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SUBJECT: Californium Production in the High Flux Isotope Reactor

TO: Distribution

FROM: H. C. Claiborne and Marjorie P. Lietzke

ABSTRACT

Californium-252 and other isotope production rates resulting from irradiating Pu-242 in the proposed High Flux Isotope Reactor were estimated. The effect of curium recycle was investigated and optimum irradiation cycles determined. Approximately 60 milligrams of Cf-252 would be available from the HFIR installation about 1-1/4 years after exposure of 200 g of Pu-242 is started. The ultimate "steady-state" production capability is about one gram/yr of Cf-252.

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INTRODUCTION

As part of the U. S. program in the construction of ultra high-flux reactors (1 to 3 x 10^{15} neutrons/cm^2-sec), the Laboratory is designing a flux-trap reactor for the production of transplutonic elements. This reactor has been designated as the High Flux Isotope Reactor (HFIR). A prime purpose of this reactor is the production of milligram quantities of Cf-252; this isotope will be produced from successive thermal and epithermal neutron captures in Pu-242 placed initially in the central moderating region (flux trap) of the HFIR.

The main purpose of this study was to estimate the production rates of Cf-252 from thermal neutron capture alone. Since ten successive neutron captures (which may proceed along several paths) are required to produce Cf-252 from Pu-242, hand calculations are impractical. A method adaptable to automatic computing machinery and capable of handling all reaction possibilities was set up and programmed for the IBM-704 computer.

In addition to Cf-252 production rates, results are presented for heat generation rates and average cross section of the target material as a function of time; recycle irradiation times which would maximize the over-all production of Cf-252 were also considered.

METHOD OF CALCULATION

The reaction paths involved in the production of Cf-252 by successive neutron capture using Pu-242 as the starting material are shown in Fig. 1. The primary production path is indicated by the solid arrows and the secondary paths by the dashed arrows.

In theory, it is possible to develop the analytical solution for the entire scheme (with all branch chains) when the neutron flux is invariant with time since the mathematical system consists of first-order linear differential equations.

The algebraic manipulations, however, rapidly become too ponderous to handle as the number of isotopes increases. It is simpler to compute the rate of Cf-252 production as the sum of the rates of production from all the various paths as shown. Using the general equations developed in this manner, it is also possible to include α-decay by treating it as a β-decay which forms a fictitious chain that repeats itself. This will be discussed later.

Consider only what might be called a straight chain, i.e., no branching; propagation of the chain is by β-decay or neutron capture, but not by both simultaneously. The general equation representing any isotope as a function of time for constant flux is

$$\frac{d\,N_i}{dt} = p_i\,N_{i-1} - r_i\,N_i \qquad (1)$$

where p_i represents a production cross section that can be either λ_{i-1} or $\sigma_{c_{i-1}}\,\phi$, and r_i a removal cross section that can be any part or all of $\lambda_i + \sigma_{c_i}\,\phi + \sigma_{f_i}\,\phi$ depending on the particular isotope. For the lowest number starting material (Pu-242 in our case) when $i = 1$, $p_i = 0$. In solving equation (1) by Laplace transforms for $i = 1, 2, \ldots i$, it becomes quickly evident that the transform of N_i is

$$\overline{N}_i = \frac{p_2\,p_3\,p_4 \cdots p_i\,N_{o1}}{(s+r_1)(s+r_2)\ldots(s+r_i)} + \frac{p_3\,p_4 \cdots p_i\,N_{o2}}{(s+r_2)(s+r_3)\ldots(s+r_i)} + \cdots \frac{N_{oi}}{s+r_i} \qquad (2)$$

where N_{oi} is the initial amount of the ith isotope and s is the transform variable.

The transform equation (2) may be inverted easily by the partial fraction technique which results in

$$N_i = p_2\, p_3 \cdots p_i\, N_{o1} \left[\frac{e^{-r_1 t}}{(r_2 - r_1)(r_3 - r_1) \ldots (r_i - r_1)} \right.$$

$$\left. + \frac{e^{-r_2 t}}{(r_1 - r_2)(r_3 - r_2) \ldots (r_1 - r_2)} + \ldots \frac{e^{-r_i t}}{(r_1 - r_i)(r_2 - r_i) \ldots (r_{i-1} - r_i)} \right]$$

$$+ p_3\, p_4 \cdots p_i\, N_{o2} \left[\frac{e^{-r_2 t}}{(r_3 - r_2)(r_4 - r_2) \ldots (r_i - r_2)} \right.$$

$$\left. + \frac{e^{-r_3 t}}{(r_2 - r_3)(r_4 - r_3) \ldots (r_i - r_3)} + \ldots \frac{e^{-r_i t}}{(r_2 - r_i)(r_3 - r_i) \ldots (r_{i-1} - r_i)} \right]$$

$$+ \ldots N_{oi}\; e^{-r_i t} \tag{3}$$

Using equation (3), it is possible to calculate the amount of each isotope in any nonbranching chain as a function of time. Equation (3) is, of course, restricted to the case of no repeated roots, i.e., $r_i \neq r_j$. Equation (3) may be written in the following succinct manner, which is a convenient form for the IBM 704 computer.

$$N_I = \sum_{k=1}^{I} \left\{ \sum_{i=k}^{I} \left[N_{ok}\, e^{-r_i t} \prod_{q=k}^{I} \frac{p_q}{(r_q - r_i)} \right] \right\} \tag{4}$$

where

$$r_i = \lambda_i + (\sigma_{ci} + \sigma_{fi})\, \phi$$

$$p_{i+1} = A_i\, \lambda_i + B_i\, \sigma_{ci}\, \phi$$

$$A = 1 \text{ or } 0,\; B = 1 \text{ or } 0$$

and λ, σ_c and σ_f are the decay constant, radiative capture cross section and fission cross section, respectively.

The restricting conditions are:

$$p_q = 1 \quad \text{when} \quad q = k$$

$$\text{and } r_q - r_i = 1 \quad \text{when} \quad q = i$$

$$\text{when } A = 0,\ B = 1 \quad \text{and vice versa.}$$

The average cross section of the material present at any time is

$$\sigma_{av} = \frac{\sum_{i=1}^{I} (\sigma_c + \sigma_f)_i \, N_i}{\sum_{i=1}^{I} N_i} \tag{5}$$

and the heat generation rate in kilowatts is

$$P = \frac{\sum_{i=1}^{I} \sigma_{fi} \, N_i \, \phi}{3.38 \times 10^{13}} \tag{6}$$

The calculations required to survey Cf-252 production as a function of various parameters were much too lengthy to do with desk computers. Equations (4) through (6) were programmed for the IBM-704. Since the program is general in nature and not confined to calculating Cf-252 production, a memorandum describing the code will be written so that it will be available for general distribution.

NUCLEAR CONSTANTS

The thermal cross sections for the transplutonic elements are not well known; consequently, some differences of opinion exist as to what constitutes the best cross section for a particular isotope. The values shown in Table 1 are a self-consistent set based on experimental data (exceptions are noted) and can be considered as good as any other self-consistent set at this date.[1]

PRODUCTION PATHS

In calculating the production of Cf-252 from Pu-242, equation (3) or (4) should be applied separately to the main path (solid arrows, Fig. 1) and to each branch path (dashed arrows). The effect of an α-decay of Cf-252 (other α-decay effects are definitely negligible) can be accounted for in theory by treating the α-decay as a β decay which forms a fictitious chain that repeats itself; i.e., the Cf-252 decays to a branch chain (starting with Cm-248) that generates Cf-252 which in turn decays to a similar branch chain, and so on until the desired degree of accuracy is attained. The sum of the contribution from each chain is the total Cf-252 production.

