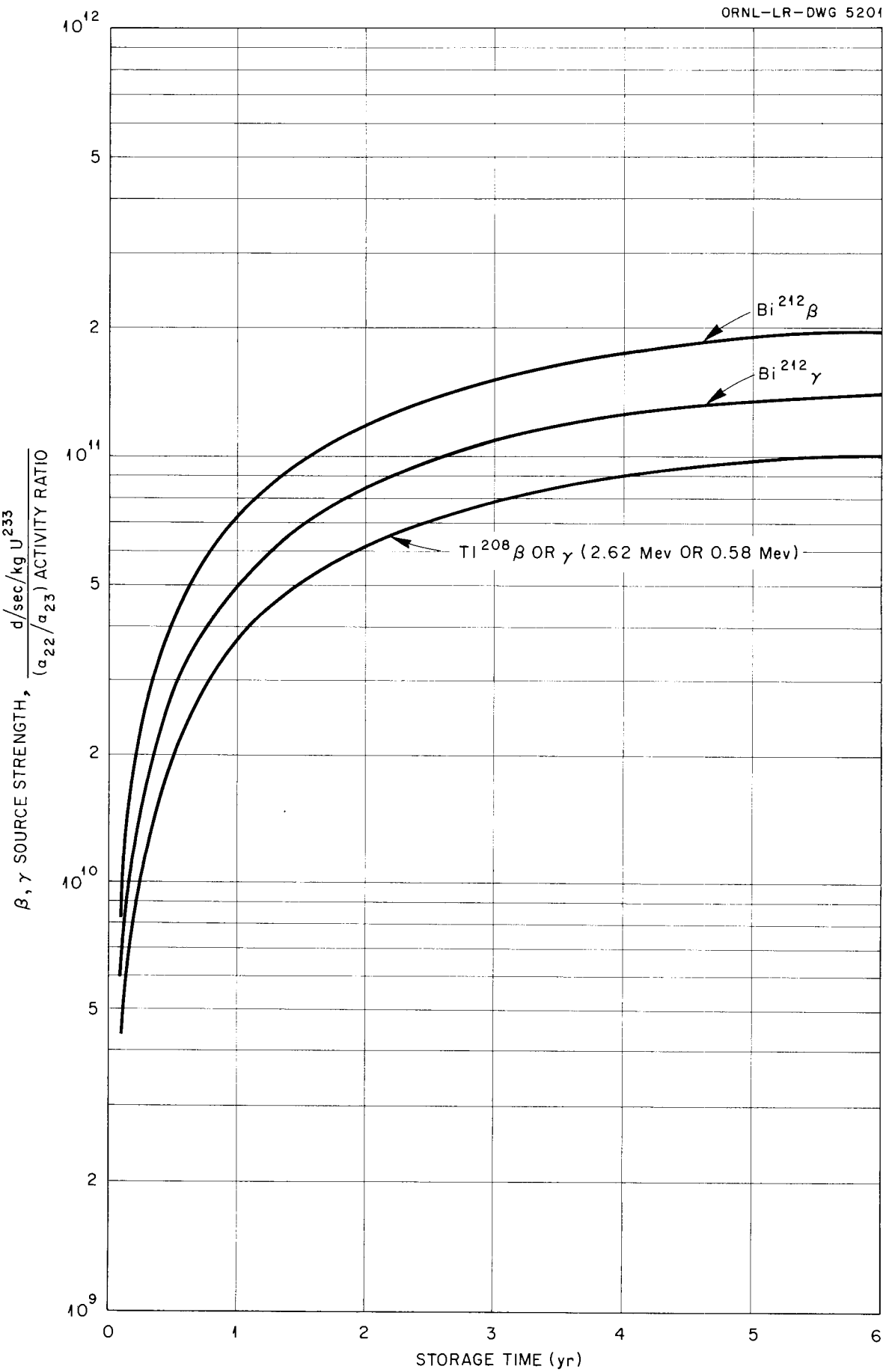

Fig. 3. Effect of Storage Time on Beta-Gamma Source Strength in Uranium-233.

Similarly,

$I_o^* = 30.4$ r/hr/cc for the Tl^{208} 2.62-Mev γ

$I_o^* = 6.74$ r/hr/cc for the Tl^{208} 0.58-Mev γ

The intensity on the surface of a 1-kg sphere, the radius, R, of which =

$\sqrt[3]{55.5 \times 3/4\pi} = 2.36$ cm, is

$$I = \frac{\pi I_o^*}{\mu^2 R}(2\mu R + e^{-2\mu R} - 1) \tag{11}$$

Using the following values[6] of the absorption coefficient, μ, in which compton scattering and buildup are considered,

$\mu = 0.86$ for 1.18-Mev γ (Bi^{212} average)

$\mu = 0.56$ for 2.62-Mev γ

$\mu = 2.25$ for 0.58-Mev γ

$$I_{Bi^{212}} = \frac{\pi \times 18.8}{\overline{0.86}^2 \times 2.36}(2 \times 0.86 \times 2.36 + e^{-0.86 \times 2.36} - 1) = 107.5 \text{ r/hr} \tag{12}$$

Similarly,

$I_{1Tl^{208}} = 246$ r/hr

$I_{2Tl^{208}} = 17$ r/hr

Intensity from surface of sphere = 370 r/hr

The activities 1 ft from this sphere may be estimated as

$$I_{Bi^{212}} = \frac{18.8 \times \pi \times \overline{2.36}^2}{0.86 \times \overline{30.48}^2} = 0.41 \text{ r/hr} \tag{13}$$

$$I_{1Tl^{208}} = \frac{3.04 \times \pi \times \overline{2.36}^2}{0.56 \times \overline{30.48}^2} = 1.02 \text{ r/hr} \tag{14}$$

$$I_{2Tl^{208}} = \frac{6.74 \times \pi \times \overline{2.36}^2}{2.25 \times \overline{30.48}^2} = 0.06 \text{ r/hr} \qquad (15)$$

Total $\overline{1.49}$ r/hr

The surface activities and the intensity at a point 1 ft from the surface may be estimated in an analogous manner for other conditions. The surface and 1-ft activities as a function of the gt level have already been given in Table 1.

4.3 Data on U^{233} Product Activities

The results of the ORNL Pilot Plant extraction of 19 specially irradiated Hanford thorium slugs, covered in HW-33019,[7] indicate that the U^{232} α pulse activity was 2% and that the U^{232} concentration of the product was 0.001%. The material contained 1132 g of Mass-233 per ton of thorium. The effective thermal cross-section for the (n,2n) reaction on thorium can be estimated from these data to be 3 mb (Fig. 2). The U^{232} pulse analyses of the last two shipments of U^{233} product from the ORNL Thorex Pilot Plant indicated that the average thermal cross-section for the (n,2n) reaction was also 3 mb. The results of the first shipment are in error because of the analytical procedure used.

A 1.7-kg hollow sphere of U^{233} (3 in. o.d., 2.5 in. i.d.) from Los Alamos gave a reading of 54 mr at 1 ft. This material had been reduced to the metal about 4 months previous to the activity check. The estimated γ activity from the Bi^{212} and Tl^{208} that would build up in the uranium product recovered from thorium containing 800 g of U^{233} per ton (estimated Interim-23 process product level) during 4 months is 70 mr at 1 ft. The assumed (n,2n) cross-section was 3 mb. A sample of 35 g of U^{233}, also produced during the Interim-23 program, as uranyl nitrate solution containing ~300 g of U^{233} per liter, read 160 mr at 6 in. Inserting lead sheet between the sample and the cutie pie gave the following readings:

Lead Sheet Thickness (in.)	Reading (mr/hr)	Lead Sheet Thickness (in.)	Reading (mr/hr)
1/8	37	1	10
1/4	29	1-1/2	5
1/2	20	1-7/8	3

This material had been separated approximately 26 months previous to the last radiation reading. The radiation was first read on an ion chamber on December 8, 1952, as 5.07 mv and the background reading was 0.6 mv. The last ion chamber reading on January 28, 1955, was 26.8 mv. The expected γ readings due to Bi^{212} and Tl^{208} from 35 g of U^{233}, considered as a point source, would be 110 mr at 6 in. Scintillation counter gamma readings with 100% geometry and counting through 5/32 in. of lead gave 2.12×10^4 c/m/mg U^{233}. The theoretical activity for Bi^{212} and the Tl^{208} γ would be 1.32×10^5 d/m/mg U^{233}. Correcting disintegrations for 5/32 in. of lead (using the reduction factor indicated by readings at 6 in.) indicated a counting rate of 2.22×10^4 c/m/mg U^{233}.

5.0 THORIUM STUDY

5.1 Activity Produced in Thorium

The principal source of high-energy gamma activity in the recycled thorium from which the U^{233} is separated is Th^{228}, a daughter of U^{232} (see Sec. 4.1 for decay scheme). The Th^{228} is produced during reactor irradiation and during the cooling period before the Thorex separation process is used. The Th^{228} concentration (see Eqs. 5 and 6, Sec. 4.1) is therefore a function of the reactor flux, the irradiation time, the Th^{232} (n,2n) cross-section, and the pre-Thorex cooling period.

The total decay time of the U^{232}, i.e., the buildup time of Th^{228}, is equal to the cooling period plus some fraction of the irradiation period. In order that this fraction may be estimated, the assumption will be made that the neutron reactions convert Th^{232} directly to Pa^{231} and Pa^{231} directly to U^{232} with no time delay. It will also be assumed that the Pa^{231} concentration is far from equilibrium in order that the approximate production equations may be used. This assumption gives a U^{232} concentration only 7% greater than that given by Eq. 4 for thorium containing 5000 g of U^{233} per ton. This is a reasonable error in light of the other uncertainties of the production scheme.

