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April 5, 1972

M E M O R A N D U M

TO: S. MIRSHAK

FROM: W. C. MOSLEY *WC Mosley*

ANALYSIS OF PRODUCING  $^{238}\text{Pu}$  LOW IN  $^{236}\text{Pu}$   
BY RECOIL DURING IRRADIATION OF  $^{237}\text{Np}$

Introduction

High purity  $^{238}\text{Pu}$  (<0.3 ppm  $^{236}\text{Pu}$ ) is required for heat sources in applications requiring power at low radiation levels, especially for in vivo bio-medical devices. <sup>(1)</sup> A primary source of gamma radiation is the daughters of  $^{236}\text{Pu}$  formed in the production of  $^{238}\text{Pu}$ . Acceptable  $^{238}\text{Pu}$  can be produced by irradiation of  $^{241}\text{Am}$  to form  $^{242}\text{Cm}$  which decays to  $^{238}\text{Pu}$  with only 1.8 ppb  $^{236}\text{Pu}$  impurity; however,  $^{241}\text{Am}$  may not be available in sufficient quantity to meet demands for  $^{238}\text{Pu}$ . Irradiation of the more abundant  $^{237}\text{Np}$  to produce  $^{238}\text{Pu}$  produces  $^{236}\text{Pu}$  by ( $\gamma, n$ ) and ( $n, 2n$ ) reactions. Special assemblies and irradiations to minimize fast neutrons and gammas have succeeded in reducing the  $^{236}\text{Pu}$  concentration to 0.3 ppm, still a fairly high concentration for medical applications. This analysis was undertaken to learn whether recoil distances of  $^{236}\text{Pu}$  precursors formed by high energy reactions are sufficiently long, compared to the recoil distances of  $^{238}\text{Pu}$  precursors, that  $^{236}\text{Pu}$  might be selectively captured in a second phase in a heterogeneous target and subsequently separated from  $^{238}\text{Pu}$  product.

Summary

Results of this analysis indicate that a recoil separation technique would require neptunium targets in the form of 100Å particles of  $\text{NpO}_2$  or 50Å Np metal foils to effect a two-fold reduction in  $^{236}\text{Pu}$  by collection in a surrounding matrix, such as aluminum. While such targets might possibly be fabricable, they are probably not subsequently separable from the matrix phase containing  $^{236}\text{Pu}$ ; damage from induced fission during irradiation would degrade the target and prevent physical separation from the matrix phase.

Discussion

Since the  $^{236}\text{Pu}$  impurity in  $^{238}\text{Pu}$  product of  $^{237}\text{Np}$  irradiations is formed by high energy nuclear reactions, the possibility arises of a separation process based on the recoil distances of  $^{236}\text{Pu}$  precursors being significantly larger than that of  $^{238}\text{Pu}$  precursors. A separation might be effected by surrounding the  $^{237}\text{Np}$  target with a matrix to trap the recoiling  $^{236}\text{Pu}$  outside the target phase. The  $^{237}\text{Np}$  target containing  $^{238}\text{Pu}$  with reduced  $^{236}\text{Pu}$  content could then be separated from the matrix material by a chemical or physical process. This separation technique requires that the  $^{237}\text{Np}$  targets maintain their integrity during fabrication and irradiation.

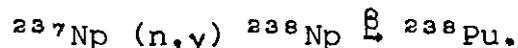
The recoil energies of  $^{236}\text{Pu}$  and  $^{238}\text{Pu}$  were determined for the significant nuclear reactions that occur during production of  $^{238}\text{Pu}$ . The  $^{236}\text{Pu}$  recoil yields from  $\text{NpO}_2$  particle and Np metal foil targets were calculated. The target sizes necessary to effect significant separation of  $^{236}\text{Pu}$  from  $^{238}\text{Pu}$  product were compared with the extent of damage to the targets from induced fission.