In using equation (3) or (4) to calculate the effect of α-decay, it is necessary to change by a small amount the removal cross sections of Cm-248 through Cf-252 in each cycle in order to avoid repeated roots. If the changes in removal cross sections are kept small enough to have negligible effect, trouble develops from loss of significance in the IBM-704 calculations (8+ significant figures are normally carried). The same trouble also occurred for small neutron exposures along the other paths. By programming the equations in double precision arithmetic (16+ significant figures are carried), this trouble was avoided.

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[1] A. Chetham-Strode, Private Communication.

Table 1

Nuclear Cross Sections and Decay Constants

Isotope	σ_c, barns	σ_f, barns	Decay half life, time units
Pu-242	52	$\leq$ 0.3	very large (α)
Pu-243	170[a]	0	4.98 hr (β)
Am-243	138	0	very large (α)
Cm-244	3000[b]	0	26 min. (β)
Cm-244	25	0	18.4 yr (α)
Cm-245	200	1500	very large (α)
Cm-246	15	0	very large (α,s.f.)
Cm-247	200	200	very large (α)
Cm-248	2	0	very large (α, s.f.)
Cm-249	3000[b]	0	50 min. (β)
Bk-249	1100	0	290 days (β)
Bk-250	5000[b,c]	0	3.1 hr (β)
Cf-249	270	630	very large (α)
Cf-250	1500	0	10 yr (α)
Cf-251	3000	3000	very large (α)
Cf-252	30	0	< 2.2 yr (α) < 66 hr (s.f.)

[a] Trilinear Chart of Nuclides, W. H. Sullivan

[b] Guesses from Contour Diagram of J. E. Evans (IDO-16163)

[c] Actually zero was always used since all captures lead to short-lived isotopes of Bk that decay to californium.

The effect of all the branch paths has not yet been calculated. It is possible to show that the difference between maximum possible and minimum possible Cf-252 production is only 15 to 25% in the range of interest, which is good enough for present purposes.

The maximum production scheme allowed no branch-chain losses. It was assumed that no radiative capture occurred in Pu-243, Am-244, Cm-249 and Bk-250 and that Cm-244, Bk-249, Cf-250 and Cf-252 did not decay. The minimum production scheme permitted all these captures and decays to occur (except for Bk-250) but did not consider the recovery of some of the losses by the branch chain contribution to Cf-252 formation, i.e., all branch chain reactions resulted in a complete loss. Captures in Bk-250 were not considered in the minimum production scheme because these captures form isotopes of very short half lives which decay to Cf-251 or Cf-252.

INITIAL IRRADIATION RESULTS

The net cumulative production of Cf-252 from an initial loading of 100 grams of Pu-242 for thermal neutron fluxes (epithermal flux effects are not considered) from 1 to 5×10^{15} is shown in Fig. 2. Production from each flux level is represented by a band with a maximum spread of about 25%. This spread must not be construed as error limits, it merely represents the difference between the maximum production scheme and the minimum production scheme described in the previous section. For the particular set of cross sections used, the actual Cf-252 production must lie between these limits.

On comparing Fig. 2 with Fig. 3, which are previously estimated[1] results, it is apparent that previous estimates of Cf-252 production are considerably lower. It is believed that the results presented in Fig. 2 are on a firmer basis and should be used in all future HFIR planning.

[1] J. A. Lane et al, High Flux Isotope Reactor Preliminary Design Study, CF-59-2-65 (March 20, 1959).

Californium-252 production is shown plotted against neutron exposure (flux x time) in Fig. 4A and 4B. It is obvious that deviations from the smooth curve drawn through the points are minor and that the net cumulative production may be considered a function of neutron exposure alone with no appreciable error and, consequently, Cf-252 production would be independent of flux level for a given neutron exposure. This is not surprising in view of the relatively small half lives of all unstable isotopes in the chain. The above result suggests that all unstable isotopes may be eliminated from consideration and neutron captures that normally produce unstable isotopes can be considered to produce instantaneously the next higher isobar. Some check calculations in the range of interest showed that this slightly simplified scheme gave results that were within 10%.

Heat generation in the fertile or target material as a function of time is shown in Fig. 5. For each 100 grams of Pu-242 exposed to a flux of 3×10^{15}, the heat generation rate rapidly rises to a maximum of 242 kw at 0.4 hr. This peaking results from fission of Cm-245 which also reaches a maximum amount at 0.4 yr (see Fig. 6B). Thus, for a 200-250 gram initial loading of Pu-242, the peak heat generation rate is about 0.5 megawatt, a rather large value for the small allowable volume of the target material. The magnitude of the heat generation rate resulting from reactions initiated with pure Pu-242 is sufficiently high to dictate the allowable limit of Pu-241 in the target material. Pu-241 has a fission cross section of 1100 barns, so even a small percentage will cause a high initial heat generation rate. If it is assumed that the peak rate indicated in Fig. 5 should not be exceeded, this demands that the Pu-241 content of the target be limited to 1% since the initial heat generation rate from one gram of Pu-241 exposed to a 3×10^{15} flux is 242 kw.

The average microscopic cross section of the target material as a function of time is also shown in Fig. 5. Note that the average cross section rapidly rises to a maximum of 70 barns and then drops off to 25 barns after 2 years. This particular behavior is similar to the behavior of the average cross section of the fission products. This, and the fact that the majority of the fission products are generated in the first half years, (see power curve of Fig. 5) will allow considerable simplification in estimating the effect of the total cross section on flux depression in the target material.

The net cumulative production of stable isotopes as a function of time, other than Cf-252, is shown in Fig. 6A and 6B. After about one year of exposure to a 3×10^{15} flux, all isotopes of lower mass number than Cm-248 have passed through a maximum. Berkelium-249 and higher isotopes continue to increase beyond two years of irradiation time. This demonstrates graphically that Cm-248 is the "bottleneck" in the production of Californium. Estimates of the cross section of Cm-248 have ranged from 2 to 10 barns. In this study the value of 2 barns was used. Using 10 barns with no changes in other cross sections would cause a large increase in the rate of Cf-252 production. Whether one is justified in changing the Cm-248 cross section without concomitant changes in the others is doubtful in view of some inherent interdependence of the cross sections that results from the method of arriving at the consistent set.

PRODUCTION RATE MAXIMIZATION

Without regard to cost of processing, production of Cf-252 can be maximized on the basis of the largest net yield over a period of time, which will be called the maximum average production rate.

The time of irradiation of Pu-242 required to give the maximum average production rate is readily found as shown in Fig. 7, if recycling of curium is not considered. The slope of a straight line drawn from the origin to a point on the net cumulative production curve is the average production rate for the particular neutron exposure. Obviously the maximum slope, and hence the maximum average production rate, is found when the line is drawn tangent to the curve. With no recycling of curium, the maximum average production rate occurs at a neutron exposure of 5.7×10^{15} n yr/cm^2 sec, which is equivalent to an initial irradiation time of 1.9 yr for an average target flux of 3×10^{15} n/cm^2 sec. This is also clearly indicated in Table 2.

