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### DISCUSSION OF AN ACTIVATION METHOD FOR MEASURING THE RATIO OF $U^{238}$ ABSORPTION TO $U^{235}$ FISSION IN A REACTOR

C. A. Preskitt

#### ABSTRACT

The theory is presented in detail of an activation method for measuring the ratio of  $U^{238}$  absorptions to  $U^{235}$  fissions in a nuclear reactor. The origin of the activities is discussed, the equations are given for interpretation of the measurements, and the expected counting rates are computed for a typical experiment in the EGCR.

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DISCUSSION OF AN ACTIVATION METHOD FOR MEASURING THE RATIO  
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Introduction

This report presents in detail the theory supporting an activation method for measuring the ratio of  $U^{238}$  absorptions to  $U^{235}$  fissions in a nuclear reactor. Experiments of this sort have been conducted for some time, but the important innovation of the method described here is the use of a coincidence counting technique due to Sher,<sup>1</sup> Weitzberg,<sup>2</sup> and Antunez<sup>3</sup> which greatly reduces the fission-product gamma-ray background while counting the  $Np^{239}$  activity for determination of the  $U^{238}$  reaction rate. In the past, chemical separation of the neptunium has been required to reduce this background to an acceptable level.

The author believes that the experiment described here would be an extremely useful part of the experimental program during startup of the EGCR.

The experiment yields directly the ratio of  $U^{238}$  absorption to  $U^{235}$  fission and would be an important aid in comparing the initial critical experiments with the design calculations. The required experimental determinations are rather simple and no elaborate counting equipment is necessary.

Discussion of Observable Activities

In essence, the experiment is performed by relating the  $U^{238}$  absorption rate to measurements of the  $Np^{239}$  decay rate as observed through the 106-kev decay gamma ray of  $Np^{239}$ , and by relating the  $U^{235}$  fission rate to the fission-product decay-gamma activity in an energy range where no  $Np^{239}$  activity is found.

The basis for the discrimination against fission-product activity by coincidence counting in the  $Np^{239}$  decay spectrum is illustrated by the decay diagrams shown in Fig. 1.<sup>4</sup> The transitions which are shown are not all of those possible, since those weak enough to be unimportant here have been omitted for simplicity. The beta decay of  $Np^{239}$  occurs

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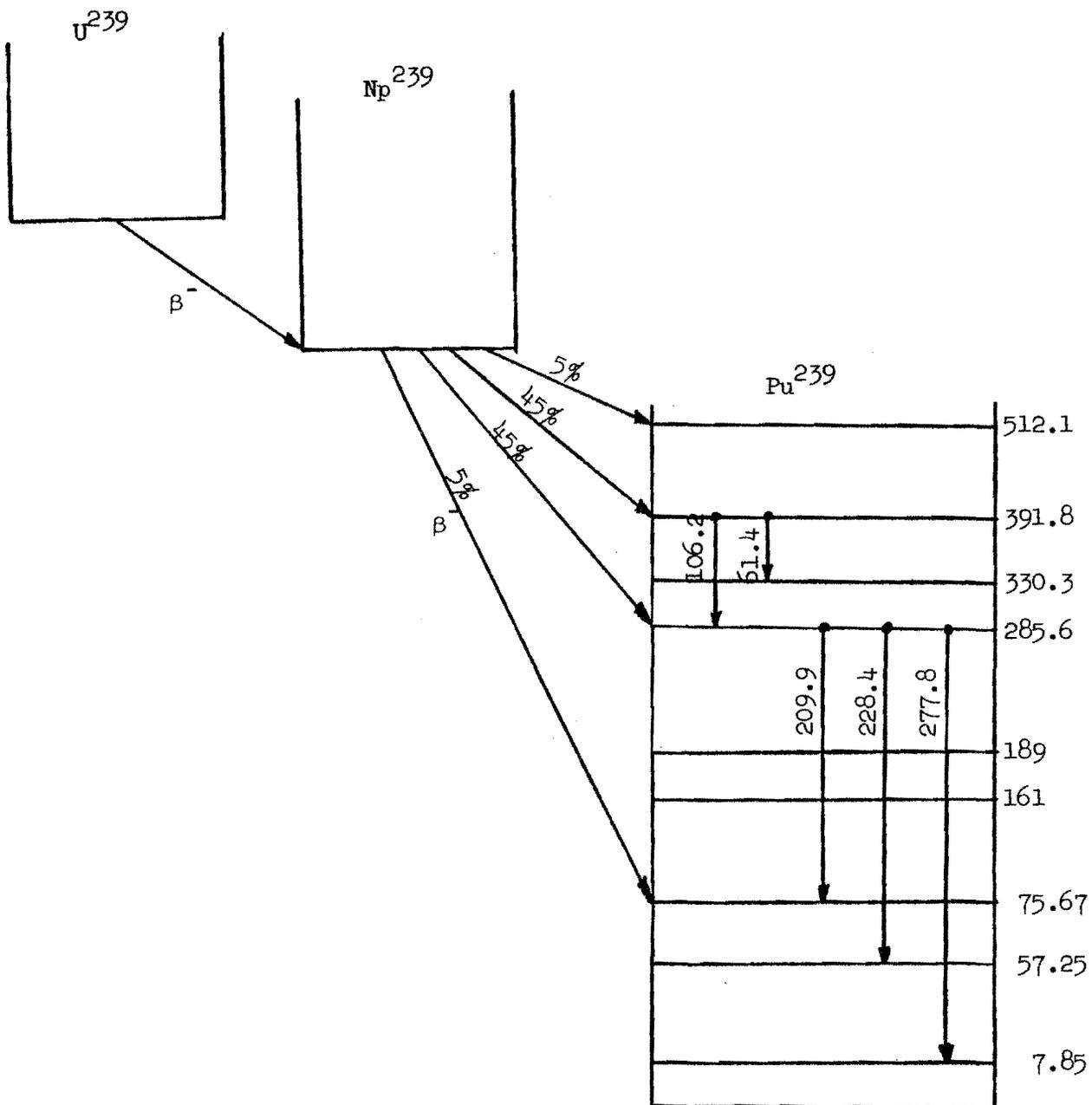


Fig. 1. Level Diagrams for the Decay of the  $U^{239}$ ,  $Np^{239}$ ,  $Pu^{239}$  Chain

with at least four beta groups, the transitions going predominately (90%) to two metastable states in  $\text{Pu}^{239}$  at 285.6 keV and 391.8 keV. The decay of the 391.8-keV state is primarily through a parity-favored E1 gamma transition to the 285.6-keV state, resulting in a 106.2-keV gamma ray. This state may also undergo a parity-favored E1 transition to a state at 330.3 keV by emission of a 61.4-keV gamma ray, but this transition occurs in only 13% of the decays of the 391.8-keV state.