Nuclear Events in  $^{238}\text{Pu}$  Production

The significant nuclear reactions involved in irradiating  $^{237}\text{Np}$  to produce  $^{238}\text{Pu}$  are  $(n,\gamma)$ ,  $(\gamma,n)$ ,  $(n,2n)$  and fission. The recoil energies for the first three reactions, listed in Table I, were calculated using the principles of conservation of momentum and known or estimated reaction energies.<sup>(2)</sup> Recoil energies are highest for  $(n,2n)$  reactions and least for  $(n,\gamma)$  reactions. The recoil ranges in  $\text{NpO}_2$  and Np metal (Table I) were determined by comparison with range-energy relations for  $^{237}\text{Np}$  published by Kaplan<sup>(3)</sup> and shown in Figure 1. Recoil ranges in  $\text{NpO}_2$  ( $\rho = 11.1\text{g/cc}$ ) are about 30% longer than in Np metal ( $\rho = 20.4\text{g/cc}$ ).

1. Production of  $^{238}\text{Pu}$  by  $(n,\gamma)$  reaction

$^{238}\text{Pu}$  is produced by capture of thermal neutrons by the following reaction:



The largest recoil of the  $^{238}\text{Np}$  nucleus occurs on emission of the gamma ray after neutron capture. The compound nucleus receives essentially no recoil from thermal neutron capture since the neutron energy is only

0.025 ev. Prompt gammas are emitted with energies ranging up to 6.5 Mev (Figure 2).<sup>(4)</sup> The recoil energy imparted to a nucleus of mass number A from emission of a gamma with energy  $E_\gamma$  is given by:

$$E_R^\gamma = \frac{(5.3 \times 10^{-10})(E_\gamma^2)}{A}.$$

Therefore, the  $^{238}\text{Pu}$  nuclei recoil from gamma emission with energies up to 90 ev.

$^{238}\text{Np}$  decays to  $^{238}\text{Pu}$  by emission of a 1.25 Mev  $\beta$ -particle. The recoil energy corresponding to emission of a  $\beta$ -particle with energy  $E_\beta$  is:

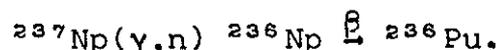
$$E_R^\beta \leq \frac{E_\beta}{1840A} \left[ 1 + \frac{E_\beta}{Mc^2} \right].$$

Maximum recoil energy from  $\beta$ -decay of  $^{238}\text{Np}$  is 12 ev.

The  $^{238}\text{Pu}$  recoil energy primarily from prompt gamma emission, is sufficient to break chemical bonds; however,  $^{238}\text{Pu}$  nuclei will recoil only several angstroms. Since higher energy reactions involved in  $^{236}\text{Pu}$  production will also break chemical bonds, a chemical separation of  $^{236}\text{Pu}$  from  $^{238}\text{Pu}$  based on selective breakage of chemical bonds, the Szilard-Chalmers effect,<sup>(2)</sup> will not be possible.

## 2. Production of $^{236}\text{Pu}$ by $(\gamma, n)$ Reaction

Significant quantities of  $^{236}\text{Pu}$  can be produced by the photonuclear  $(\gamma, n)$  reaction with gamma energies above a threshold energy of 6.5 Mev according to the reaction:



The emission of a neutron by the excited  $^{237}\text{Np}$  nucleus is the predominant source of recoil energy. The energy of the neutron is equal to the energy of the incident gamma ray in excess of the threshold energy of 6.5 Mev. The recoil will therefore depend on the gamma energy spectrum, which is influenced by reactor components such as aluminum.

Three prompt gamma rays<sup>(5)</sup> from thermal neutron capture by  $^{27}\text{Al}$  with unattenuated energies of 6.72, 7.21 and 7.72 Mev (Figure 3) will predominate in producing  $^{236}\text{Pu}$ . The emitted neutrons will have energies of 0.22, 0.71 and 1.22 Mev.