When curium is recycled, maximizing the production rate involves the time for chemical processing and target fabrication. It was assumed that a 3-month period was sufficient. The procedure for determining the optimum irradiation time for each curium recycle is similar to that shown in Fig. 7 except allowance must be made for processing time. The result of optimizing the curium recycle time from three different initial irradiation times for a 3×10^{15} flux is shown in Fig. 8 (in which all and only curium resulting from the initial irradiation of Pu-242 was considered). The recycle irradiation time is measured from zero on the abscissa; -0.25 yr. represents the discharge time after the initial irradiation. Allowance for chemical processing time is made simply by drawing the tangent line through the latter point. Each succeeding recycle was optimized in the same manner and the results for three recycles are shown in Table 2. Eventually, however, the production rate from recycled material would fall off due to Cm-248 depletion. Consequently, in order to attain full utilization of the potential production from each target tube, blending or mixing of the recycle material becomes necessary. This leads to a complicated irradiation

Table 2

Maximum Average Cf-252 Production Rates[a]

Initial irrad. time, yr.	Cycle irrad. time, yr.	Total time,[b] yr.	Cycle production, mg	Cum. prod. all cycles, mg	Average prod. rates,[c] mg/yr.
		Initial Irradiation of 100 g Pu-242			
1.0	1	1.25	30.2	30.2	24.2
1.5	1.5	1.75	68.8	68.8	39.3
2.0	2.0	2.25	97.5	97.5	43.3
2.5	2.5	2.75	112	112	40.7
		1st Curium Recycle			
1.0	0.60	2.10	70.5	101	48.0
1.5	0.42	2.42	74.2	143	59.1
2.0	0.37	2.87	76.7	174	60.7
2.5	0.35	3.35	76.0	188	56.1
		2nd Curium Recycle			
1.0	0.40	2.75	74.5	175	62.5
1.5	0.37	3.04	76.0	219	72.0
2.0	0.35	3.47	75.8	250	72.0
2.5	0.34	3.94	73.2	261	66.2
		3rd Curium Recycle			
1.0	0.37	3.37	76.3	251	74.5
1.5	0.35	3.64	75.4	294	80.8
2.0	0.35	4.06	71.5	322	79.3
2.5	0.34	4.53	71.5	333	73.5

[a] Assumes uniform flux of 3×10^{15} in target

[b] Includes 3-month processing time

[c] Cumulative production divided by total time

schedule. A "steady-state" production rate may be estimated by extrapolating the data of Table 2. When heat transfer limitations and processing time are considered, on an average a little over half the target tube will be filled with Pu-242 during initial irradiation of one to 1.5 years while the remainder will be filled with recycle material that on an average has been irradiated somewhat over two years. For an initial 200 gram Pu-242 loading, this will lead to an eventual production rate of about one gram/yr of Cf-252.

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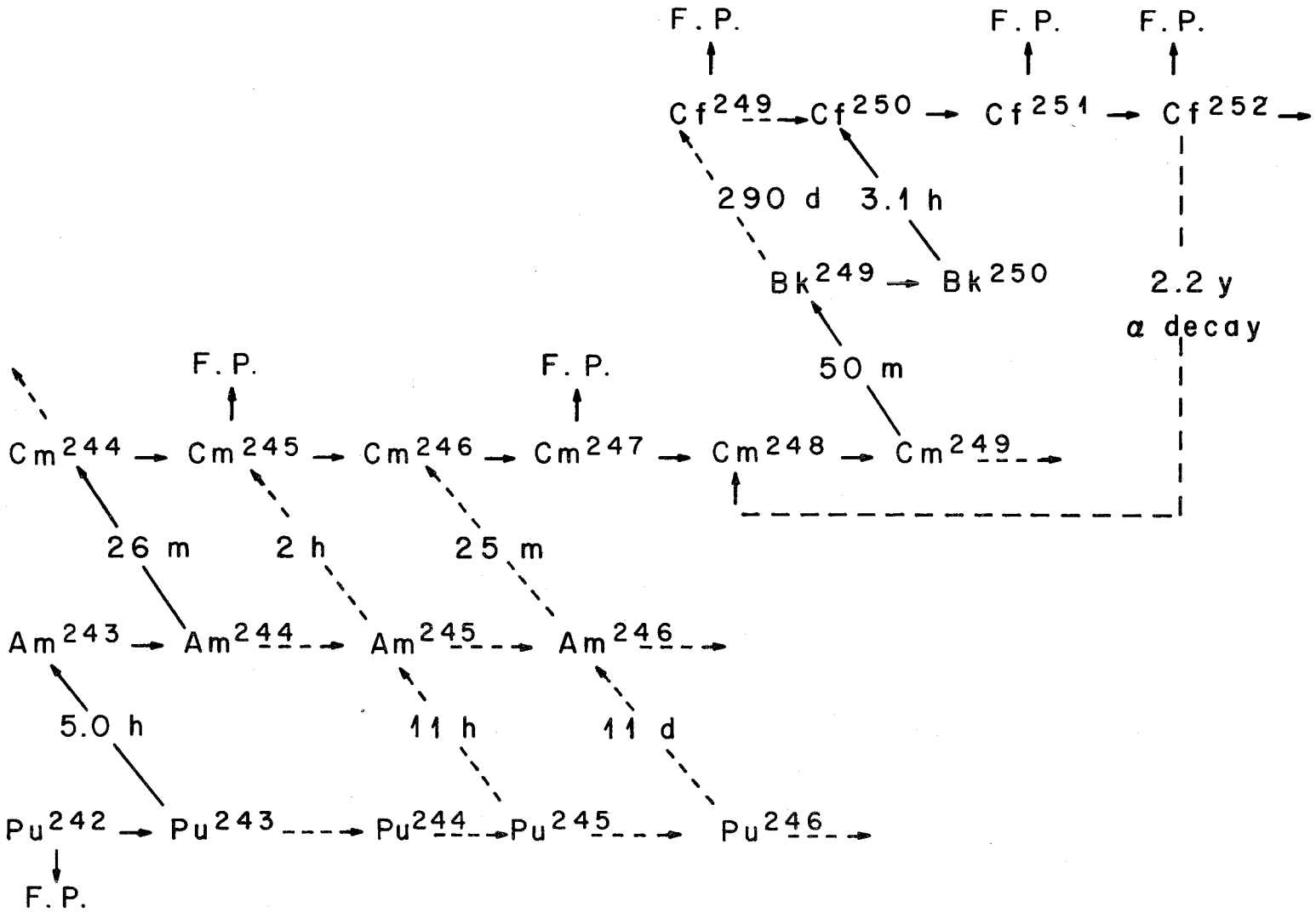