Focussing attention now on the state at 285.6 keV we see that this state is populated through two routes, one directly by 45% of the beta transitions from  $\text{Np}^{239}$ , and the other by about 87% of the decays of the 391.8-keV state. All states below 391.8 keV have even parity with the result that the lowest order radiative transitions possible in this range are types M1 and E2. The decay of the 285.6-keV state consequently proceeds primarily by internal conversion. The relative transition intensities for all of the important transitions are given in Table 1.<sup>4</sup>

Table 1. Relative Transition Intensities for the Important Transitions in  $\text{Pu}^{239}$  When Populated by the Beta Decay of  $\text{Np}^{239}$

Transition Energy (keV)	Relative Transition Intensity		
	Internal Conversion	Radiative	Total
61.4	10	340	350
106.2	500	1800	2300
209.9	700	140	840
228.4	1980	440	2420
277.8	1400	600	2000

The x-ray resulting from the three internal conversion transitions from the 285.6-keV state has an energy of 103 keV, and the coincidence between this x-ray and the 106.2-keV decay gamma ray is a convenient indicator for the  $\text{Np}^{239}$  beta decay. From the beta transition probabilities in Fig. 1 and the relative intensities in Table 1, it is apparent that the 106.2-keV gamma ray results in  $45\% \times 87\% = 39\%$  of the

$\text{Np}^{239}$  decays and that it will be in coincidence with the 103-keV x-ray in  $39\% \times (700 + 1980 + 1400)/(840 + 2420 + 2000) = 30\%$  of the  $\text{Np}^{239}$  decays.

### Interpretation of the Measurements

The material to be irradiated in the experiment and subsequently counted could be uranium foils with the same enrichment as the reactor fuel, or could be pellets of the fuel itself. In either case the sample would be dissolved and the concentration and volume adjusted to obtain a convenient geometry and counting rate.

A typical setup for the counting apparatus could be as shown in Fig. 2. The integral discriminator is set with a base line of about 600 keV so that it will discriminate against all gamma rays except those from fission-product decay. The coincidence channel is set with both windows centered at 104 keV with widths of about 25-40 keV, depending on the energy resolution of the counters. After correcting for counting losses and accidental coincidences, we may write for the true coincidence rate ( $L_c$ ) and the true singles rate ( $L_s$ )\*

$$L_c = E_1^{39} P_{39} + E_1^{25} F_{25}, \quad (1)$$

$$L_s = E_2^{25} F_{25} + E_2^{28} F_{28}. \quad (2)$$

If we divide Eq. (1) by Eq. (2) and let

$$\delta = \frac{E_2^{28} F_{28}}{E_2^{25} F_{25}} \quad (3)$$

then, solving for  $P_{39}/F_{25}$ , we obtain

$$\frac{P_{39}}{F_{25}} = \left\{ \frac{L_c}{L_s} (1 + \delta) - \frac{E_1^{25}}{E_2^{25}} \right\} \cdot \frac{E_2^{25}}{E_1^{39}}. \quad (4)$$

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\* All nomenclature is explained in the Appendix.

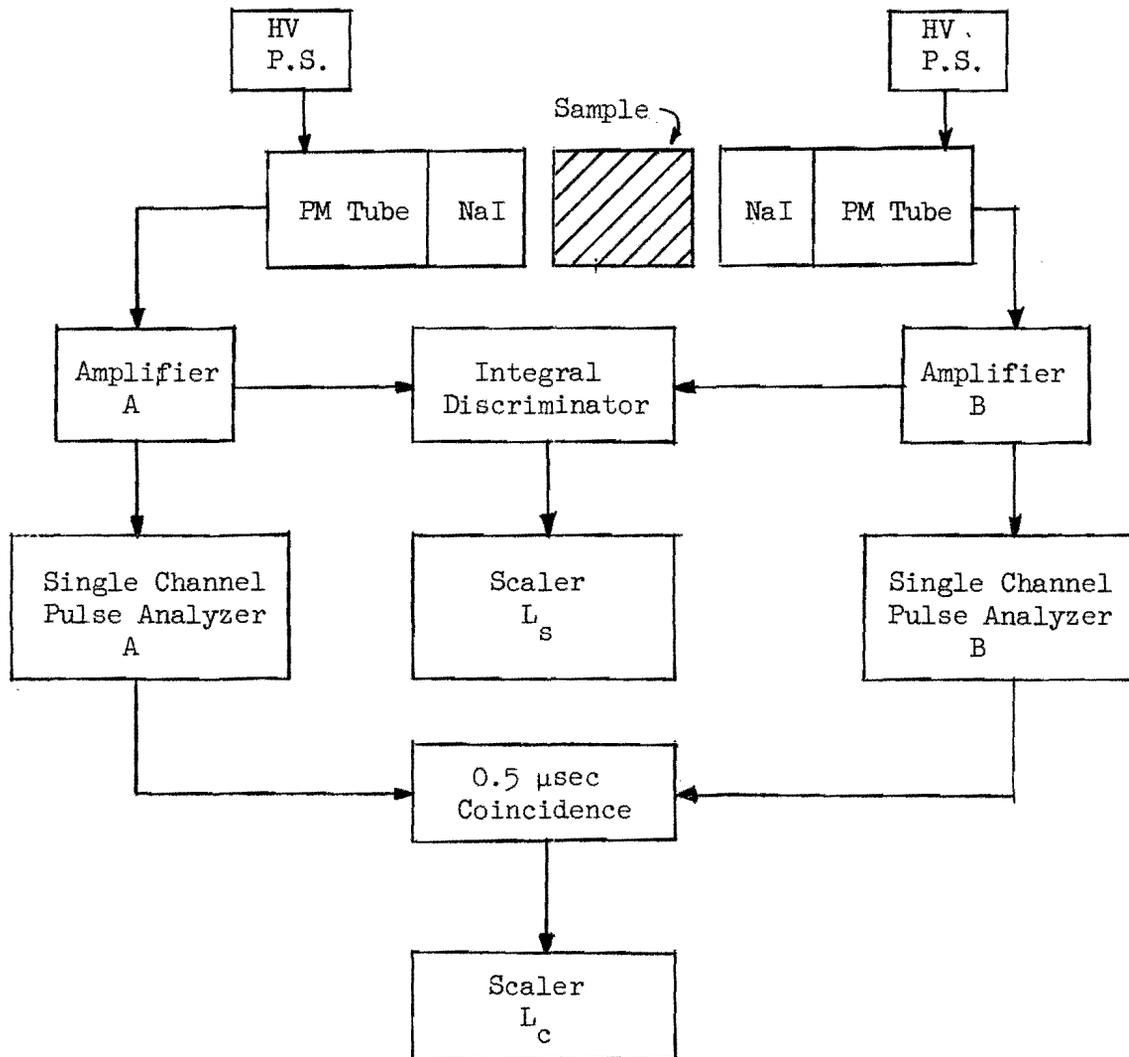
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Fig. 2. Typical Counting Setup

This ratio is the desired quantity since it is directly equal to the  $U^{238}$  absorption rate to the  $U^{235}$  fission rate in the reactor.

It should be pointed out that a term proportional to  $F_{28}$  should appear in Eq. (1) as it does in Eq. (2). This term is at most  $\sim 2\%$  of the second term in Eq. (1) and is, therefore, entirely negligible compared to  $E_1^{39} P_{39}$ .