The concentration of Pa^{231} as a function of time is

$$N_{11} = N_{02}\phi\ \sigma_{02}(n,2n)t \tag{16}$$

and the U^{232} concentration in terms of the Th^{232} concentration is

$$N_{22} = \int_0^t N_{11}\phi\ \sigma_{11}dt = \int_0^t N_{02}\phi\ \sigma_{02}(n,2n)t\phi\ \sigma_{11}\ dt$$

$$= \frac{N_{02}\ \sigma_{02}(n,2n)\ \sigma_{11}\phi^2 t^2}{2} \tag{17}$$

The concentration of Th^{228} as a function of time is

$$N_{08} = \int_0^t N_{22}\lambda_{22}\ dt = \int_0^t \frac{N_{02}\ \sigma_{02}(n,2n)\ \sigma_{11}\phi^2 t^2}{2}\ \lambda_{22}\ dt$$

$$= \frac{N_{02}\ \sigma_{02}(n,2n)\ \sigma_{11}\phi^2 t^3\ \lambda_{22}}{6} \tag{18}$$

Then the concentration of Th^{228} as a function of the U^{232} concentration at the time of reactor discharge would be, letting t become T_R, the total reactor irradiation time,

$$N_{08} = \frac{N_{02}\,\sigma_{02}(n,2n)\,\sigma_{11}\phi^2 T_R{}^2}{2} \times \frac{T_R}{3}\ \lambda_{22}$$

$$N_{08} = N_{22}\lambda_{22}T_R/3 \tag{19}$$

The total U^{232} decay time or the Th^{228} buildup time, T_B, is therefore approximately one third the total reactor irradiation time plus the cooling time:

$$T_B = T_R/3 = T_C \tag{20}$$

where T_C = pre-Thorex cooling time. The Th^{228} buildup factor is shown as a function of the U^{232} decay time in Table 6 and Fig. 4.

The daughters of Th^{228}, specifically Bi^{212} and Tl^{208}, are the chief sources of the high-energy gamma activity. The daughter activities build in with essentially a half-life of 3.64 days (that of Ra^{224}). After equilibrium has been reached, in about 25 days, they then decay with the 1.9-yr half-life of Th^{228}. The calculation of the buildup of the daughters of Th^{228} in thorium was performed in a manner analogous to that of Th^{228} from U^{232}. The effect of decay time on the decay and buildup factors of Th^{228} and its daughters is shown in Fig. 5 and the source strength of these important daughters in terms of gt level, α_{22}/α_{23} activity ratio, and the Th^{228} buildup factor is shown in Fig. 6 as a function of decay time. The actual activity may be found by multiplying the source strength (Fig. 6) by the product of the gt level/10,000, the α_{22}/α_{23} activity ratio, and the Th^{228} buildup factor for the desired conditions. The decay and buildup factors for Th^{228} and its daughters are listed in Table 7 as a function of time. The source strength of the Bi^{212} β, the Bi^{212} γ, and the Tl^{208} β, γ activities are given in Table 8 as a function of the time, the gt level, the α_{22}/α_{23} activity ratio, and the Th^{228} buildup factor.

Table 6

Thorium^{228} and Daughters in Thorex Thorium Product

Decay Time Before Separation (yr)	Fraction of Th^{228} Activity, $N_{08} \lambda_{08}/N_{22}^{o} \lambda_{22}$
0.1	0.0365
0.2	0.0729
0.3	0.1035
0.4	0.1354
0.5	0.1662
0.7	0.2244
1.0	0.3039
1.2	0.3523
1.5	0.4185
1.7	0.4581
2.0	0.5121
2.5	0.5895
3.0	0.6535

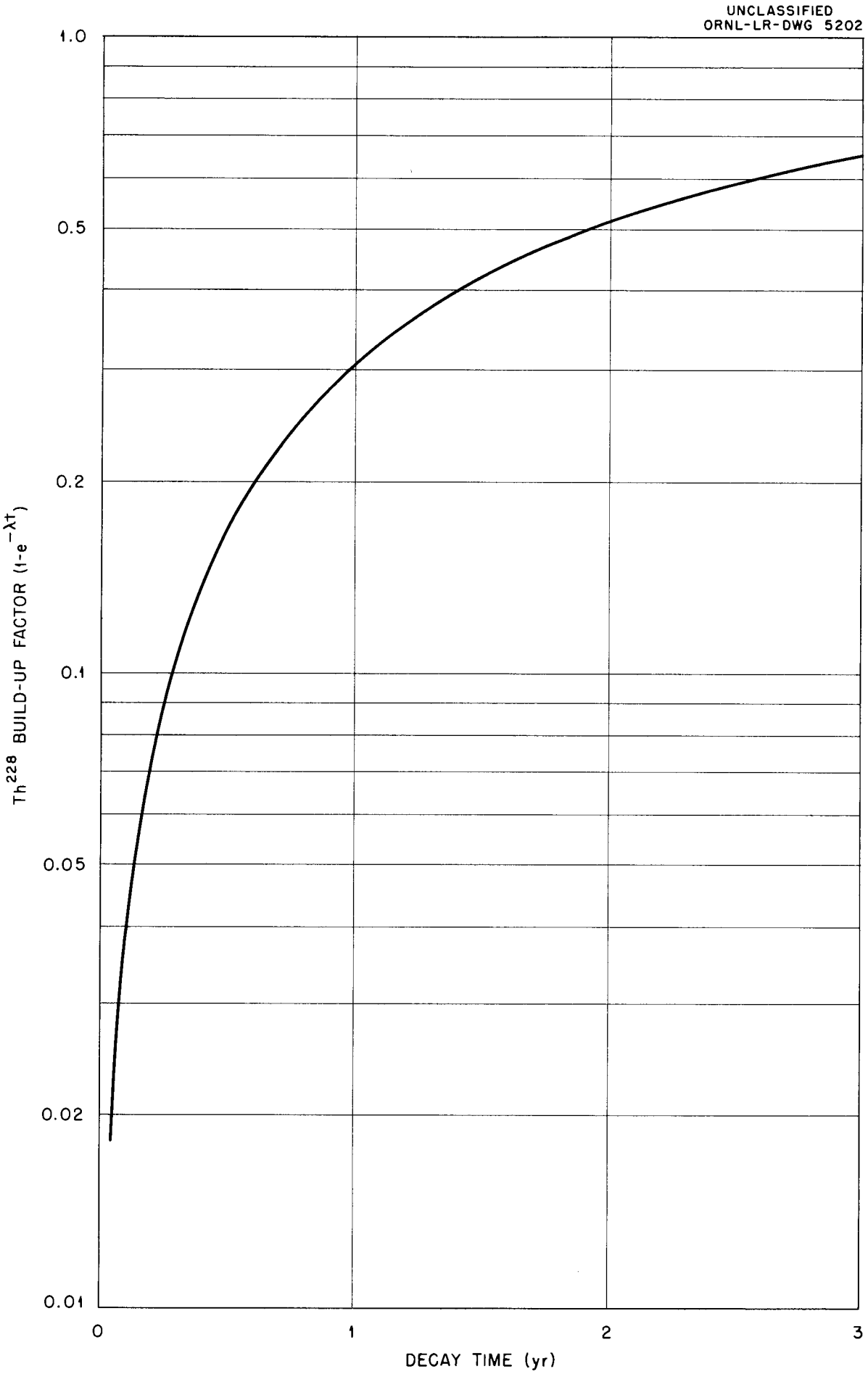

Fig. 4. Effect of Decay Time Before Separation on Thorium-228 Build-Up Factor.

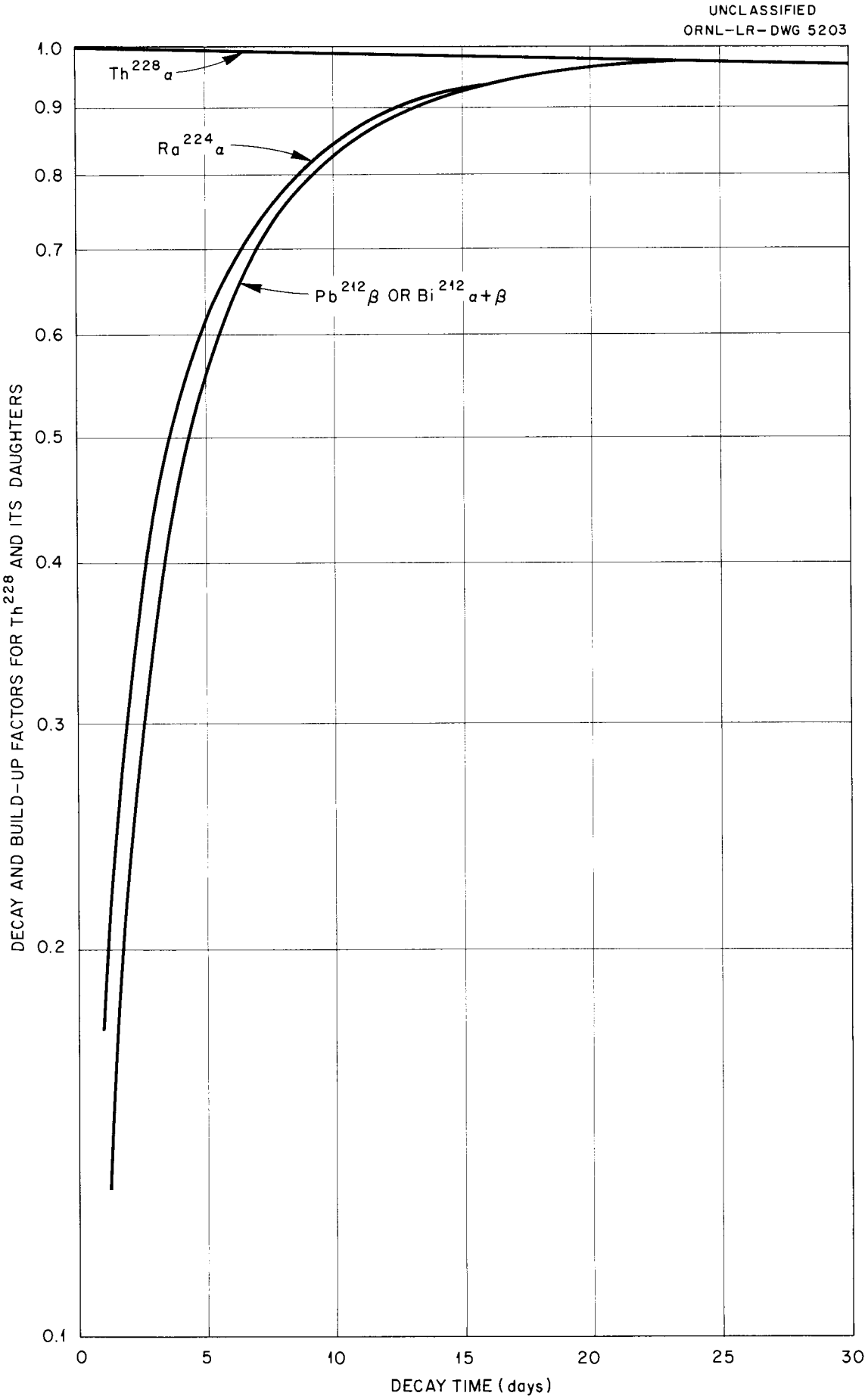

Fig. 5. Effect of Decay Time on Thorium-228 Alpha Activity and the Alpha and Beta Activities of Its Daughters.