Fig. 1. Reaction Paths in the Production of Cf^{252} from Pu^{242}

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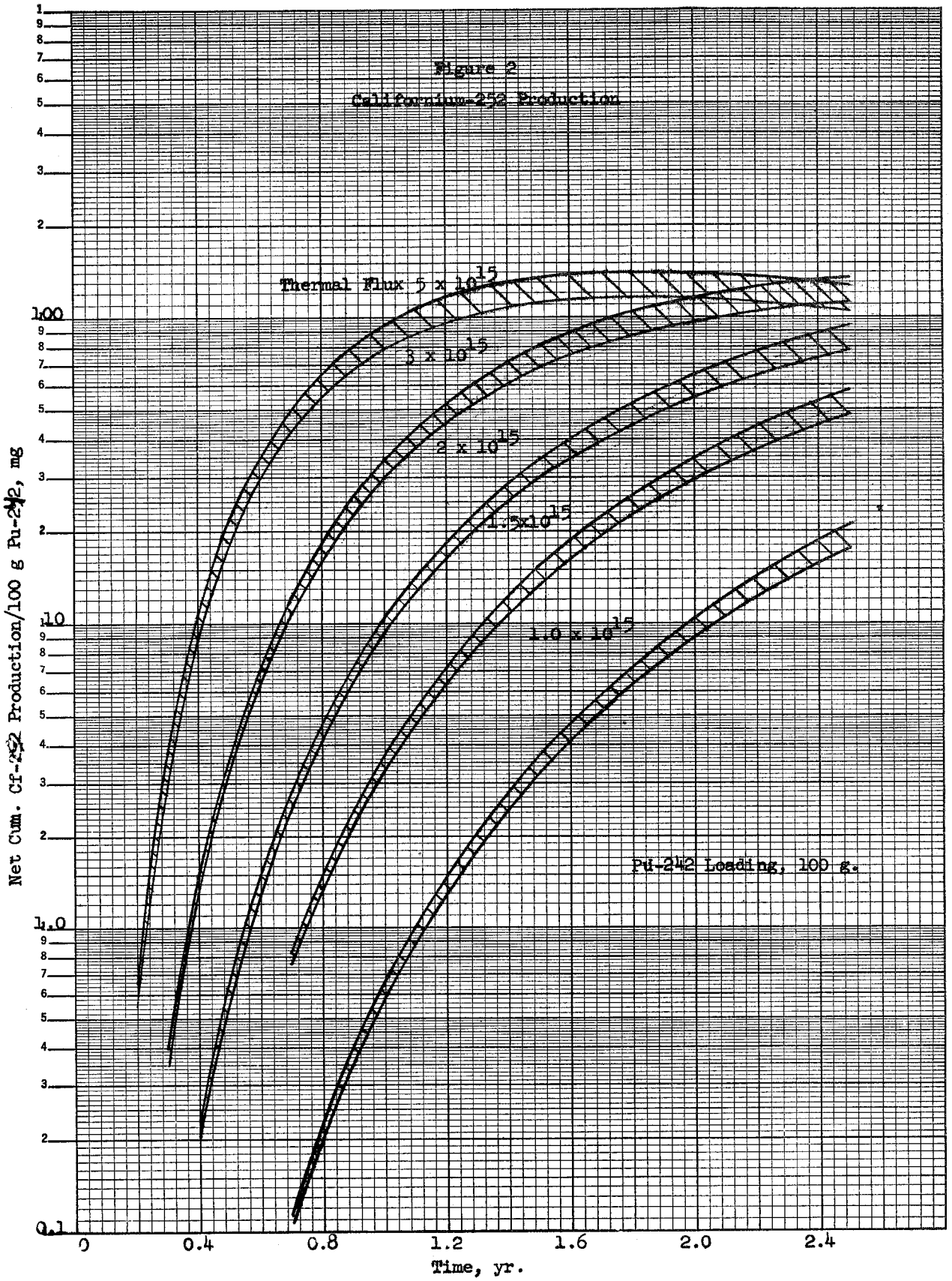

Figure 2
Californium-252 Production

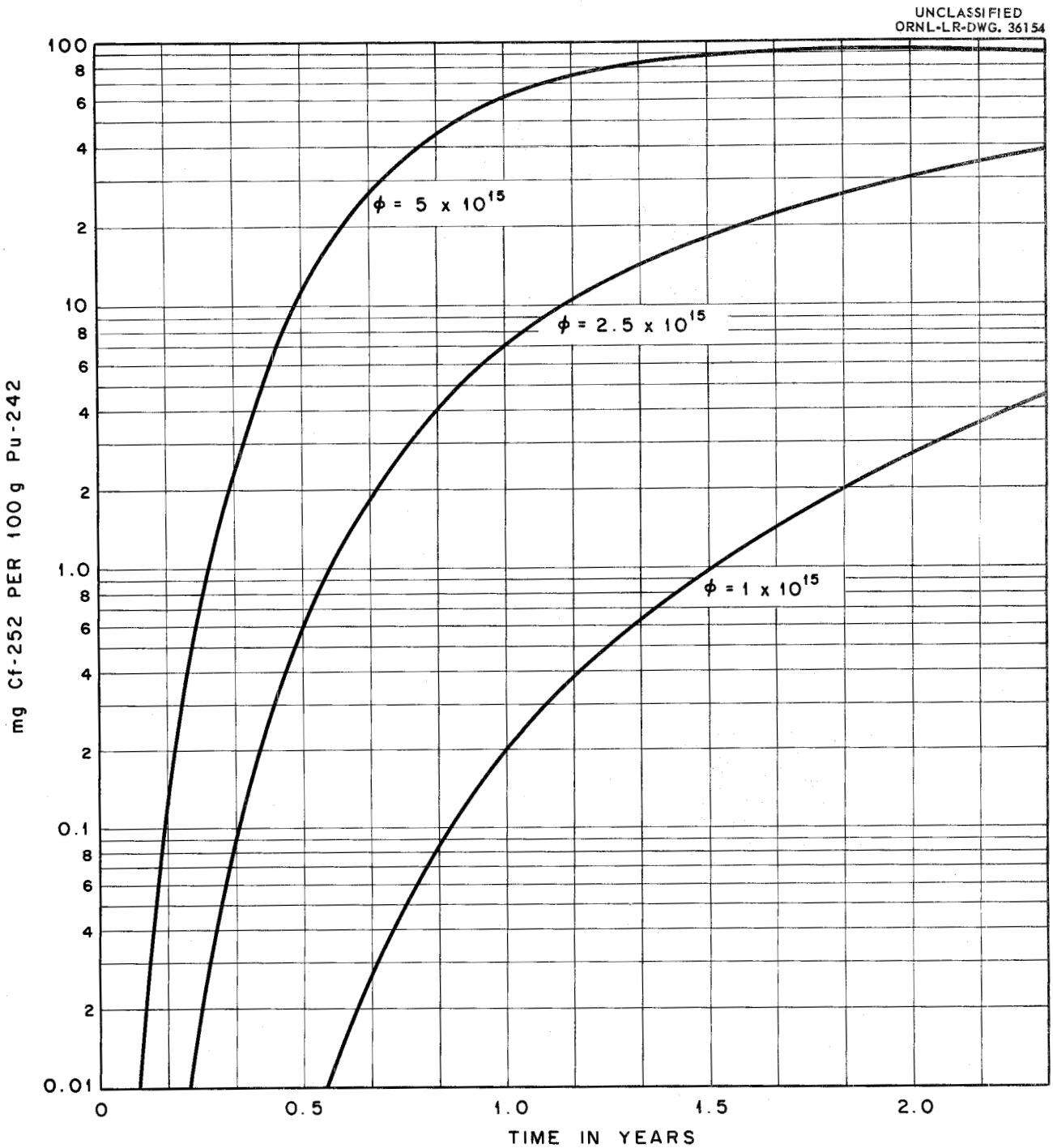

Figure 3. Production of Cf-252 from Pu-242 (from CF 59-2-65)