Typical values of the terms in Eq. (4) are:<sup>5</sup>

$$\frac{E_1^{25}}{E_2^{25}} \sim 0.01$$

$$\frac{E_1^{39}}{E_2^{25}} \sim 0.8$$

$$\delta \sim 0.02$$

$$\frac{L_c}{L_s} \sim 0.4$$

As indicated later, each of these will be known to within counting statistics so that the uncertainties in  $L_c/L_s$  and  $E_1^{39}/E_2^{25}$  are the principal limitations in accuracy.

Each of the four terms listed above which appear in Eq. (4) may be measured experimentally by irradiating additional samples of other enrichments as described below.

Measurement of  $E_1^{39}/E_2^{25}$  and  $E_1^{25}/E_2^{25}$

In order to determine this ratio we irradiate samples of two enrichments in a pure thermal flux. This could be, for example, at the thermal column of a reactor like the ORNL Graphite Reactor. We write Eqs. (1) and (2) for each enrichment, using single and double primes to designate the two cases. Note that the term involving  $F_{28}$  will vanish due to the assumption of a pure thermal flux, and that the E factors depend on the counting conditions but not the irradiation conditions.

$$L'_c = E_1^{39} P'_{39} + E_1^{25} F'_{25} \quad (5)$$

$$L'_s = E_2^{25} F'_{25} \quad (6)$$

$$L''_c = E_1^{39} P''_{39} + E_1^{25} F''_{25} \quad (7)$$

$$L''_s = E_2^{25} F''_{25} \quad (8)$$

Dividing Eq. (5) by Eq. (6) and Eq. (7) by Eq. (8), we obtain two equations containing the ratios  $E_1^{39}/E_2^{25}$  and  $E_1^{25}/E_2^{25}$ . Solving the equations for these two quantities give

$$\frac{E_1^{39}}{E_2^{25}} = \frac{L''_c/L''_s - L'_c/L'_s}{P''_{39}/F''_{25} - P'_{39}/F'_{25}} \quad (9)$$

and

$$\frac{E_1^{25}}{E_2^{25}} = \frac{\frac{L'_c}{L'_s} \frac{P''_{39}}{F''_{25}} - \frac{L''_c}{L''_s} \frac{P'_{39}}{F'_{25}}}{\frac{P''_{39}}{F''_{25}} - \frac{P'_{39}}{F'_{25}}} \quad (10)$$

Since the irradiation was done in a known spectrum, purely thermal, we may write the  $P_{39}/F_{25}$  ratios in terms of known cross sections,

$$\frac{P''_{39}}{F''_{25}} = \frac{\Sigma''_{28}}{\Sigma''_{f,25}}$$

$$\frac{P'_{39}}{F'_{25}} = \frac{\Sigma'_{28}}{\Sigma'_{f,25}}$$

and Eqs. (9) and (10) may be evaluated.

Several significant observations can be made from the above relationships. From Eqs. (5-8) it may be seen that

$$\frac{L_c}{L_s} \approx \frac{E_1^{39}}{E_2^{25}} \frac{P_{39}}{F_{25}} \quad (11)$$

so that the coincidence to singles counting rate is approximately proportional to  $P_{39}/F_{25}$ . With this it is clear from Eqs. (9) and (10) that  $E_1^{25}/E_2^{25}$  is near zero and  $E_1^{39}/E_2^{25}$  is near unity. In fact, from Eq. (4) we can see that it is desirable for accuracy to make  $E_1^{25}/E_2^{25}$  as nearly zero as possible. This may be accomplished either by reduction of the base line of the singles counting channel, by reducing the window width of the coincidence channel, or by increasing the detection efficiency for counting the high energy gamma rays in the singles channel. In selecting and setting up the counting equipment we should, therefore, take the following precautions.

1. Set the base line in the singles channel as low as possible without counting the highest energy gamma rays of the neptunium decay.
2. Set the window widths of the coincidence channel as narrow as possible without causing a significant reduction in the coincidence counting rate.
3. Select reasonably large NaI crystals for high detection efficiency.
4. Count the samples over an interval of time when the ratio of fission-product activity to neptunium activity is at a minimum. This later point will be discussed quantitatively in a later section.

Measurement of  $\frac{E_{28}^{28} F_{28}}{E_2^{25} F_{25}}$

In order to determine this quantity we irradiate a sample of a second enrichment in the reactor of interest and count the singles and coincidences as before. Then, in addition to Eqs. (1) and (2), we write the same relationships for the second enrichment and obtain by division

$$\frac{L_c}{L_s} = \frac{\frac{E_1^{39}}{E_2^{25}} \frac{P_{39}}{F_{25}} + \frac{E_1^{25}}{E_2^{25}}}{1 + \frac{E_2^{28} F_{28}}{E_2^{25} F_{25}}}, \quad (12)$$

and for the second enrichment

$$\frac{L'_c}{L'_s} = \frac{\frac{E_1^{39}}{E_2^{25}} \frac{P'_{39}}{F'_{25}} + \frac{E_1^{25}}{E_2^{25}}}{1 + \frac{E_2^{28} F'_{28}}{F_2^{25} F'_{25}}} \quad (13)$$

Since the enrichment alone has changed we may accurately ascribe the difference between Eqs. (12) and (13) to changes in the  $F_{25}$  factors and assume that  $P_{39}$  and  $F_{28}$  are unaffected by the change in enrichment. The result of this will be to make

$$\frac{P'_{39}}{F'_{25}} = \frac{1}{\alpha} \frac{P_{39}}{F_{25}} \quad (14)$$

and

$$\frac{E_2^{28} F'_{28}}{E_2^{25} F'_{25}} = \frac{1}{\alpha} \frac{E_2^{28} F_{28}}{E_2^{25} F_{25}} \quad (15)$$

where the factor  $\alpha$  is very nearly just the ratio of enrichments, the difference arising from any changes in the self shielding of the sample as the enrichment is changed. That is,

$$\alpha = \frac{e'}{e} \frac{f'}{f}, \quad (16)$$

where  $f'/f$  is the ratio of the two self-shielding factors.

If Eqs. (14) and (15) are substituted into Eqs. (12) and (13) we can solve directly for the desired quantity. We obtain

$$\frac{E_2^{28} F_{28}}{E_2^{25} F_{25}} = \frac{\frac{L_c}{L_s} - \alpha \frac{L'_c}{L'_s} + (\alpha - 1) \frac{E_1^{25}}{E_2^{25}}}{\frac{L'_c}{L'_s} - \frac{L_c}{L_s}} \quad (17)$$

This is the quantity  $\delta$  appearing in Eq. (4). Since  $E_1^{25}/E_2^{25}$  is nearly zero for reasons given previously and since Eq. (14) implies that

$$\frac{L_c}{L_s} \cong \alpha \frac{L'_c}{L'_s},$$

it is easily seen that  $\delta$  is small compared to unity.

### Analysis of Required Counting Rates and Neutron Exposure

In order to estimate the sample exposures necessary to obtain reasonable counting rates the results of Perkins and King<sup>6</sup> have been used to estimate the gamma source strengths due to fission-product decay. These data are presented for the interval up to 1000 hr after irradiation in Figs. 3 through 7. For the EGCR core the absorption in  $U^{238}$  is  $\sim 0.27$  per source neutron or  $\sim 0.66$  per fission. Based on this value for the  $Np^{239}$  production rate, the  $Np^{239}$  decay will follow the curve shown in Fig. 8.