$^{237}\text{Np}$  recoil energies are related to neutron energies by:

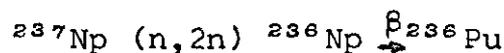
$$E_R^n = \frac{E_n}{A}.$$

The  $^{237}\text{Np}$  recoil energies from  $(\gamma, n)$  reactions are, then, 0.93, 3.0, and 5.1 kev. Corresponding recoil ranges and the fractions of nuclei with each energy, as determined by the relative intensities of the prompt gamma rays, are given in Table I.

The recoil energies imparted to the target nuclei by the incident gamma rays ( $E_\gamma = 6.5\text{--}10$  Mev) and by beta emission ( $E_\beta = 0.52$  Mev) are 90–200 ev and 2.4 ev, respectively. These energies are insignificant compared to recoil energies from neutron emission.

### 3. Production of $^{236}\text{Pu}$ by $(n, 2n)$ Reaction

Capture of fast fission neutrons with energies greater than the neutron binding energy ( $\sim 6.5$  Mev) produces  $^{236}\text{Pu}$  by the following reaction:



The amount of  $^{236}\text{Pu}$  produced by  $(n, 2n)$  reactions will depend on the neutron spectrum at the target position. The largest recoil is imparted to the compound neptunium nuclei upon capture of fast neutrons. Recoil from the emission of two neutrons is not insignificant, but is of secondary importance. Since the energies of the captured neutrons must be greater than the threshold energy of the reaction at 6.5 Mev, the recoil energy will be  $E_r > 27$  kev.

The two neutrons emitted by the compound nucleus to form  $\text{Np}^{236}$  will share the energy of the captured fast neutron that is in excess of the 6.5 Mev neutron binding energy. Their energies will range from zero to several Mev. For these calculations, each neutron was assumed to be emitted with 1 Mev of energy. Depending on the neutron angular correlation, the  $^{236}\text{Np}$  will receive additional recoil energy varying from 0 to about 8 kev.

Decay of  $^{236}\text{Np}$  to  $^{236}\text{Pu}$  by  $\beta$ -emission results in a recoil energy of 2.4 ev, which is insignificant. Thus, the minimum recoil energy for fast neutron capture will be  $\sim 27$  kev.

### 4. Fission of $^{237}\text{Np}$

Fission can cause extensive damage in insulating materials like  $\text{NpO}_2$  and in thin metallic foils.  $^{237}\text{Np}$  targets are usually irradiated to about 10% burnup. The fission cross sections of  $^{237}\text{Np}$  and  $^{238}\text{Np}$ , the precursors of  $^{238}\text{Pu}$ , are sufficiently high that fissioning occurs with about the same probability as neutron capture. About  $10^6$  fission and neutron capture events occur for each  $^{236}\text{Pu}$  producing event. Induced fission will produce two heavy and highly energetic, charged particles for each event. Fission fragment damage appears as tracks about  $100\text{\AA}$  wide and  $10\ \mu$  ( $10^6\text{\AA}$ ) in length in which the structure of the material has become highly disordered<sup>(6)</sup> and, on this scale, would intermix target material with the surrounding matrix phase.

## $^{236}\text{Pu}$ Recoil from Spherical Particles and Foil

The amount of  $^{236}\text{Pu}$  that will be ejected from a particle or foil target was estimated by integrating the probability of ejection from a position within the target over the volume of the target.