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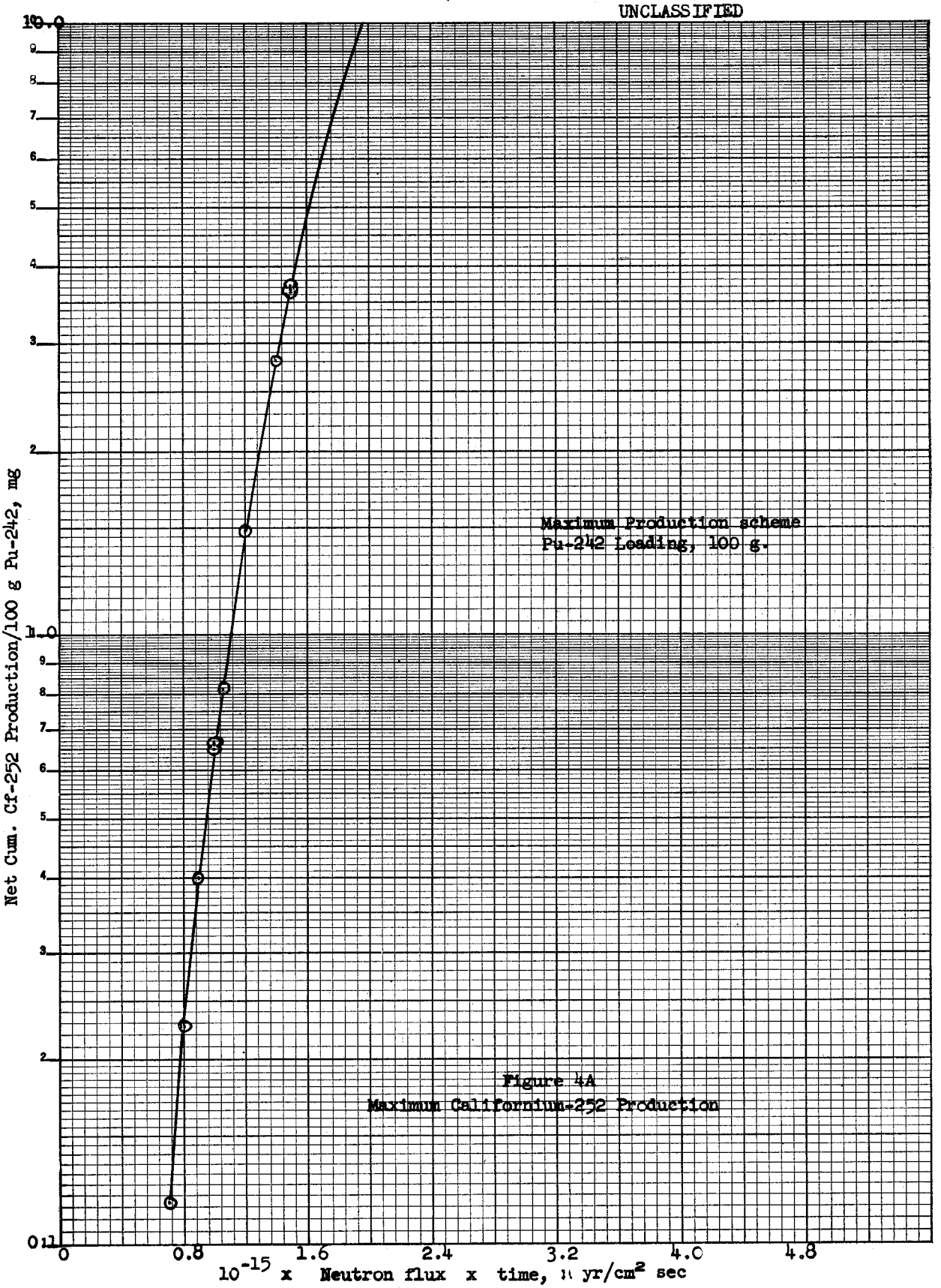