In order to determine the optimum counting interval we wish to know the period after irradiation during which  $L_c/L_s$  is most nearly independent of time and during which the ratio of  $Np^{239}$  activity to fission-product activity near 100 kev is a maximum. This may be determined from the data shown in Figs. 3 through 8 if we assume that the singles rate,  $L_s$ , is proportional to the total gamma decay rate above 400 kev, and assume that the intensity of fission-product gamma rays near 100 kev follows the decay curve of the group from 100 kev to 400 kev. The variations resulting from these assumptions are shown in Fig. 9. The ratio  $L_c/L_s$  increases rapidly at first due to the decay of short lived fission products and goes through a maximum at about 100 hr after the irradiation. The ratio of  $Np^{239}$  activity to fission-product activity at 100 kev (actually 100 kev to 400 kev) shows a similar variation.

In view of these variations the optimum counting interval appears to lie between 50 and 200 hr after the irradiation. During this period the variation of  $L_c/L_s$  will be only about 25%. The importance of this

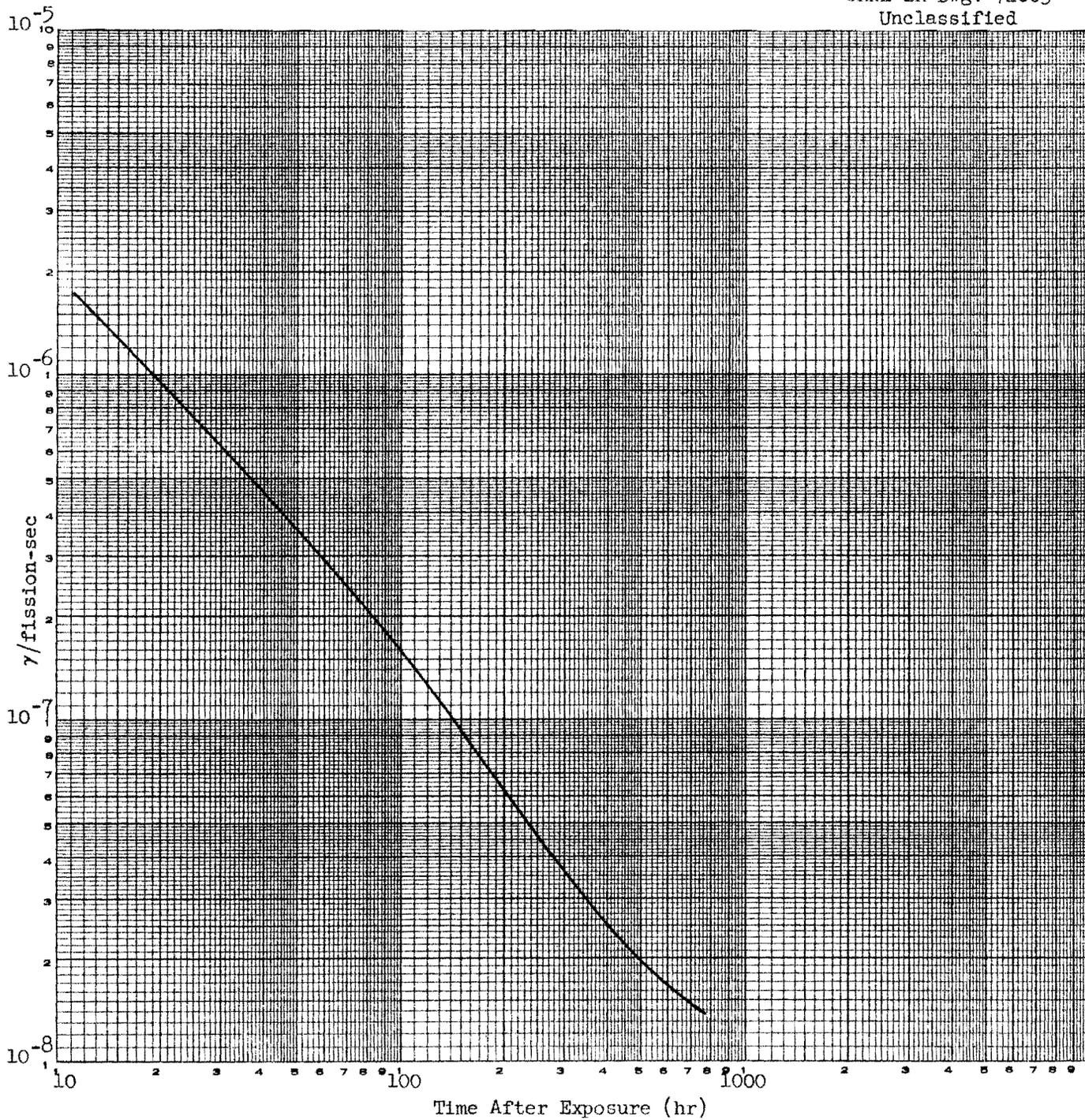


Fig. 3. Decay Curve for Fission-Product Gamma Rays in the Range 0.1-0.4 Mev.