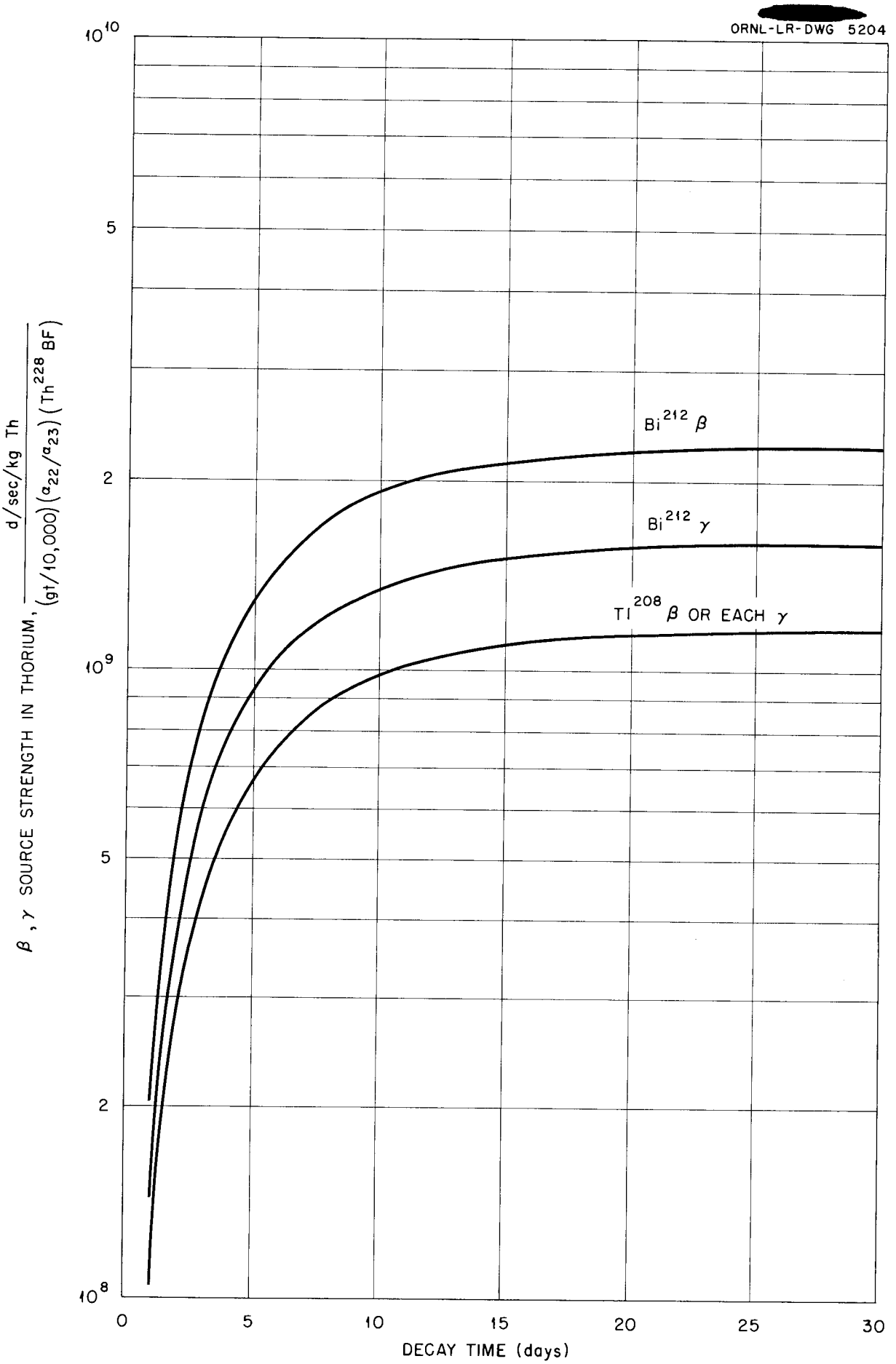

Fig. 6. Effect of Decay Time on Beta-Gamma Source Strength in Thorium-232.

Table 7

Decay and Buildup of Th^{228} and/or Daughters

Time (days)	Th^{228} α Activity Fraction, $N_{08}\lambda_{08}/N^{o}_{08}\lambda_{08}$	Ra^{224} α Activity Fraction, $N_{Ra^{224}}\lambda_{Ra^{224}}/N^{o}_{08}\lambda_{08}$	Pb^{212} β Activity Fraction = Total Bi^{212} α + β Activity Fraction, $N_{Pb^{212}}\lambda_{Pb^{212}}/N^{o}_{08}\lambda_{08}$	Bi^{212} β Activity Fraction, 0.66 Total Bi^{212} Activity Fraction	Bi^{212} γ Activity Fraction, 0.708 Total Bi^{212} β Activity Fraction	Tl^{208} γ Activity Fraction, 0.34 Total Bi^{212} Activity Fraction
0	1.0000	0	0	0	0	0
1	0.999	0.1733	0.0878	0 0579	0.0410	0.0298
2	0.998	0.3165	0.2280	0.1505	0.1065	0.775
3	0 997	0 4345	0.3578	0 2361	0 1671	0 1216
4	0.986	0.5319	0 4679	0.3088	0.2186	0 1596
5	0 995	0.6123	0 5593	0.3691	0.2613	0.1901
6	0.994	0.6786	0.6349	0.4190	0.2903	0.2158
7	0.993	0.7332	0.6972	0.4601	0.3258	0.2370
8	0.992	0 7781	0.7485	0 4940	0.3497	0.2545
9	0.991	0 8151	0.7907	0 5218	0.3694	0.2688
10	0.9901	0.8456	0.8255	0 5448	0.3857	0.2807
12	0.9881	0.8910	0.8755	0.5792	0.4100	0.2983
15	0.9851	0.9325	0.9252	0.6106	0.4323	0.3146
17	0.9832	0 9488	0 9440	0.6230	0.4411	0.3209
20	0.9802	0.9631	0.9602	0.6337	0.4486	0.3265
25	0.9753	0.9719	0.9713	0 6410	0.4538	0.3302
30	0.9705	0.9723	0.9724	0 6418	0.4544	0.3306
35	0.9656	0 9694	0 9699	0.6401	0.4532	0.3297
40	0.9608	0.9654	0.9659	0 6375	0.4514	0.3284
45	0.9560	0.9609	0.9615	0.6346	0.4493	0.3269
50	0.9513	0.9562	0.9568	0.6315	0.4471	0.3253

Table 8

Effect of Buildup Time on Daughter Source Strengths in Thorex Thorium Product

Based on an α_{22}/α_{23} ratio of 1 at time of reactor discharge

10,000 g of U^{233} per ton of thorium

$\sigma_{02}(n,2n) = 12$ mb

Th^{228} buildup factor = 1

Time (days)	Bi^{212} β Activity (d/s/kg Th)	Bi^{212} γ Activity (d/s/kg Th)	Tl^{208} β and each γ Activity (d/s/kg Th)
0	0	0	0
1	2.03×10^8	1.44×10^8	1.04×10^8
2	5.28×10^8	3.74×10^8	2.72×10^8
3	8.28×10^8	5.86×10^8	4.27×10^8
4	1.08×10^9	7.64×10^8	5.60×10^8
5	1.29×10^9	9.13×10^8	6.67×10^8
6	1.47×10^9	1.04×10^9	7.57×10^8
7	1.61×10^9	1.14×10^9	8.32×10^8
8	1.73×10^9	1.22×10^9	8.93×10^8
9	1.83×10^9	1.29×10^9	9.43×10^8
10	1.91×10^9	1.35×10^9	9.85×10^8
12	2.03×10^9	1.44×10^9	1.047×10^9
15	2.14×10^9	1.51×10^9	1.104×10^9
17	2.18×10^9	1.54×10^9	1.126×10^9
20	2.22×10^9	1.57×10^9	1.146×10^9
25	2.25×10^9	1.59×10^9	1.159×10^9
30	2.25×10^9	1.59×10^9	1.160×10^9
35	2.24×10^9	1.58×10^9	1.157×10^9
40	2.24×10^9	1.58×10^9	1.153×10^9
45	2.23×10^9	1.58×10^9	1.147×10^9
50	2.21×10^9	1.56×10^9	1.142×10^9

5.2 Effect of Recycle on Activity of Thorium

Thorium^{228} is formed during one irradiation cycle; however, the discharged thorium will probably be reprocessed and recycled back to the reactor before this Th^{228} has decayed. The lengths of the Th^{228} buildup period and decay period must be known if the increase in activity after successive cycles and/or infinite cycles is to be determined. It has been shown (Sec. 5.1, Eq. 20) that the Th^{228} buildup period is equal to the cooling period plus one-third of the reactor period. The decay period will then be the remainder of the cycle, or equal to the post-Thorex process period plus two-thirds of the reactor period. These terms are T_B and T_D, as defined in Sec. 5.1.