Figure 4A
Maximum Californium-252 Production

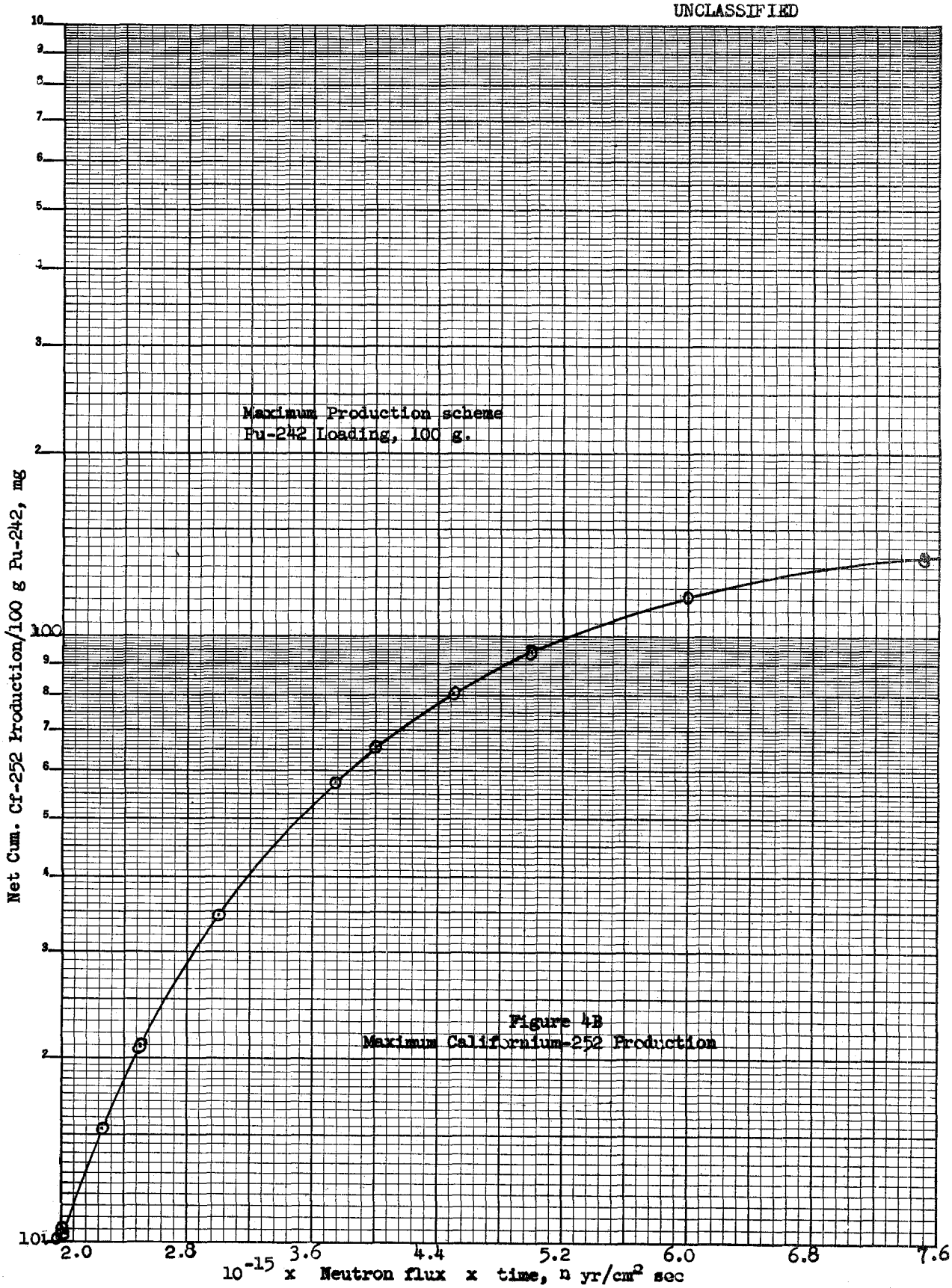

Figure 4B
Maximum Californium-252 Production

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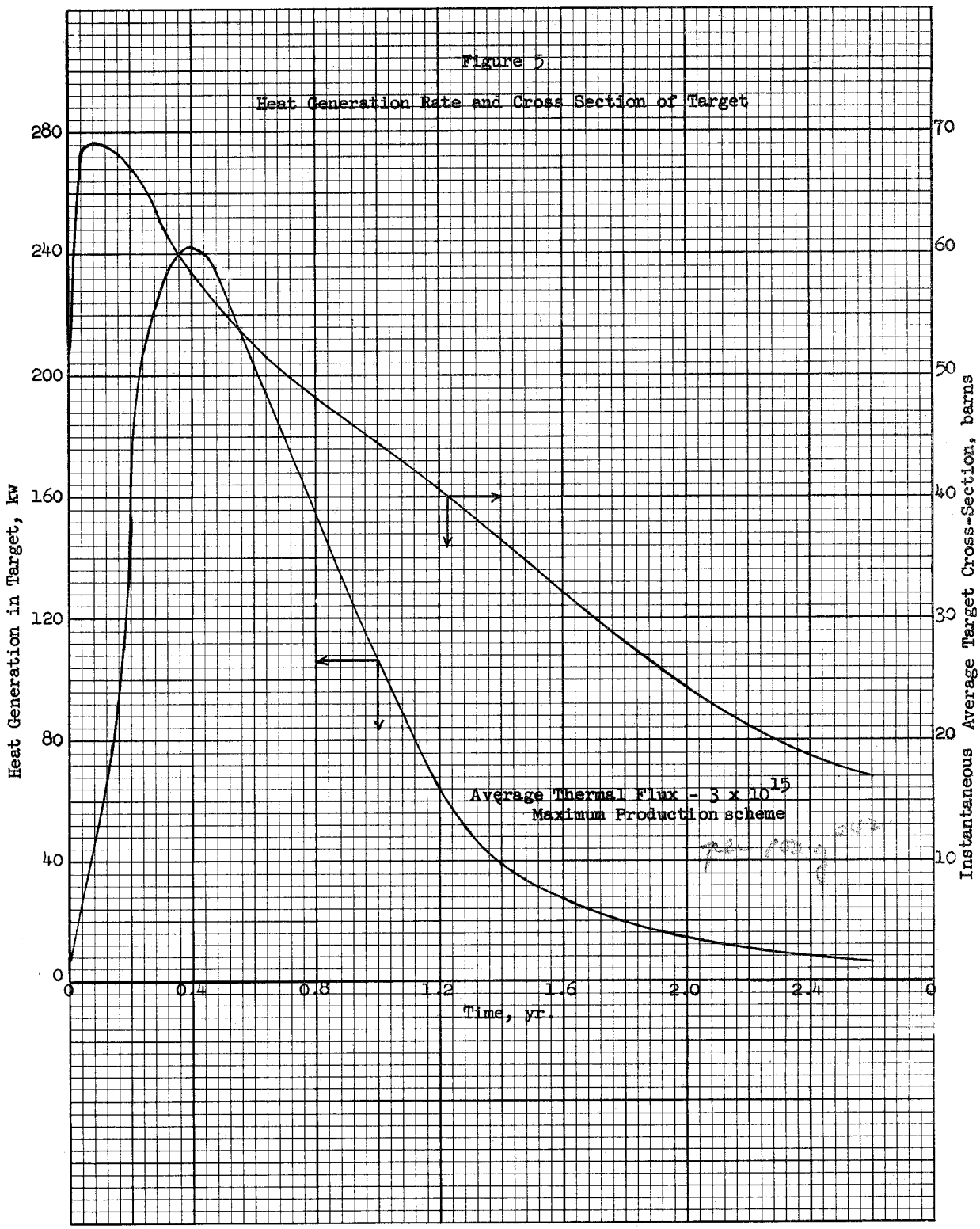

Figure 5

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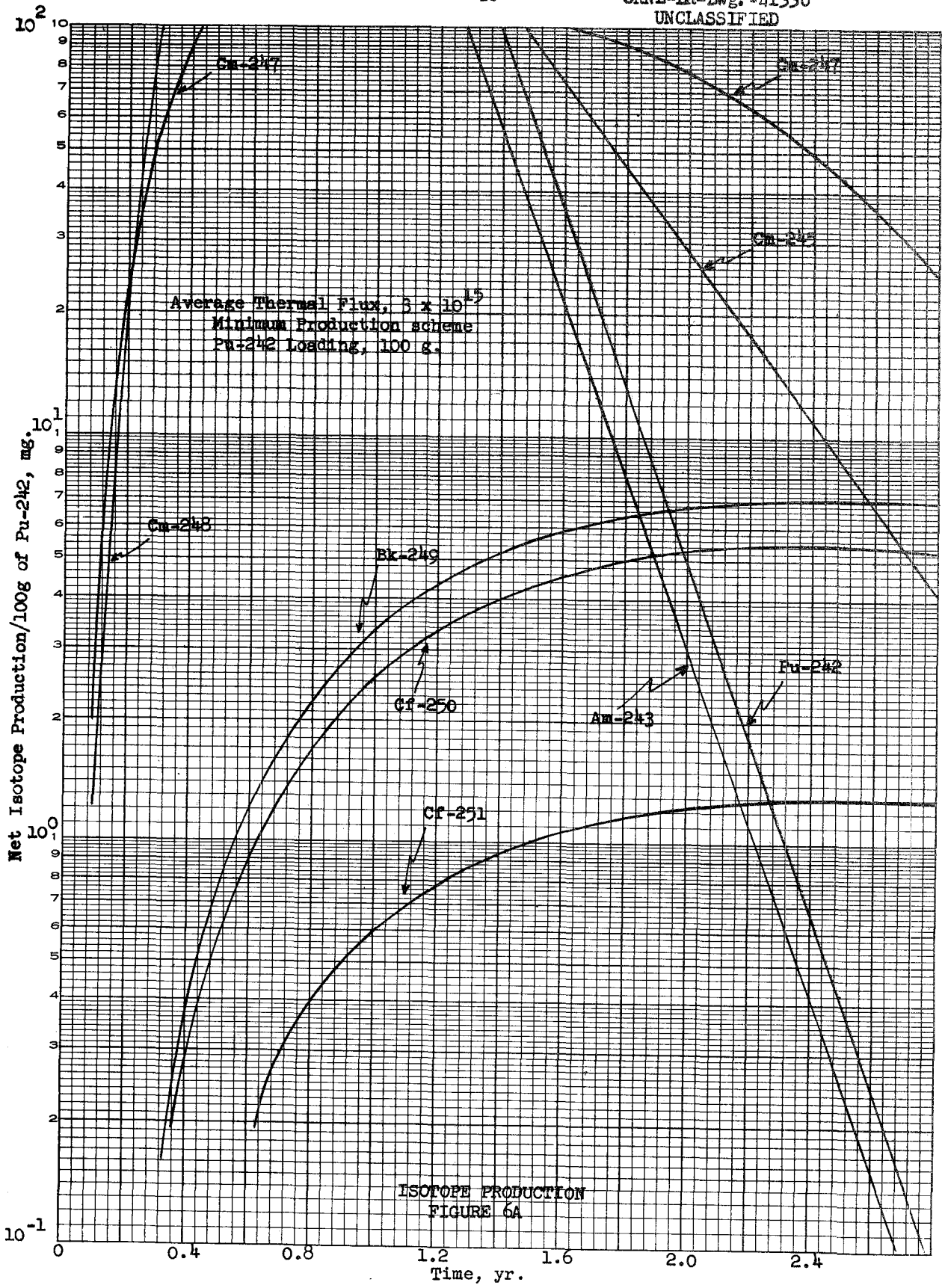