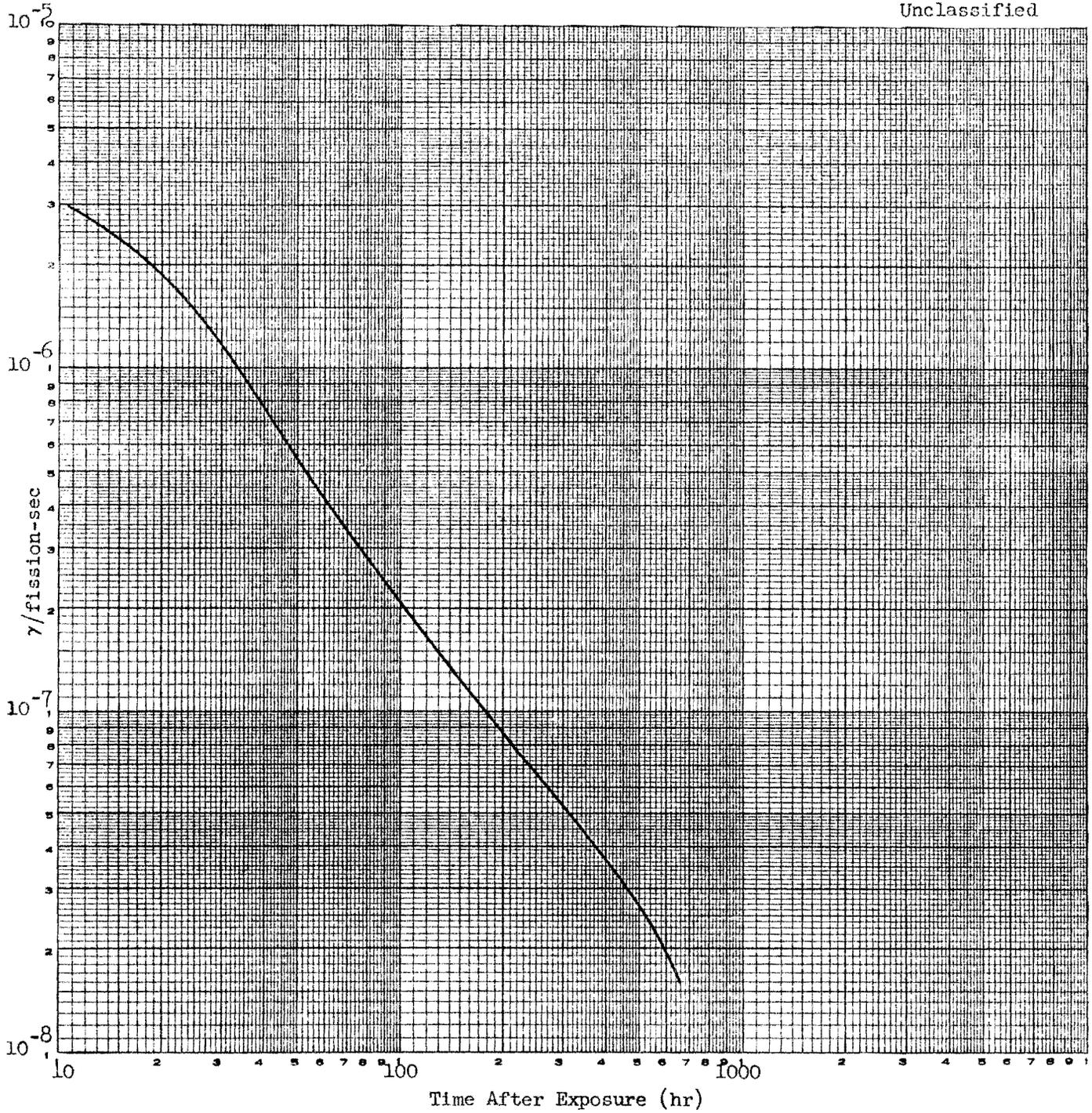


Fig. 4. Decay Curve for Fission-Product Gamma Rays in the Range 0.4-0.9 Mev.

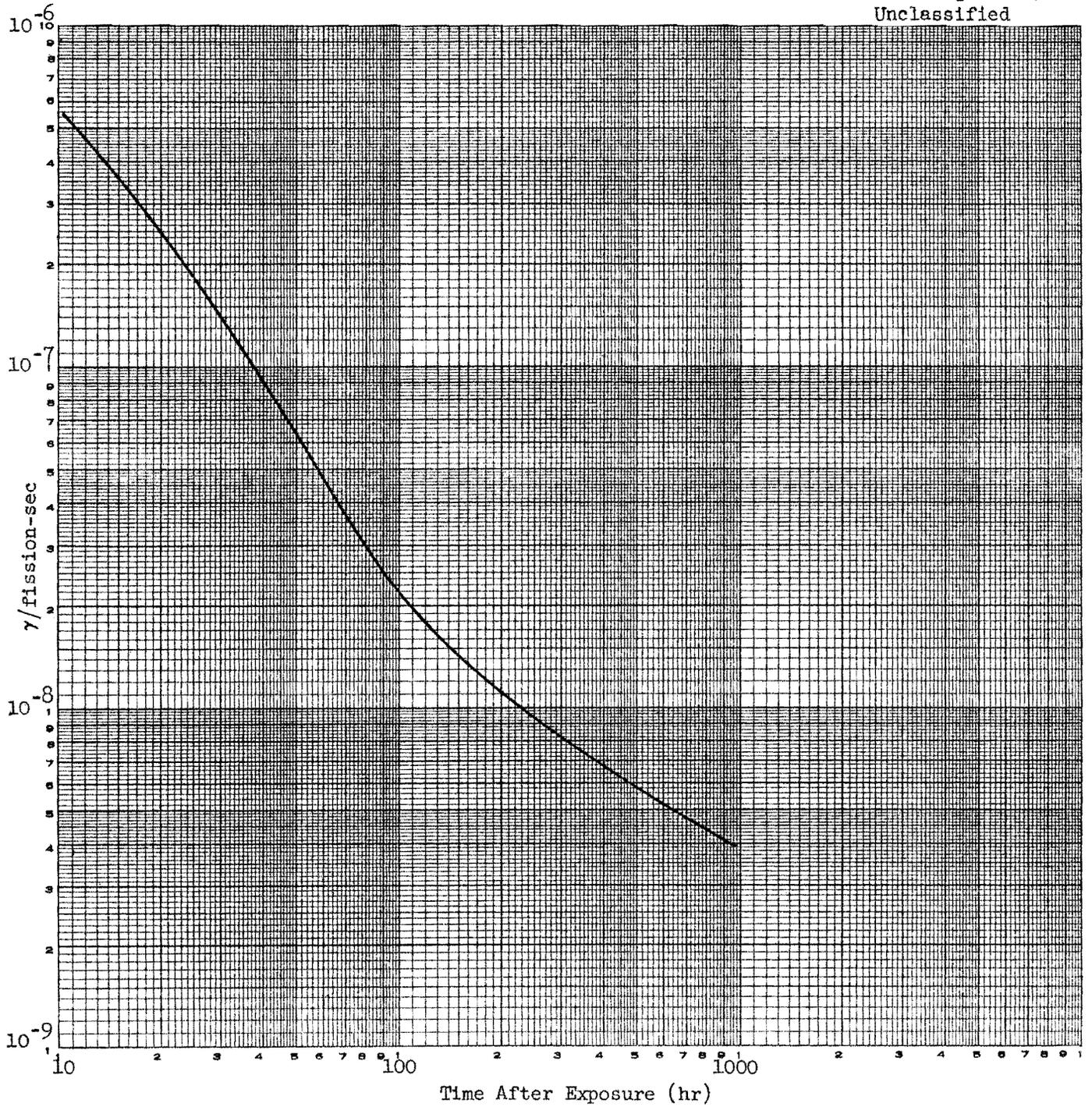


Fig. 5. Decay Curve for Fission-Product Gamma Rays in the Range 0.9-1.35 Mev.