### 1. Spherical Target (Figure 4)

The probability that a nucleus with range  $R$  will escape on recoil from a position at radius  $r$  in a spherical target particle of radius  $a$ , is  $P(r) = \frac{1}{2}(1 - \cos \theta)$ , where  $\theta$  is the angle between the radius through the nucleus and the intersection of a sphere of radius  $R$  with particle surface (Figure 4a). The probability varies with position in the particle and size of the particles as shown in Figure 4b. The fraction of  $^{236}\text{Pu}$  ejected from the  $^{237}\text{Np}$  target particle is given by:

$$^{236}\text{Pu ejected} = 3/a^3 \int_0^a P(r)r^2 dr.$$

The fraction of  $^{236}\text{Pu}$  ejected from  $(n,2n)$  and  $(\gamma,n)$  reactions as a function of target particle size is shown in Figure 5. Because of higher recoil energy the amount ejected by  $(n,2n)$  reactions is significantly higher than from  $(\gamma,n)$  reactions for a given target diameter or thickness.

Figure 6 shows the ejected fraction to be expected for several particle sizes as a function of relative amounts of  $^{236}\text{Pu}$  produced by the two reactions. The fraction of  $^{236}\text{Pu}$  produced by  $(\gamma,n)$  reaction varies from 50% in the core<sup>(7)</sup> to near 90% in the reflector of the reactor.<sup>(8)</sup> A 50% reduction in  $^{236}\text{Pu}$  content within the target will therefore require target particles with diameters of about 100Å or less.

### 2. Foil Targets (Figure 7)

The probability of a nucleus being ejected from a foil is:

$$P = \frac{1}{2} [(1 - \cos \theta_1) + (1 - \cos \theta_2)]$$

where  $\theta_1$  and  $\theta_2$  are the angles between a line perpendicular to the surfaces and the intersections of a sphere of radius  $R$  with the surfaces of the foil (Figure 7a). This probability varies with position in the foil and foil thickness  $t$  as shown in Figure 7b. The fraction of  $^{236}\text{Pu}$  ejected is:

$$^{236}\text{Pu Ejected} = 1/t \int_0^t P(\tau) d\tau,$$

where  $\tau$  is the distance from surface of the foil.

The fraction of  $^{236}\text{Pu}$  that recoils from neptunium metal foils is shown in Figure 8 as a function of foil thickness for  $(n,2n)$  and  $(\gamma,n)$  reactions. Again more  $^{236}\text{Pu}$  would be ejected by  $(n,2n)$  reactions than by  $(\gamma,n)$  reactions.

The  $^{236}\text{Pu}$  ejected for several foil thicknesses as a function of the relative amounts of  $^{236}\text{Pu}$  produced by  $(\gamma, n)$  and  $(n, 2n)$  reactions is shown in Figure 9. Since  $(\gamma, n)$  reactions are estimated to predominate, (7,8) significant reduction in  $^{236}\text{Pu}$  content (>50%) will require foil targets with thicknesses of  $50\text{\AA}$  or less.

### Conclusions

A 50% reduction of  $^{236}\text{Pu}$  in  $^{238}\text{Pu}$  product by matrix trapping of recoiling nuclei from  $(\gamma, n)$  and  $(n, 2n)$  events would require  $\text{NpO}_2$  particles of  $<100\text{\AA}$  diameter or  $\text{Np}$  foils  $<50\text{\AA}$  thick. The size of the  $\text{NpO}_2$  particle targets is so small that each particle would be completely disordered and intermixed with matrix material by a single fission event. Likewise, a fission event in  $50\text{\AA}$  thick foil would disorder the foil over a range larger than the recoil distance for a  $(\gamma, n)$  or  $(n, 2n)$  event. About  $10^6$  fissions will occur for each  $^{236}\text{Pu}$  producing event.

Thus, the target sizes required for significant reduction in  $^{236}\text{Pu}$  content by selective recoil appear to be too small for partial target fabrication and subsequent separation of matrix and target phases. Fissioning which occurs during production of  $^{236}\text{Pu}$  will prevent reduction in the  $^{236}\text{Pu}$  content by recoil separation since the damage from fission will degrade the small targets during irradiation and prevent their subsequent separation from the  $^{236}\text{Pu}$  trapping matrix.