It can be shown that the concentration of Th^{228} after one complete cycle of one buildup period plus one decay period is

$$N_{08}^{1c} = \frac{\lambda_{22}}{\lambda_{08} - \lambda_{22}} N_{22}^{o} (e^{-\lambda_{22}T_B} - e^{-\lambda_{08}T_B}) e^{-\lambda_{08}T_D} \tag{21}$$

where T_D is the decay time. The activity after two such cycles can be shown to be

$$N_{08}^{2c} = \frac{\lambda_{22}}{\lambda_{08} - \lambda_{22}} N_{22}^{o} (e^{-\lambda_{22}T_B} - e^{-\lambda_{08}T_B}) e^{-\lambda_{08}T_D} e^{-\lambda_{08}T_B}$$

$$+ (e^{-\lambda_{22}T_B} - e^{-\lambda_{08}T_B}) e^{-\lambda_{08}T_D} \tag{22}$$

or the ratio of activity after two cycles to the activity after one cycle is

$$\frac{N_{08}^{2c}}{N_{08}^{1c}} = 1 + e^{-\lambda_{08}T_D} e^{-\lambda_{08}T_B} \tag{23}$$

For convenience K will be used to represent the product of the exponentials, $e^{-\lambda_{08}T_D} e^{-\lambda_{08}T_B}$. Then

$$\frac{N_{08}^{2c}}{N_{08}^{1c}} = 1 + K \tag{24}$$

$$\frac{N_{08}^{3c}}{N_{08}^{1c}} = (K+1)K + 1 = K^2 + K + 1 \tag{25}$$

$$\frac{N_{08}^{4c}}{N_{08}^{1c}} = \left[(K + 1)\ K + 1\right] K + 1 = K^3 + K^2 + K + 1 \tag{26}$$

and, for n cycles,

$$\frac{N_{08}^{nc}}{N_{08}^{1c}} = K^{n-1} + K^{n-2} + \cdot\ \cdot\ \cdot\ K + 1 = \sum_{m=1}^{n} K^{m-1} \tag{27}$$

If n is allowed to approach infinity,

$$\frac{N_{08}^{\infty c}}{N_{08}^{1c}} = \sum_{m=1}^{\infty} K^{m-1} = \frac{1}{1 - K} = \frac{1}{1 - e^{-\lambda_{08} T_B}\ e^{-\lambda_{08} T_D}} \tag{28}$$

Fugure 7 is a nomograph which may be used to calculate the activity after infinite recycle when the buildup time and the 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.

5.3 Activity from Thorium Cylinders

In order to indicate the effect of U^{232} daughters on the activity of thorium that is being recycled, the surface activity of (1) a cylinder 10 by 60 cm and (2) of a thorium slug 1.36 by 6 in. will be calculated. The following assumptions are made:

Production = 4000 g of Mass-233 per ton of thorium

Flux = 3×10^{13} n/cm^2/sec

Irradiation period = 270 days

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$T_B = T_C + 1/3\,T_R$; where T_C = COOLING PERIOD
$T_D = T_{PT} + 2/3\,T_R$ T_R = REACTOR PERIOD
T_{PT} = POST THOREX PROCESS PERIOD

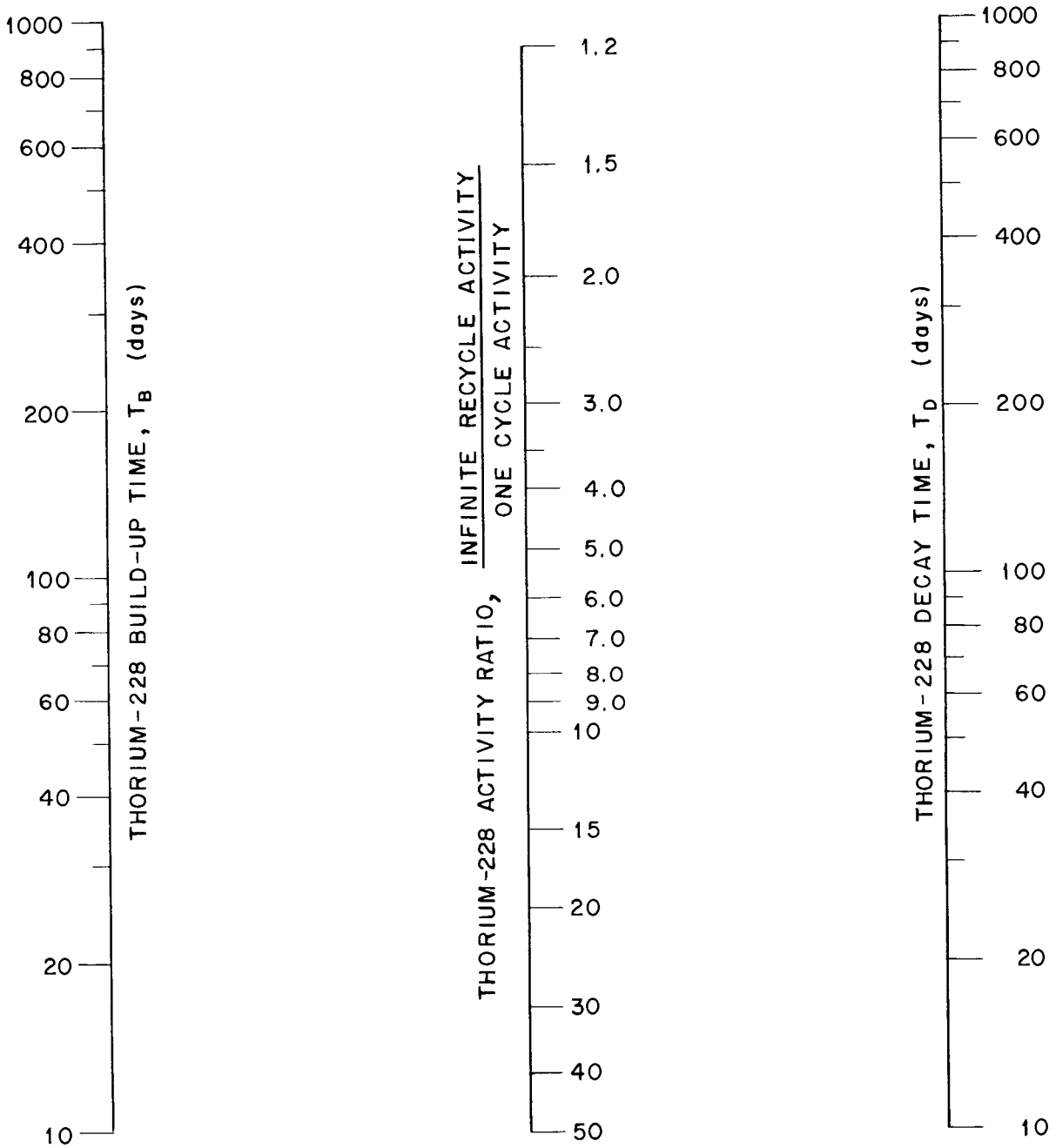

Fig. 7. Nomograph for Computing Infinite Recycle Thorium-228 Activity in Thorium-232.

$\sigma_{02}(n,2n) = 0.003$ barn

Cooling period = 130 days

Post-Thorex period, T_{PT}, = 100 days

Other information which may be found is

$\alpha_{22}/\alpha_{23} = 0.07$ (Fig. 1)

$T_B = T_R/3 + T_C = 220$ days

Th^{228} buildup factor = 0.2 (Fig. 4)

$T_D = 2T_R/3 + T_{PT} = 280$ days

The expected source strengths after the Th^{228} daughters have come to equilibrium (25 days after the Thorex process) are:

$$Bi^{212}\ \gamma = 1.6 \times 10^9 \times 0.4 \times 0.07 \times 0.2 = 8.97 \times 10^6 \text{ d/s/kg Th} \tag{29}$$

$$Tl^{208}\ \gamma \text{ (each)} = 1.16 \times 10^9 \times 0.4 \times 0.07 \times 0.2 = 6.5 \times 10^6 \text{ d/s/kg Th} \tag{30}$$

Similar source strengths for natural thorium would be

$$Bi^{212}\ \gamma = 1.92 \times 10^6 \text{ d/s/kg and } Tl^{208}\ \gamma \text{ (each)} = 1.4 \times 10^6 \text{ d/s/kg Th}$$

The radiation intensities (Sec. 7) are

$$I_o^* = AN_o/4\pi X^3 \text{ (Eq. 9)}$$

$$= \frac{8.97 \times 10^6}{3.7 \times 10^{10}} \times \frac{1.18}{169} \times \frac{1.18 \times 10^7}{4\pi \times 89.3} = 0.0178 \text{ r/hr/cc for } Bi^{212} \text{ for 1 steradian} \tag{31}$$

Similarly,

$$I_o^* = 0.0286 \text{ r/hr/cc for } Tl^{208} \text{ 2.62-Mev } \gamma \text{ for 1 steradian}$$

$$I_o^* = 0.0063 \text{ r/hr/cc for } Tl^{208} \text{ 0.58-Mev } \gamma \text{ for 1 steradian}$$

The intensities on the surface of the 10- by 60-cm cylinder will be

$$I = 4\pi R I_o^* F_p(\mu a) \tag{32}$$

Using the following values[6] for μ (estimated from lead)

$\mu = 0.42$ for 1.18-Mev γ (Bi^{212} average)

$\mu = 0.33$ for 2.62-Mev γ

$\mu = 1.0$ for 0.58-Mev γ

Then

$$I_{Bi212} = 4\pi \times 5 \times 0.0178 \times 0.284 = 0.318 \text{ r/hr} \quad (33)$$

$$I_{1Tl\,208} = 4\pi \times 5 \times 0.0286 \times 0\ 510 = 0.620 \text{ r/hr} \quad (34)$$

$$I_{2Tl\,208} = 4\pi \times 5 \times 0.0063 \times 0.156 = 0.050 \text{ r/hr} \quad (35)$$

Total 0.988 r/hr

Infinite recycle will increase this activity to 2.5 x 0.99 = 2.5 r/hr. The intensities on the surface of a 1.36- by 6-in. thorium slug would be

$$I_{Bi212} = 4\pi \times 1.73 \times 0.0178 \times 0.585 = 0.226 \text{ r/hr} \quad (36)$$

$$I_{1Tl\,208} = 4\pi \times 1.73 \times 0.0286 \times 0.775 = 0.405 \text{ r/hr} \quad (37)$$

$$I_{2Tl\,208} = 4\pi \times 1.73 \times 0.0063 \times 0.405 = 0.045 \text{ r/hr} \quad (38)$$

Total 0.676 r/hr

Infinite recycle will increase this activity to 2.5 x 0.676 = 1.69 r/hr

The expression (Sec. 7) for the intensity produced by a finite cylinder at a given distance from the detector is

$$I = \frac{4\pi I_o * R^2}{2(a+Z)} \mathcal{J}(\theta, \phi) \quad (39)$$

It can be shown that the intensities 5 cm from the midpoint of the surface of the thorium slug are

$$I_{Bi}212 = \frac{4\pi \times 0.0178 \times \overline{1.73}^2}{2(5 + 0.95)} (0.54) = 0.030 \text{ r/hr} \quad (40)$$

$$I_{1Tl}208 = \frac{4\pi \times 0.0286 \times \overline{1.73}^2}{2(5 + 0.90)} (0.63) = 0.058 \text{ r/hr} \quad (41)$$

$$I_{2Tl}208 = \frac{4\pi \times 0.0063 \times \overline{1.73}^2}{2(5 + 0.84)} (0.34) = 0.007 \text{ r/hr} \quad (42)$$

Total 0.095 r/hr

Similarly it may be shown that the total intensity at 1 ft would be 0.0048 r/hr.