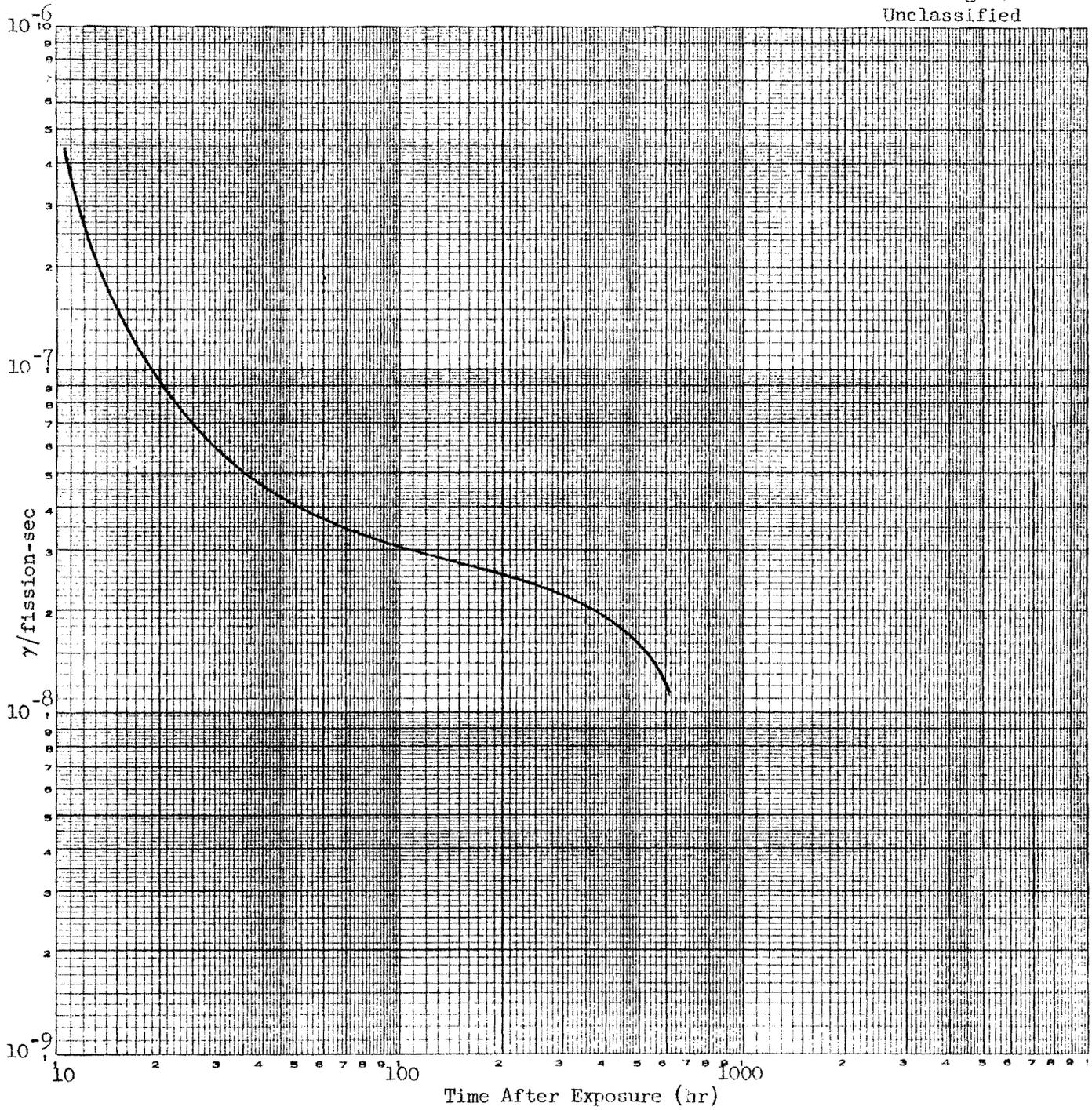


Fig. 6. Decay Curve for Fission-Product Gamma Rays in the Range 1.35-1.8 Mev.