ISOTOPE PRODUCTION
FIGURE 6A

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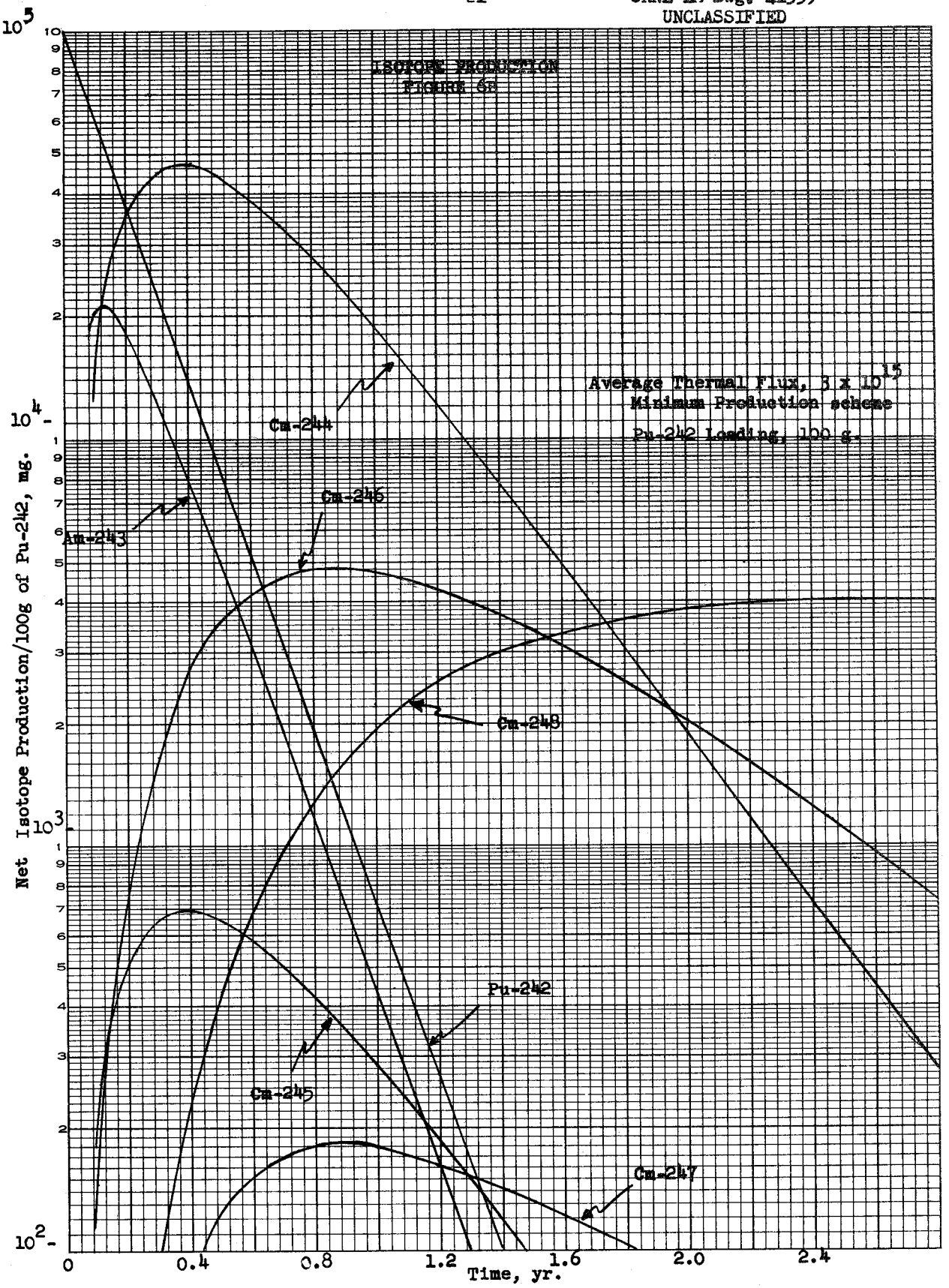
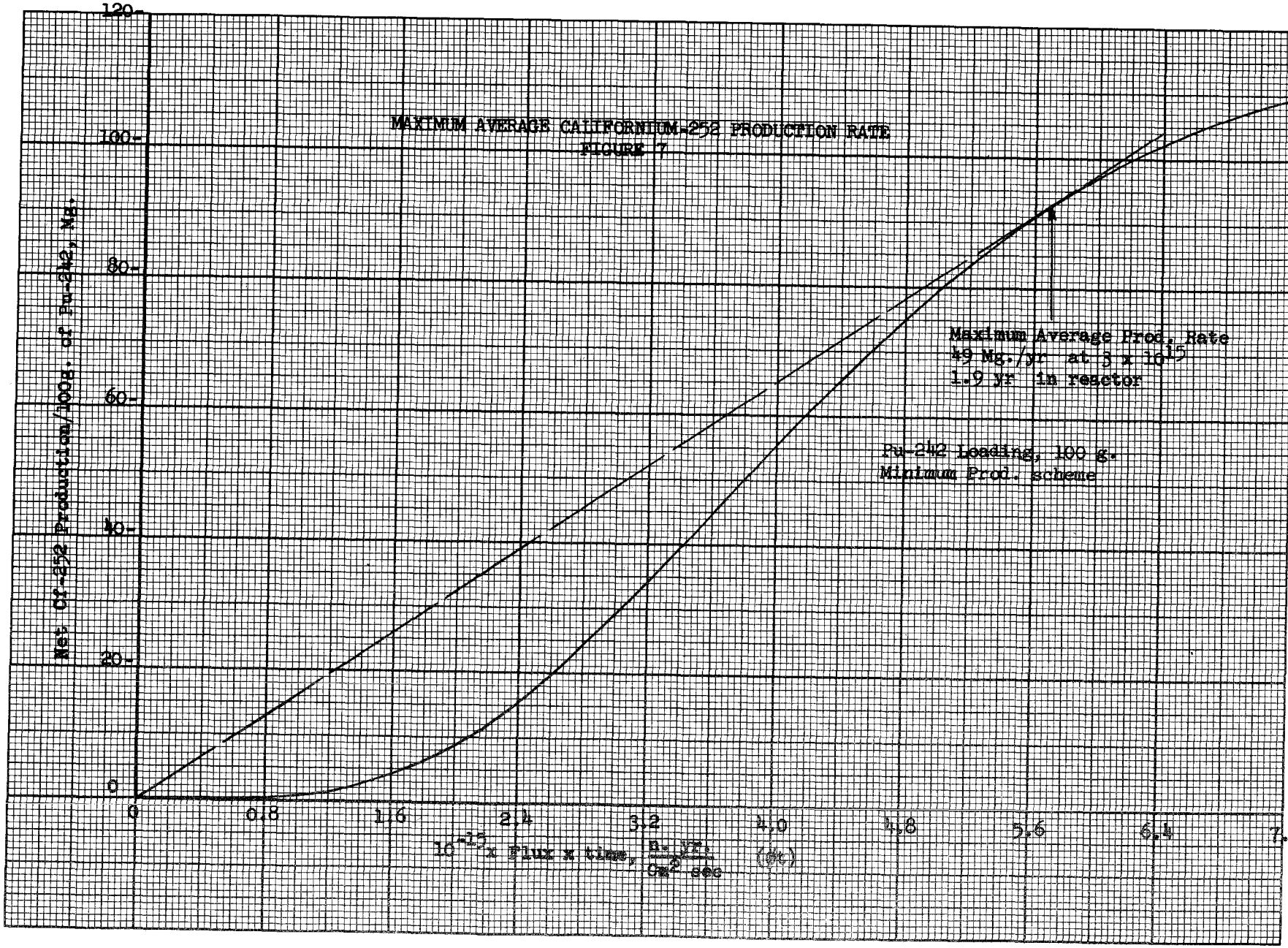