The activities for other shapes may be calculated similarly. The activities for other reactor and cooling conditions may easily be estimated from the above calculations since the activity is proportional to the gt level, the α_{22}/α_{23} activity ratio, and the Th^{228} buildup factor. The effect that the gt level and the pre-Thorex cooling period have on the activity of thorium slugs has been indicated in Tables 2 and 3.

5.4 Data on Thorium Activities

An analytical-chemical study of the thorium products from the recent ORNL Thorex Pilot Plant is now underway, but no data have been obtained. The material must go through a series of separation steps in order to remove fission products and Pa^{233}, which might mask the U^{232} daughter activities. The activity of this material will be studied over a period of about 6 months.

Several checks on Fernald thorium, a quarter section of an ingot approximately 1 in. thick, read 11 mr at 5 cm and 0.75 mr at 1 ft. A 2.56- by 15.5-cm slug read 4.5 mr at 5 cm and 0.35 mr at 1 ft. The ingot material had stood 1 yr since metallurgical processing (it is listed on Fernald production order 231). The age of the slug since reduction is unknown. The theoretical readings from a 1.36- by 6-in. natural thorium slug would be 33 mr at 5 cm and 1.6 mr at 1 ft.

6.0 CROSS-SECTIONS

6.1 Theoretical Cross-sections of the (n,2n) Reaction of Th^{232}

The proportion of the total reaction represented by the (n,2n) reaction involved in conversion of Th^{232} to U^{232} (see Sec. 4.1 for reactions) varies with different reactors and even in different regions of the same reactor as a result of the extent of moderation. Because of the high energy necessary for reaction, only a small fraction of neutrons enter into (n,2n) reactions, and this small fraction decreases with increased moderation.

Since the (n,2n) reaction involves fast neutrons, the fast-neutron cross-section of Th^{232} should be known. The cross-section for the (n,2n) reaction, S(E), as a function of incident neutron energy E for a heavy element may be obtained from the equation derived by Weisskoff and Ewing:[8]

$$S(E) = \pi R^2 \left[1 - \left(1 + \frac{\Delta E}{T}\right)e^{-\Delta E/T}\right] \tag{43}$$

where E = energy of the incident neutron minus the binding energy of the last neutron in the target; in the case of Th^{232} that binding energy is 6.37 Mev, which constitutes a threshold for the (n,2n) reaction

R = nuclear radius of the target = $1.5 \times 10^{-13} A^{1/3}$

T = the nuclear temperature defined by $T = 2\left(\frac{5E}{A}\right)^{1/2}$

A = the atomic weight of the target

Equation 43 accounts for the probabilities of forming an excited compound nucleus and for the decay of the compound nucleus by the emission of two neutrons. It must, however, be corrected for the probability of the neutron fission reaction. S(E) must be averaged over the neutron energy distribution inside a slug in order to calculate S(n,2n) for the sample. In this study it was assumed that the thorium sample is placed within a Hanford-type receptacle slug (hollow-center standard uranium slug). It was assumed that only fission spectrum neutrons play an important part in the (n,2n) reaction and the averaging was accomplished by using the following equation[9] for the energy distribution of fission spectrum neutrons:

$$N(E) = 1.058 \times 10^3 e^{-E/0.965} \sinh \sqrt{2.29E} \qquad (44)$$

where N(E) is the number of neutrons of energy E. The fast neutron cross-section is then calculated:

$$S(n,2n + \text{fission}) = 0.96 \text{ b}$$

In order to determine the average fission cross-section of Th^{232} for pile neutrons of energy higher than 6.37 Mev, the curve of $\sigma_{02}(n,f)$ vs. neutron energy given by Barschall and Henkel[10] is weighted by means of Eq. 44 to give

$$\overline{\sigma}_{02}(n,f) = 0.42 \text{ b}$$

Then $S(n,2n) = 0.96 - 0.42 = 0.54$ b.

The calculated cross-section differs from the published values in that the published values are expressed in terms of thermal flux. In order to compare the two, the ratio of the mean flux of neutrons with energy above 6.37 Mev to the mean thermal flux must be determined. The best available data for the flux in the Hanford pile[11] give the ratio

$$\frac{\text{Mean flux above 3.5 Mev}}{\text{Mean thermal flux}} = 0.14$$

for the section of a Hanford slug between 0.39 in. and 0.55 in. from the center. To determine the mean flux of neutrons with energy above 6.37 Mev, Eq. 44 and numerical integration are used to get the ratio of the flux above 6.37 to the flux above 3.5 Mev. This ratio is 0.12, and thus the ratio of mean neutron flux above 6.37 Mev to the mean flux is $0.14 \times 0.12 = 0.0168$. The theoretical value of the $Th^{232}(n,2n)$ cross-section, in terms of the thermal flux within the receptacle slug, is 0.0091 barn:

$$\sigma(n,2n) = S(n,2n) \times 0.0168 = 0.54 \times 0.0168 = 0.0091 \text{ b}$$

The experimental value for the (n,2n) cross-section of Th^{232} based on the known $U^{238}(n,2n)$ cross-section in a Hanford receptacle slug has been published as 0.009 ± 0.004 b in terms of the thermal flux.[12]

The $Th^{232}(n,2n)$ cross-section in a Hanford process tube would be much less than the calculated value for a receptacle slug. The curves of Gast[13] indicate that there is about a factor of 5 less neutrons above the $Th^{232}(n,2n)$ threshold in a thorium slug located in a Hanford process channel than inside a receptacle slug. There is also an increase by a factor of about 2 in the thermal flux in a thorium slug over that inside a receptacle slug. These two factors reduce the cross-section for the (n,2n) reaction in a thorium-loaded process channel to approximately 0.001 barn.

6.2 Effects of Different Reactor Loadings and Geometries on Cross-sections

The effective thermal cross-section for the (n,2n) reaction for other reactors and loadings would have to be estimated in a similar manner. The effective (n,2n) thermal cross-section for alternate thorium and J slugs in a Hanford channel has been experimentally determined as 4.3 mb.[14] From the above discussion it should be expected that this cross-section value would be somewhat between the receptacle case and the separate channel case because of end effects. If the depth of the end effects is equivalent to a slug radius on each end, then the apparent cross-section would be 2.9 mb; if the depth of these end effects approaches a slug diameter, then the apparent cross-section would be 4.6 mb.

Determination[14] of the (n,2n) cross-section in the ORNL Low Intensity Test Reactor indicates that:

1. A thorium slug arrangement, in a beryllium reflector, 1-2 in. from the active lattice and within a few inches of the vertical center gave an effective cross-section of 9 ± 3 mb.
2. A thorium slug arrangement, identical radial position to the above but positioned near bottom and top of reactor, gave an effective cross-section of 6 ± 2 mb.

For the case of Savannah River loadings the following probable cross-sections may be expected:

1. With an arrangement of two channels of thorium slugs and two of U^{235}-

aluminum alloy slugs in each quatrefoil assembly—somewhat similar to alternate thorium and J slugs at Hanford and also similar to the ORNL LITR cases—a probable value for the (n,2n) cross-section would be 5 to 10 mb.[14]

2. With an arrangement of thorium slugs surrounded by U^{235}-aluminum alloy cylinders or shells—this scheme would be similar to a Hanford receptacle slug—an (n,2n) cross-section of 9 mb should be probable.

3. With an arrangement of thorium shells outside and surrounding U^{235}-aluminum alloy slugs, a cross-section somewhat higher than 10 mb would be expected since essentially all the fast neutrons would have to go through the thorium to reach the moderator; an (n,2n) cross-section of 20 mb would be the upper limit for this scheme.[14] The true value of the (n,2n) cross-section would depend upon the thorium shell thickness and would be less than 20 mb for comparatively thin shells.

6.3 Effect of Buildup of U^{233} in a Thorium Slug

The theoretical (n,2n) cross-section of 9 mb for Th^{232} based on thermal flux was calculated for a Hanford natural uranium receptacle slug which is 0.71% U^{235}. The equivalent level of U^{233} in thorium to give the same fission rate within a slug, if uniform distribution of U^{233} throughout the slug is assumed, would be 7.5 g of U^{233} per kilogram of thorium. Such a condition would create the same fast flux level within the thorium slug as that experienced in the receptacle slug, and the thermal flux depressions should be comparable in the two cases. It therefore should be safe to assume that the (n,2n) cross-section in thorium placed in a Hanford process tube would become 9 mb when the N_{23}/N_{02} ratio is 7.5×10^{-3}. This assumption also requires that U^{233} have the same fission spectrum as U^{235}. The effective cross-section for lower N_{23}/N_{02} levels would be between 1 and 9 mb and should be proportional to the actual N_{23}/N_{02} ratio. The average (n,2n) cross-section during an irradiation should be proportional to the average N_{23}/N_{02} ratio during the irradiation period. An approximation that may be assumed for the average (n,2n) cross-section is

$$\sigma(n,2n) = 1 + \frac{N_{23}^{f}/2N_{02}}{7.5 \times 10^{-3}} \times 8 \text{ mb} \qquad (45)$$

The average (n,2n) cross-section for an irradiation in which the final U^{233} concentration (N_{23}^{f}) is 3 atoms per 1000 atoms of thorium would be

$$\sigma(n,2n) = 1 + \frac{3.0 \times 10^{-3}/2}{7.5 \times 10^{-3}} \times 8 = 2.6 \text{ mb} \qquad (46)$$

The increase in the (n,2n) cross-section for a thorium irradiation using a different type of loading would be somewhat similar, although possibly not of the same magnitude. The increase in this case would depend on the percentage increase of fast flux by U^{233} fissioning over the existing fast flux produced by the U^{235} elements in proximity of the thorium.