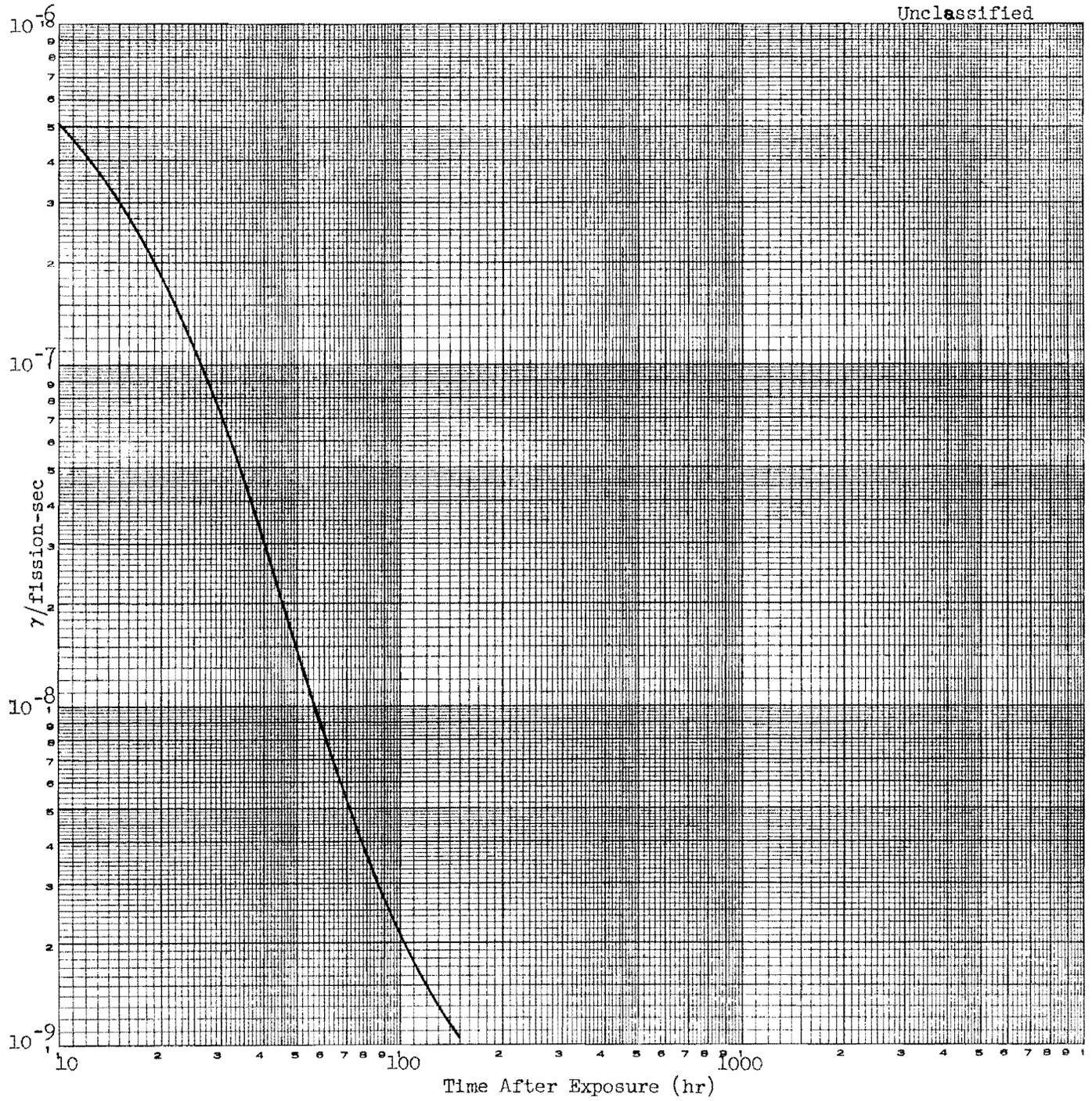
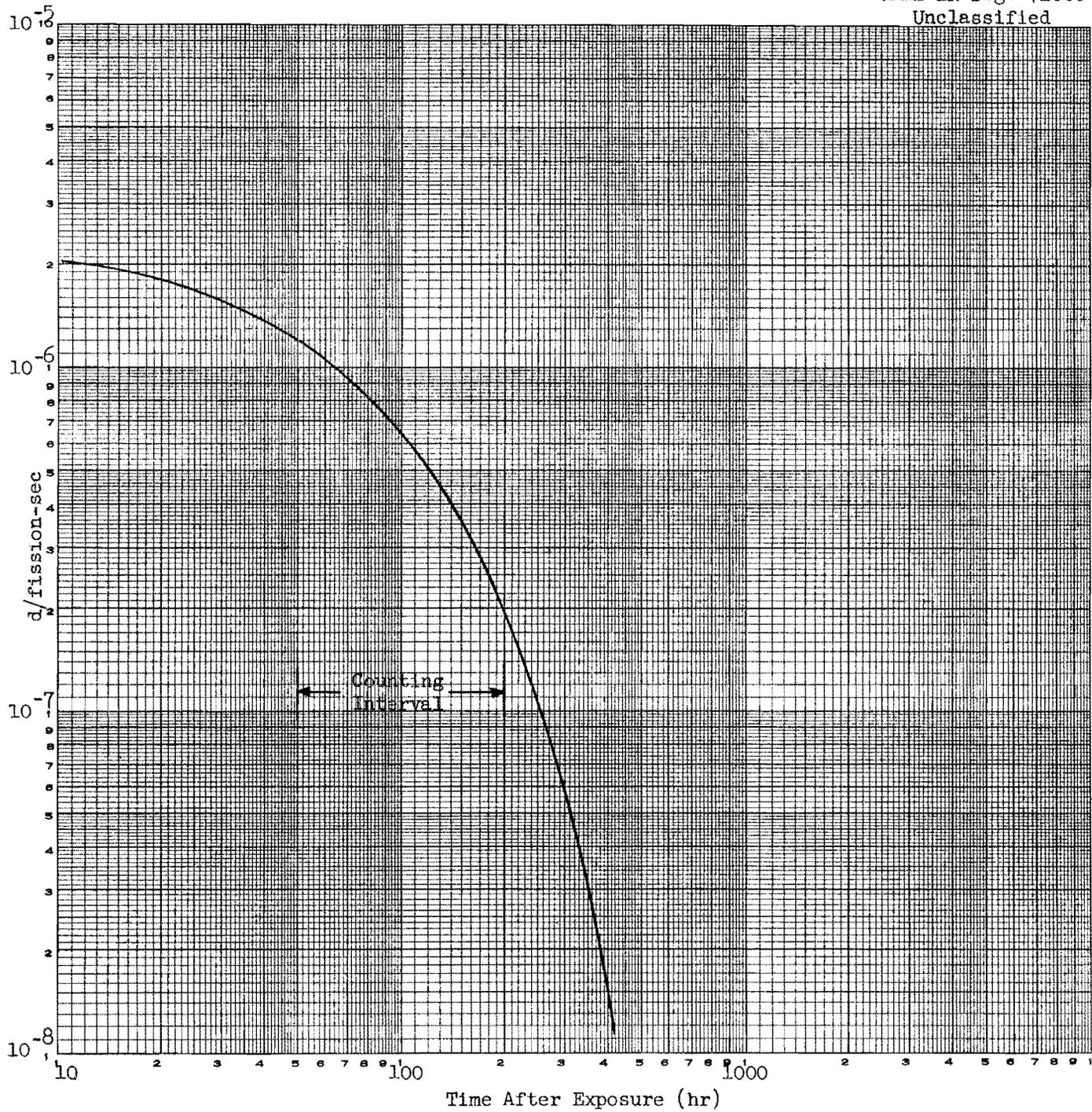
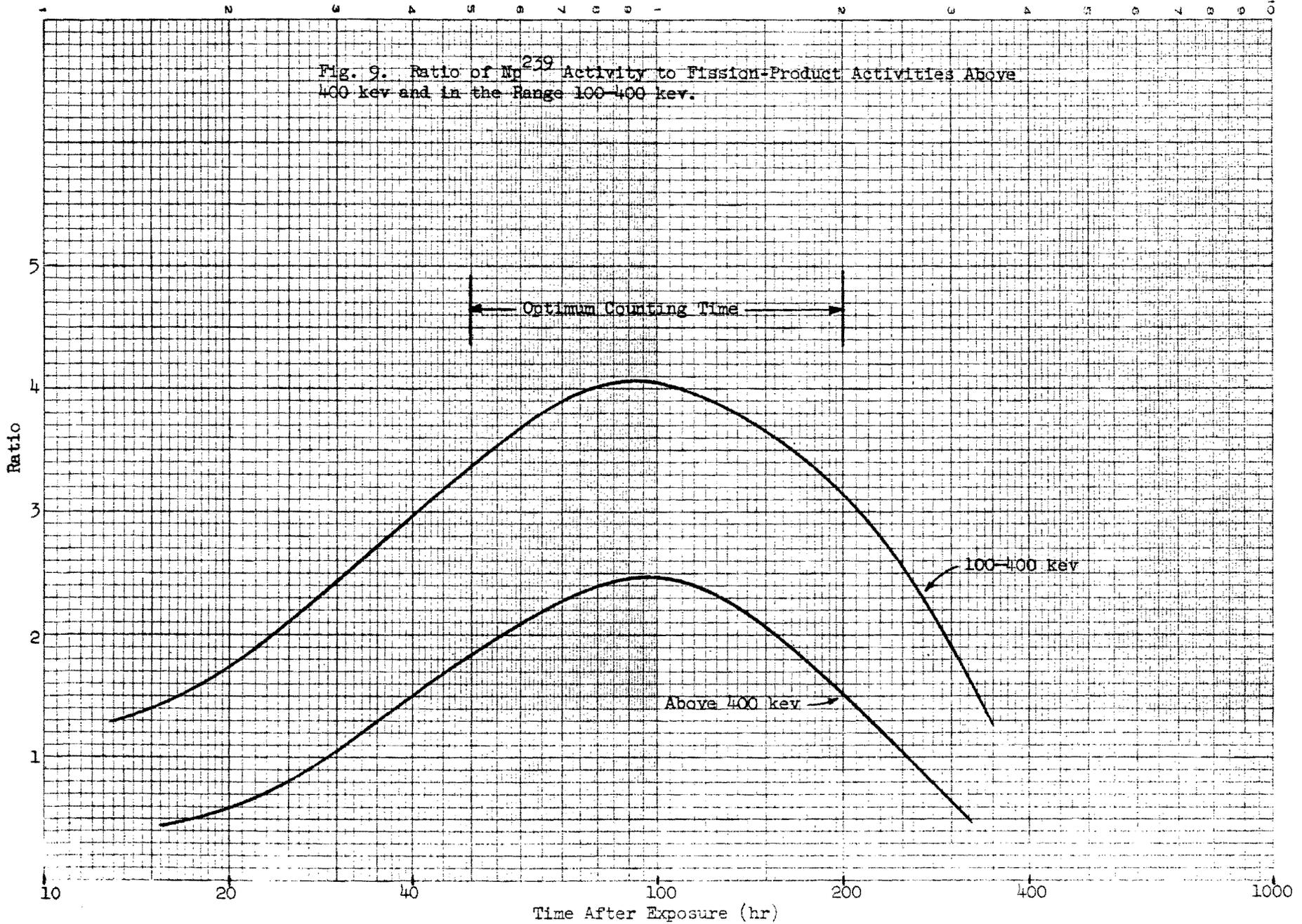


Fig. 7. Decay Curve for Fission-Product Gamma Rays in the Range 1.8-2.2 Mev.