MAXIMUM AVERAGE CALIFORNIUM-252 PRODUCTION RATE
FIGURE 7



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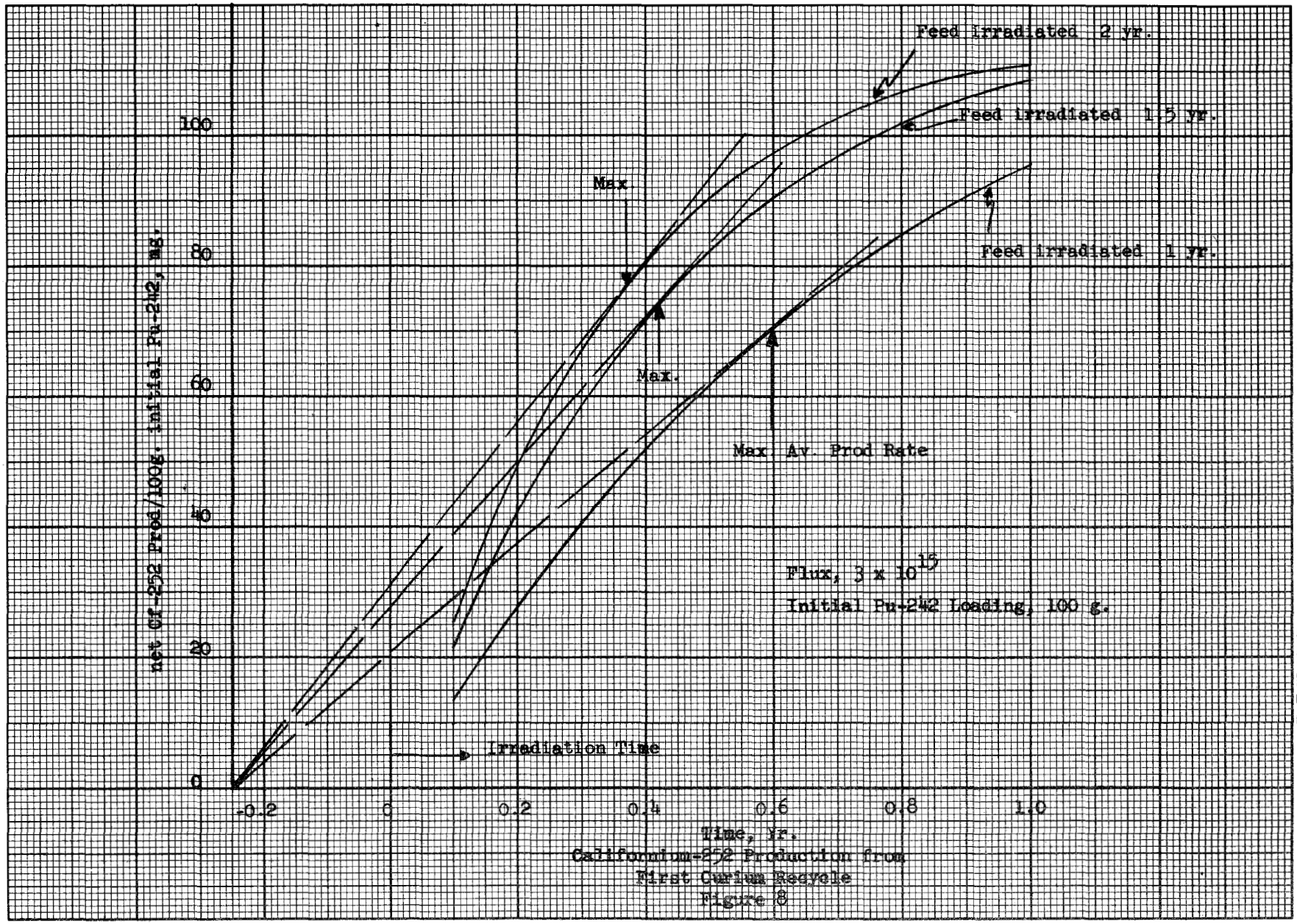

Californium-252 Production from
First Curium Recycle
Figure 8

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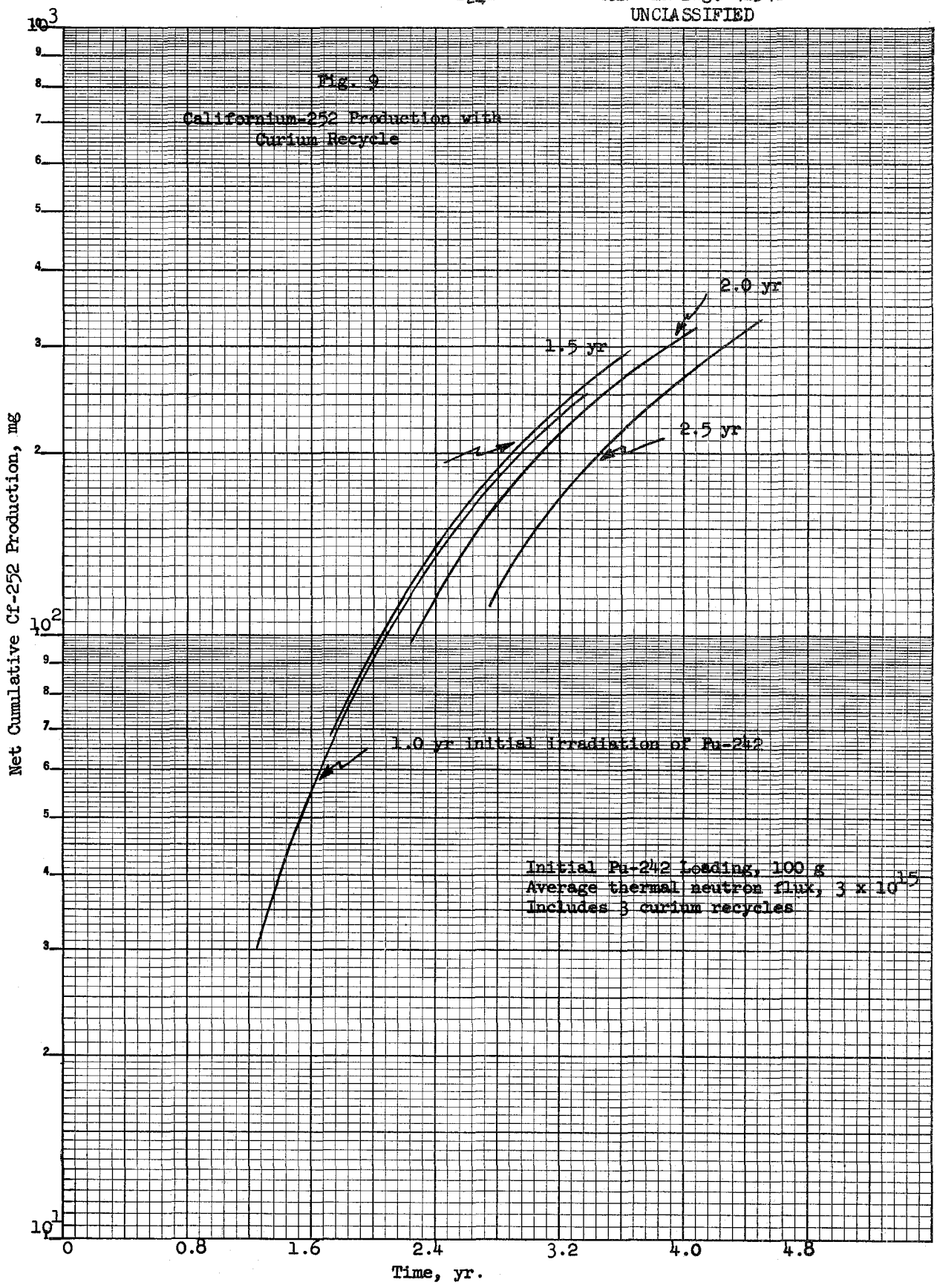

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