6.4 Effects of Other Paths for U^{232} Formation

Uranium^{232} may also be formed by the reactions

$$Pa^{233}(n,2n)Pa^{232} \xrightarrow[1.32d]{\beta^-} U^{232} \text{ and } U^{232}(n,2n)U^{232}$$

The (n,2n) cross-sections for these reactions may be assumed to be equal to the cross-section of the (n,2n) reaction on Th^{232}. The ratio of the U^{232} formed by the sum of these two paths to that formed by the $Th^{232}(n,2n)$ path may be estimated to be $\sim 2.5 \times 10^{-2}$ for a final Mass-233 concentration of 4000 g per ton of thorium.

7.0 RADIATION HAZARD CALCULATIONS

In order to calculate the intensity of radiation at the surface of and at some distances from various shapes, a few basic formulas and definitions should be understood.[2] To keep the system dimensionally consistent, it will be convenient to define four types of I_o, the intensity at a unit distance from the source. If a unit of the source emits N_o photons per second, then where A is a constant of proportionality and X a linear dimension,

$I_o = AN_o/4\pi$ = intensity at a unit distance from a point source

$I_o' = AN_o/4\pi X$ = intensity at a unit distance from a unit length of a line source.

$I_o'' = AN_o/4\pi X^2$ = intensity at a unit distance from a unit area of a plane source

$I_o^* = AN_o/4\pi X^3$ = intensity at a unit distance from a unit volume of a solid source.

When the depth of the source, as viewed by the receptor, is small in comparison to the mean free path of the radiation in the source, self-absorption is negligible and the following equations may be used for shapes that are of interest in the thorium-U^{233} program:

Point source, receptor at distance a,

$$I = I_o/a^2$$

Spherical source of radius R with receptor at surface,

$$I = 2\pi I_o^* R$$

$$\frac{\text{Dosage at surface of a homogeneous sphere}}{\text{Dosage from point source at center}} = \frac{2\pi I_o^* R}{\frac{4}{3}\pi R^3 \frac{I_o^*}{R^2}} = \frac{3}{2}$$

Infinitely long cyclinder, radius R, with receptor at surface,

$$I = 4\pi R I_o^*$$

When self-absorption within a source is considered the expression becomes:

Spherical source of radius R with receptor at surface,

$$I = \frac{\pi I_o^*}{\mu^2 R} \left(2\mu R + e^{-2\mu R} - 1\right)$$

$$I = \frac{2\pi I_o^*}{\mu} \quad \text{for a very large sphere}$$

When the receptor is located a few diameters (X cm) from the surface of the sphere,

$$I \approx \frac{I_o^* \pi R^2}{\mu X^2}$$

Other expressions may be used for volume sources with self-absorption. The most desirable, and probably least cumbersome, is the expression for volumes without self-absorption multiplied by a self-absorption coefficient:[15]

$$I_{SA} = I_H F_p(\mu a)$$

where I_{SA} = intensity of radiation with self-absorption

I_H = intensity of radiation assuming a homogeneous volume source

$F_p(\mu a)$ = self-absorption coefficient

$$F_p(\mu a) = \frac{\Gamma(p + 1)}{\Gamma(3/2)\Gamma(p + 1/2)} \int_0^{\pi/2} e^{-2\mu a \cos\theta} \sin^{2p}\theta \, d\theta$$

p = 1 for a cylinder, $\frac{3}{2}$ for a sphere

a = radius of sphere or cylinder

μ = absorption coefficient of source material

Values for self-absorption coefficients for both a cylinder and a sphere as a function of μa are given in Table 9.[15]

The expression for intensity at the surface of a self-absorbing cylinder would then be

Table 9

Self-Absorption Coefficients for Cylinder and Sphere

	F_p (μa)	
μa	Cylinder	Sphere
0	1.0	1.0
0.25	0.810	0.830
0.50	0.675	0.700
0.75	0.575	0.605
1.00	0.490	0.525
1.25	0.425	0.460
1.50	0.370	0.410
1.75	0.330	0.367
2.00	0.295	0.330
2.50	0.242	0.275
3.00	0.205	0.235
3.50	0.177	0.205
4.00	0.156	0.183
4.50	0.140	0.163
5.00	0.125	0.145

$$I = 4\pi R I_o * F_p(\mu a)$$

Another expression for intensity at the surface of a self-absorbing sphere is

$$I = 2\pi R I_o * F_p(\mu a)$$

An expression which may be used for the intensity produced by a finite cylinder (see Fig. 8) at a distance a from the surface is

$$I = \frac{4\pi I_o * R^2}{2(a + Z)} \mathcal{F}(\theta, b)$$

where $b = \mu Z = \frac{1}{2}\mu R$ for values of $\mu R \leq 3$

$$\mathcal{F}(\theta, b) = \int_0^{\theta} e^{-b \sec \theta} \, d\theta$$

Z = effective distance due to self-absorption

θ = angle subtended by Z line of one half of cylinder at point A

Numerical values for constants are given in references 2, 6, and 15.

The following numerical constants may be used to determine the magnitude of radiation:

1 watt = 1.18×10^7 r/hr at 1 cm = 169/E curies = 6.24×10^{12} Mev/sec

where E = energy of particle or photon in mega electron volts.

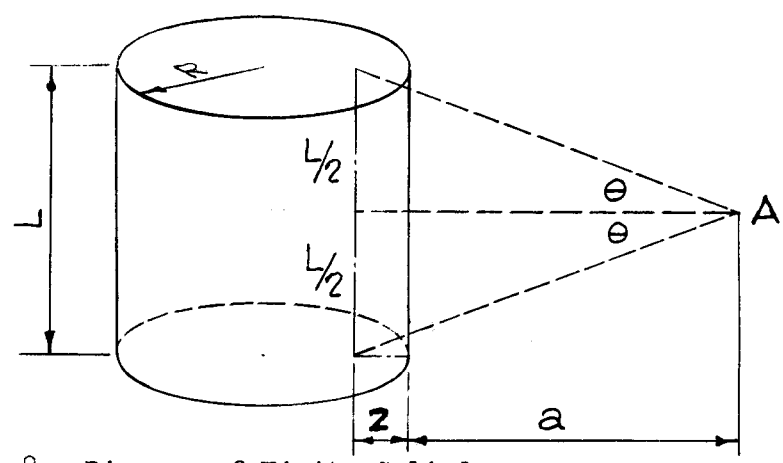

Fig. 8. Diagram of Finite Cylinder.

8.0 COOLING PERIODS AND ACTIVITY CONSIDERATIONS

Short pre-Thorex cooling periods will be necessary in order to limit the Th^{228} chain activities in the thorium product. These short cooling periods (30 - 60 days) will create other activity problems while reducing the Th^{228} chain activity. The Pa^{233} beta activity will be the most important contributor to solvent irradiation. Fission products and other activities will contribute less than 10% additional activity. The activity of Th^{234}-Pa^{234} will be important from the standpoint of direct metallurgical reprocessing of the thorium product and will add the requirement of a long post-Thorex cooling period for the thorium before it is recycled.

8.1 Activity of Pa^{233} and Fission Products

The buildup and decay of Pa^{233} (27.4 days) in thorium irradiations at thermal flux values of 10^{13}, 10^{14}, and 10^{15} n/cm^2/sec have been studied separately.(3) The effect of flux and extent of irradiation on the Pa/U^{233} ratio is shown in Fig. 9 and the effect of extent of irradiation and cooling time in Fig. 10. From the figures it is possible to determine the Pa^{233} activity for any irradiation level and cooling time. For example, with a flux of 3×10^{13}, material containing 4000 g of Mass-233 per ton of thorium, and 130 days cooling, Pa^{233} will be 20% of the total Mass-233 at the time of reactor discharge (Fig. 9), and decay over 130 days will reduce this Pa^{233} content to 0.8% of the total Mass-233 (Fig. 10). The Pa^{233} activity of this material can then be calculated:

$$\frac{4 \text{ g Mass-233} \times 0.008}{1000 \text{ g Th} \times 233} \times \frac{6.023 \times 10^{23} \times 0.693}{8.64 \times 10^{4} \times 27.4} = 2.42 \times 10^{10} \text{ d/s/g Th}$$

The activity in beta-watts per kilogram of thorium due to Pa^{233} would be

$$\frac{4 \text{ g Mass-233}}{\text{kg Th}} \times 0.008 \times 19 \frac{\beta\text{-watts}}{\text{g } Pa^{233}} = 0.608 \frac{\beta\text{-watts}}{\text{kg Th}}$$

On the other hand, the Pa^{233} activity after 60 days' cooling would be

Dwg. 23354 [redacted]