Fig. 8. Disintegration Curve for  $\text{Np}^{239}$ .



result may be seen by referring to Eq. (4). Due to the decay and shift in spectrum of the activities observed each factor on the right-hand side of this equation will be a function of the time elapsed since the irradiation. Since, however,  $P_{39}/F_{25}$  is independent of time the variation of  $L_c/L_s$  and  $E_2^{25}/E_1^{39}$ , the principal terms, almost exactly cancel to form a time-independent product. If the counting is done over a period of time when  $L_c/L_s$  is nearly constant, then  $E_2^{25}/E_1^{39}$  will also be nearly constant and errors in the time scales of the two separate counting experiments determining these factors will have less impact on the over-all accuracy of the measurement. All irradiations should be made at the same time, including those in the pure thermal flux, and the resulting four samples should be counted alternately several times over the period from 50 to 200 hr after the irradiations. From the curve of  $Np^{239}$  decay shown in Fig. 8, the neptunium disintegration rate during the counting period is shown to vary from  $10^{-6}$  to  $2 \times 10^{-7}$  d/sec per fission in the sample.

In order to determine the necessary exposure time, we note from Eq. (4) that

$$\frac{\Delta(P_{39}/F_{25})}{P_{39}/F_{25}} \approx \frac{\Delta L_c}{L_c} + \frac{\Delta L_s}{L_s} + \frac{\Delta(E_1^{39}/E_2^{25})}{E_1^{39}/E_2^{25}} \quad (18)$$

and from Eq. (9), if the two enrichments differ enough to prevent a severe subtraction, then

$$\frac{\Delta(E_1^{39}/E_2^{25})}{E_1^{39}/E_2^{25}} \approx \frac{\Delta L_c}{L_c} + \frac{\Delta L_s}{L_s} \quad (19)$$

Speaking approximately, one could thus say that the over-all experimental error is roughly four times the percentage error in the coincidence or singles gamma count. If an over-all accuracy of 1% is desired for each determination, then the statistical accuracy of  $L_c$  and  $L_s$  must be about 0.25%, requiring  $1.6 \times 10^5$  counts. An average counting time of two hours should not be unreasonable since this would allow the

counting of all samples in an 8-hr shift. The counting period would cover a 6-day interval and this would allow each sample to be counted a maximum of 18 times which is certainly more than necessary. The final accuracy is, of course, improved by counting each sample as many times as possible.

The average counting rate implied by the above considerations is 1300 counts per minute, or 22 counts per second. From Fig. 8, the average disintegration rate of neptunium in the counting interval is found to be  $5.5 \times 10^{-7}$  d/sec per fission, so that the number of fissions required (at the EGCR enrichment of 2.46%) is

$$F = \frac{4 \times 10^7}{\omega_c \epsilon y}, \quad (20)$$

where

$\omega$  = effective fraction of a  $4\pi$  solid angle subtended by the coincidence counters,

$\epsilon$  = detection efficiency of the crystal,

$y$  = yield per disintegration of the observed activity.

In terms of the incident neutron flux on a sample of enrichment,  $e$ , containing  $m$  grams of uranium, we can write

$$F = \Sigma_f V \phi t = \left(\frac{m e}{235}\right) (0.6023) \sigma_f \phi t. \quad (21)$$

Combining Eqs. (20) and (21) we obtain the incident flux required for the desired counting rate,

$$\phi t = \frac{2.7 \times 10^7}{m e \omega_c \epsilon y} \text{ n/cm}^2. \quad (22)$$

We now evaluate Eq. (22) for the special case of a 0.005-in.-thick uranium foil cut to the outline of the EGCR fuel pellet (0.705 in. o.d., 0.323 in. i.d. For the required factors we obtain the following values:

1. For the total mass,  $m = 0.48$  grams.
2. For gamma energies near 100 kev, a detection efficiency near unity should be possible since the total attenuation cross section for NaI at this energy equals  $5.8 \text{ cm}^{-1}$ . We pick, conservatively,  $\epsilon = 0.5$ .

3. The yield of the 103-kev x-ray in coincidence with the 106.2-kev gamma was discussed earlier. The result is  $y = 0.30$ .

4. We assume that each counter subtends a fraction 0.3 of the total solid angle. For coincidence counting with an isotropic angular correlation the resulting effective solid angle is twice the square of this value. We obtain  $\omega_c = 0.18$ .

5. For the enrichment,  $e = 0.0246$   
The resulting exposure obtained with Eq. (22) is

$$\phi t = 0.85 \times 10^{11} \text{ n/cm}^2 . \quad (23)$$

At a flux level of  $5 \times 10^7 \text{ n/cm}^2 \cdot \text{sec}$  this exposure will be reached in 1/2 hr.

#### Test of Counting Equipment and Experimental Method

In order to minimize the possibility of failure it would be desirable to make a prior check of the equipment and experimental method. This would, in addition, give a more accurate basis for estimating the exposure time than the calculation presented above. This check would be amply provided by performing the experiment in the lattice of the Oak Ridge Graphite Reactor (OGR). In addition to checking the equipment and procedures the measurement would, in fact, be of distinct interest in itself.

APPENDIX

## Summary of Nomenclature

$L_c$	Coincidence counting rate of the 106.2-keV decay gamma ray and the 103-keV internal conversion X-ray of $\text{Pu}^{239}$ .
$L_s$	Singles counting rate for fission-product gamma rays at energies above a selected level.
$P_{39}$	Production rate of $\text{Np}^{239}$ .
$F_{25}$	Fission rate of $\text{U}^{235}$ .
$F_{28}$	Fission rate of $\text{U}^{238}$ .
$E_1^{39}, E_1^{25}$	Proportionality factors for the coincidence counting of gamma rays at 100 keV. These factors include the time decay of the activities as well as the counting efficiencies of the crystals and solid angles subtended by the counters. $E_1^{39}$ refers to activities in the $\text{Np}^{239}$ decay chain and $E_1^{25}$ refers to activities in the $\text{U}^{235}$ fission-product decay.
$E_2^{28}, E_2^{25}$	Proportionality factors for the singles counting of fission product gamma rays from $\text{U}^{238}$ and $\text{U}^{235}$ at energies above a selected level.
$\delta$	A ratio of counting rates defined by Eq. (3).
$\Sigma_{28}$	Effective macroscopic absorption cross section of $\text{U}^{238}$ in a pure thermal flux.
$\Sigma_{f,25}$	Effective macroscopic fission cross section of $\text{U}^{235}$ in a pure thermal flux.
$e$	Sample enrichment, uranium wt % of $\text{U}^{235}$ .
$f$	Sample self-shielding factor.
$\alpha$	Ratio of $\text{U}^{235}$ fission rate for samples of two different enrichments.
$F$	Total number of fissions in sample.
$\omega_c$	Effective fraction of $4\pi$ solid angle subtended by coincidence counters.

## APPENDIX (contd.)

$\epsilon$	Detection efficiency of crystal.
$y$	Yield per disintegration of observed activity.
$V$	Sample volume.
$t$	Length of sample irradiation, sec.
$\Sigma_f$	Macroscopic fission cross section of sample.
$\phi$	Effective thermal flux at sample position.
$m$	Mass of sample.

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