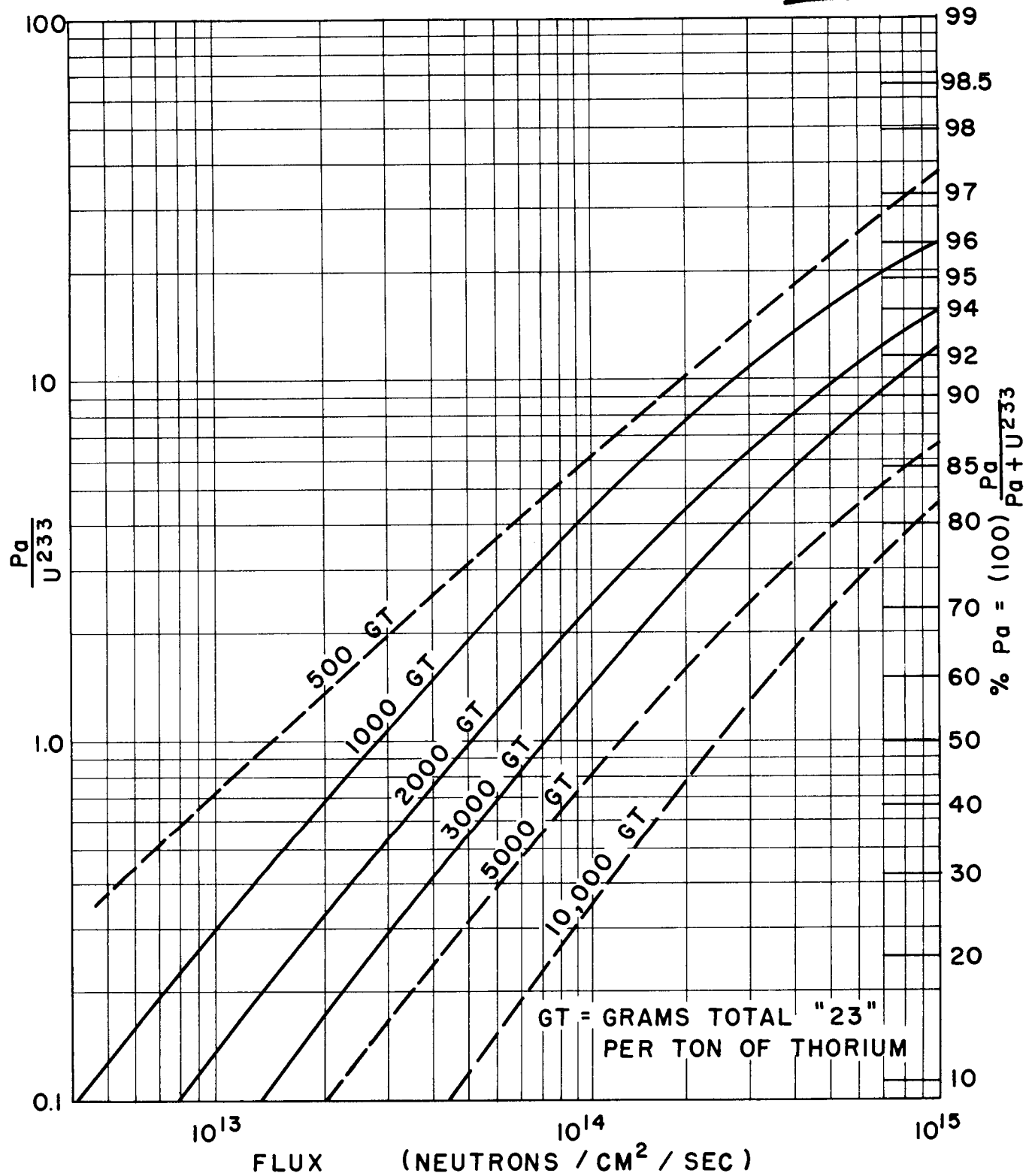

FIGURE 9

EFFECT OF FLUX AND EXTENT OF IRRADIATION ON Pa TO U^{233} RATIO

Dwg. 23355 [redacted]

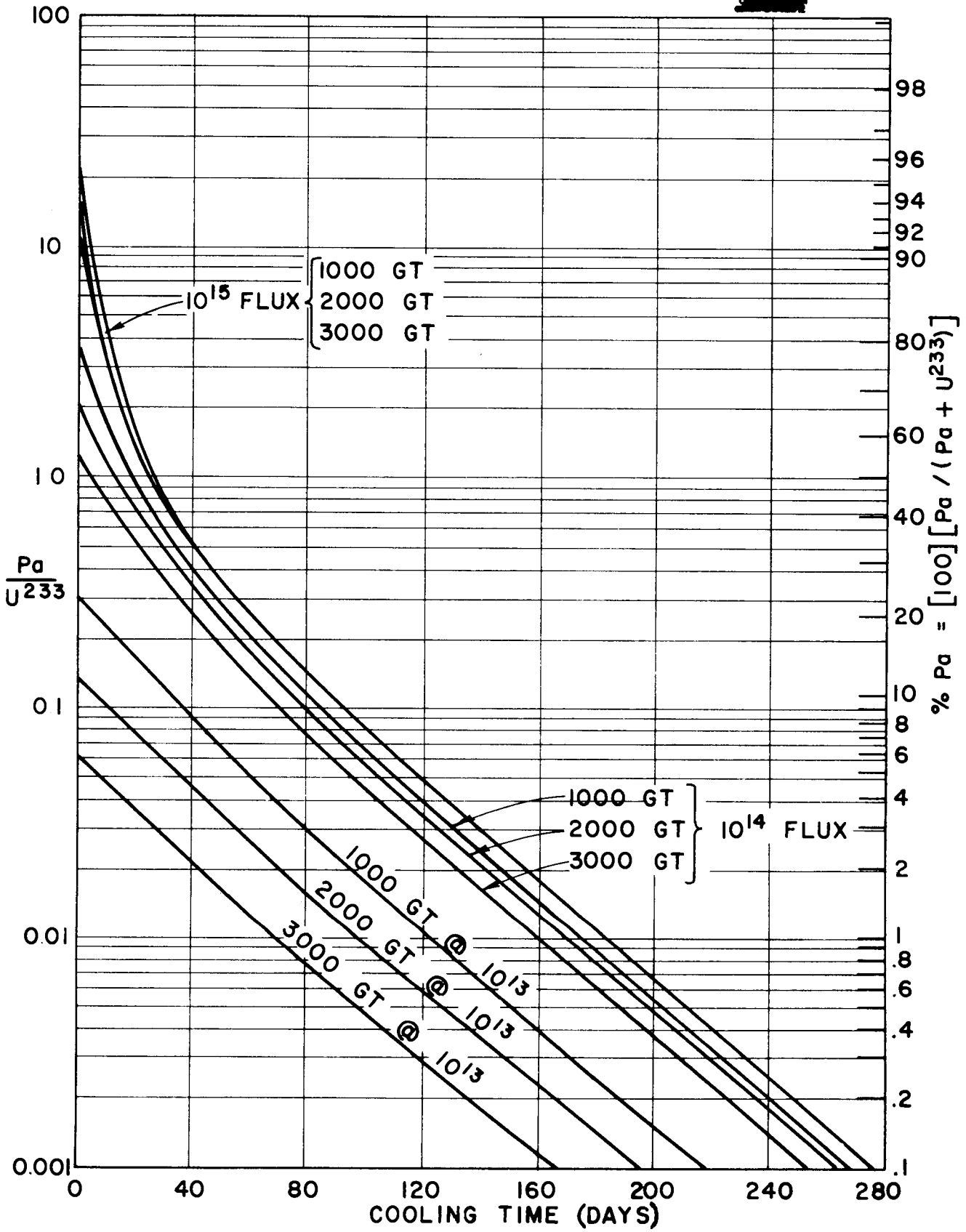

FIGURE 10
EFFECT OF EXTENT OF IRRADIATION
AND COOLING TIME ON Pa TO U^{233} RATIO

$$\frac{4 \text{ g Mass-233}}{\text{kg Th}} \times 0.045 \times 19 \frac{\beta\text{-watts}}{\text{g Pa}^{233}} = 3.42 \ \beta\text{-watts/kg Th}$$

and after 30 days

$$Pa^{233} \ \beta \text{ activity} = 6.8 \text{ watts/kg Th}$$

It can be estimated(3) that the at-time-of-discharge mass ratio of fission products to Pa^{233} for 4000-gt material produced at a flux of 3×10^{13} n/cm^2/sec will be 0.59. Using an average specific power of 3 β-watts per gram of mixed fission products, it may be shown that the fission product beta activity level will be a factor of 11 below that of Pa^{233}. The fission product activity will not be an important factor in Thorex processing after short cooling periods except that shielding must be provided for the La^{140} activity.

8.2 Activity of Th^{234} and Pa^{234}

The buildup of Th^{234} can be shown(3) to follow the following equation:

$$N_{04} = \frac{N_{02}\sigma_{02}\sigma_{03}\phi^2}{\lambda_{04}\lambda_{03}^2} \left[\lambda_{03}(1 - e^{-\lambda_{04}t}) - \lambda_{04} \right]$$

The buildup of Th^{234} as a function of flux and extent of irradiation is shown in Fig. 11. The decay of Th^{234} is shown in Fig. 12, which does not show all decay lines but the initial Th^{234} activity may be interpreted from the activity axis of Fig. 11 or from the relation indicated in Fig. 11 that 5.78×10^{-9} moles of Th^{234} per mole of Th^{232} corresponds to an activity of 3×10^{11} d/m/kg Th.

The activity of Th^{234} after irradiation of the thorium metal at a flux of 3×10^{13} n/cm^2/sec to 4000 g of Mass-233 per ton and a 130-day pre-Thorex cooling period would be 6×10^{10} d/m/kg Th. Since the half-life of Pa^{234} is short, it may be assumed that Pa^{234} is in equilibrium with Th^{234} and the total activity of $Th^{234} + Pa^{234}$ will be 1.2×10^{11} d/m/kg Th. These activities may be compared with the Pb^{212}, Bi^{212}, and Tl^{208} activities indicated in Sec. 5.3, which were:

Bi^{212} $\beta = 7.6 \times 10^8$ d/m/kg Th, Tl^{208} $\beta = 3.9 \times 10^8$ d/m/kg Th and

Pb^{212} $\beta = 1.15 \times 10^9$ d/m/kg Th

ORNL-LR-DWG 1665 [redacted]

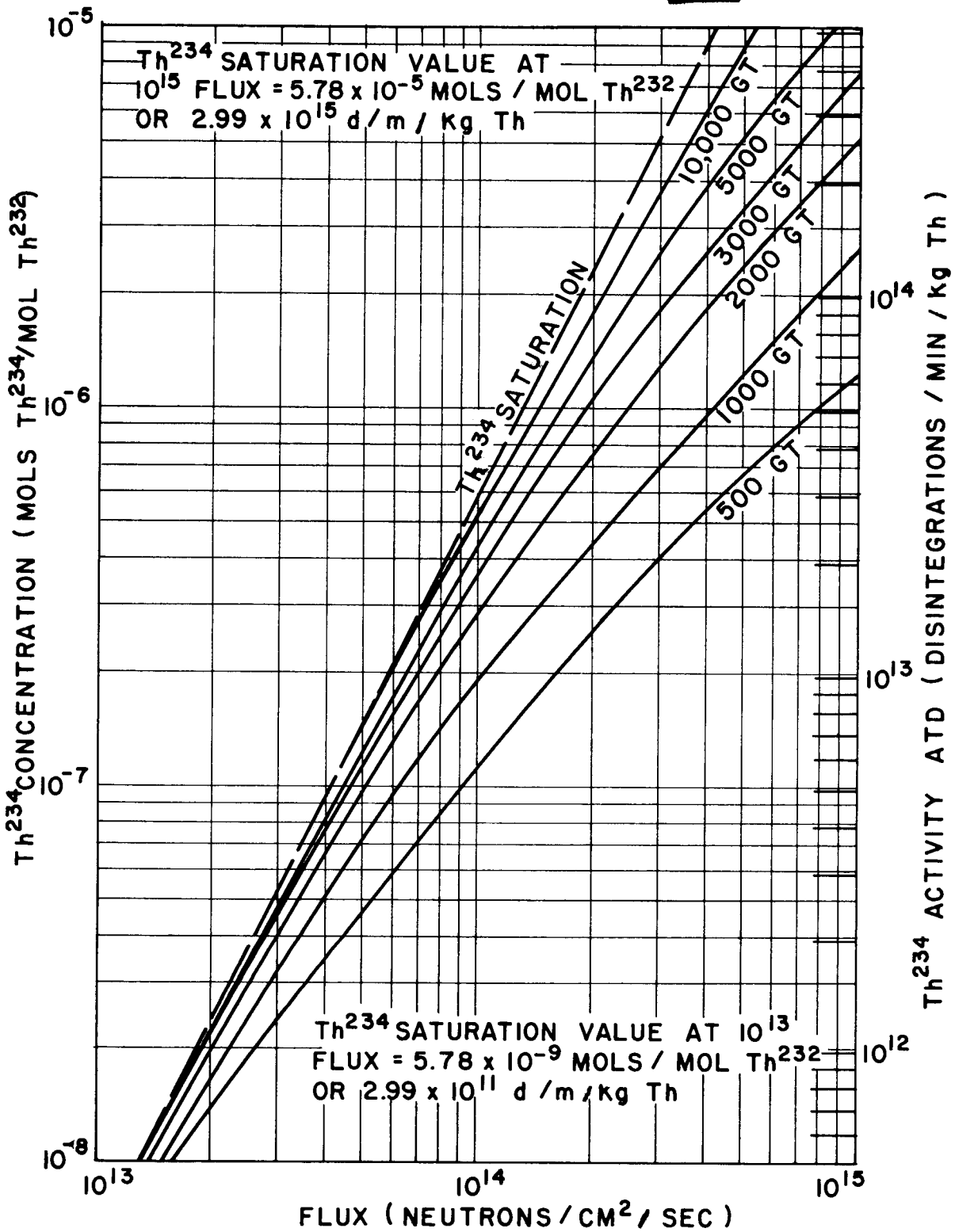

FIGURE 11

EFFECT OF FLUX ON Th^{234} CONCENTRATION AND ACTIVITY

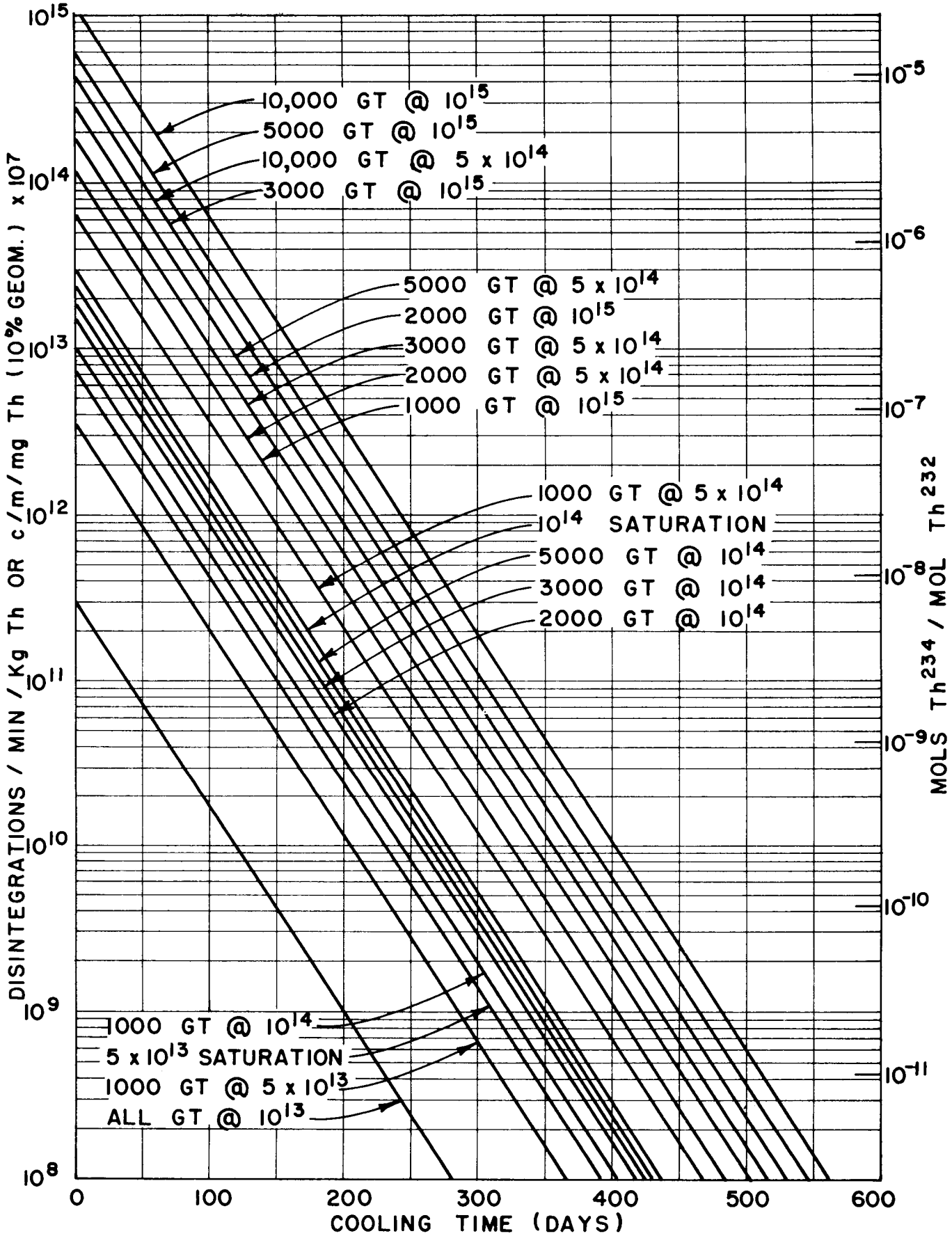

FIGURE 12
Th^{234} DECAY

The limiting activity for direct metallurgical processing would be 10^9 d/m/kg Th, which indicates that the thorium product would need a total cooling time of 290 days to reduce Th^{234} to this level. A 160-day post-Thorex cooling period would be required for the thorium product if a 130-day pre-Thorex cooling period is used.

The β activities contributed by the Th^{228} chain are greater than 5.0×10^8 d/m/kg Th for all U^{233} concentrations above 2000 g per ton of thorium, and for pre-Thorex cooling periods greater than 130 days at U^{233} concentrations of 2000 g per ton of thorium. This indicates that the Th^{228} chain activities will become controlling after several recycles even if the total thorium cooling time (pre-Thorex plus post-Thorex) is taken to reduce Th^{234} + Pa^{234} activities to 10^9 d/m/kg Th. The β activities of the Th^{228} chain and Th^{234} + Pa^{234} and the total β activity from these two major sources are shown in Table 10 as a function of irradiation level and pre-Thorex cooling time.

Table 10

Activities in Thorium Metal

Component	gt Level g U^{233} per Ton of Thorium	Beta Activity in Thorium Metal (d/m/kg Th)			
		60 days Pre-Thorex Cooling	130 days Pre-Thorex Cooling	200 days Pre-Thorex Cooling	300 days Pre-Thorex Cooling
Th^{228} chain[a]	1000	5.8×10^{7}	1.1×10^{8}	1.5×10^{8}	2.0×10^{8}
$Th^{234} + Pa^{234}$		7.2×10^{11}	8.6×10^{10}	1.14×10^{10}	6.4×10^{8}
Total		7.2×10^{11}	8.6×10^{10}	1.16×10^{9}	8.4×10^{8}
Th^{228} chain[a]	2000	2.8×10^{8}	4.6×10^{8}	6.2×10^{8}	8.3×10^{8}
$Th^{234} + Pa^{234}$		9.0×10^{11}	1.08×10^{10}	1.44×10^{10}	8.0×10^{8}
Total		9.0×10^{11}	1.13×10^{10}	1.5×10^{10}	1.63×10^{9}
Th^{228} chain[a]	3000	7.8×10^{8}	1.17×10^{9}	1.51×10^{9}	2.0×10^{9}
$Th^{234} + Pa^{234}$		9.6×10^{11}	1.16×10^{11}	1.54×10^{10}	8.8×10^{8}
Total		9.6×10^{11}	1.17×10^{11}	1.69×10^{10}	2.88×10^{9}
Th^{228} chain[a]	4000	1.6×10^{9}	2.3×10^{9}	2.9×10^{9}	3.7×10^{9}
$Th^{234} + Pa^{234}$		1.0×10^{12}	1.2×10^{11}	1.6×10^{10}	9.0×10^{8}
Total		1.0×10^{12}	1.22×10^{11}	1.9×10^{10}	4.6×10^{9}

[a] Includes β activities of Pb^{212}, Bi^{212} and Tl^{208}.

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