WCM:rbw

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2. E. Broda, Advances in Radiochemistry, Cambridge (1950)
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5. Ibid, p. 127
6. P. B. Price and R. M. Walker, J. Appl. Phys. 33 3400 (1962)
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8. SRL Monthly Report, June 1970, DP-70-1-6, p. 9

TABLE I

RECOIL FROM NUCLEAR REACTIONS DURING PRODUCTION OF  $^{238}\text{Pu}$ 

<u>Reaction</u>	<u>Source of Energy</u>	<u>Recoiling Nucleus</u>	<u>Recoil Energy</u>	<u>Range in <math>\text{NpO}_2</math></u>	<u>Range in Np</u>
1. (n, $\gamma$ )	Thermal neutron capture (.025 ev)	$^{238}\text{Np}^*$	0	0	0
	Prompt gamma emission (E<6.5 Mev)	$^{238}\text{Np}$	$\leq 90$ ev	$< 1\text{\AA}$	$< 1\text{\AA}$
	$\beta$ -emission (E=1.25 Mev)	$^{238}\text{Pu}$	$\leq 20$ ev	$\sim 0\text{\AA}$	$\sim 0\text{\AA}$
2. ( $\gamma$ ,n)	Gamma capture (E=6.5-10 Mev)	$^{237}\text{Np}^*$	90-200 ev	$\sim 1\text{\AA}$	$\sim 1\text{\AA}$
	Neutron emission 0.22 Mev	$^{236}\text{Np}$ (24%)	0.9 kev	$5\text{\AA}$	$4\text{\AA}$
	0.71 Mev	$^{236}\text{Np}$ (43%)	3.0 kev	$15\text{\AA}$	$13\text{\AA}$
	1.22 Mev	$^{236}\text{Np}$ (33%)	5.1 kev	$30\text{\AA}$	$25\text{\AA}$
	$\beta$ -emission (E=0.52 Mev)	$^{236}\text{Pu}$	2.4 ev	$\sim 0\text{\AA}$	$\sim 0\text{\AA}$
3. (n,2n)	Fast neutron capture (E=6.5-10 Mev)	$^{238}\text{Np}^*$	27-41 kev	$120-180\text{\AA}$	$75-115\text{\AA}$
	Neutron emission (1 Mev)	$^{236}\text{Np}$	0-8 kev	$0-50\text{\AA}$	$0-37\text{\AA}$
	$\beta$ -emission	$^{236}\text{Pu}$	2.4 ev	$\sim 0\text{\AA}$	$\sim 0\text{\AA}$

\*Compound nucleus

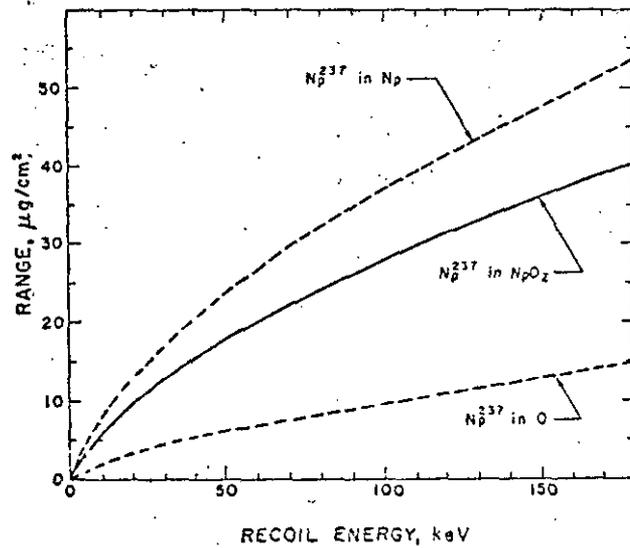


FIG. 1.—Range-energy relation for  $^{237}\text{Np}$  recoiling in  $\text{NpO}_2$ . The dashed curves give the corresponding ranges if the stopping medium consisted solely of neptunium atoms or oxygen atoms, respectively.

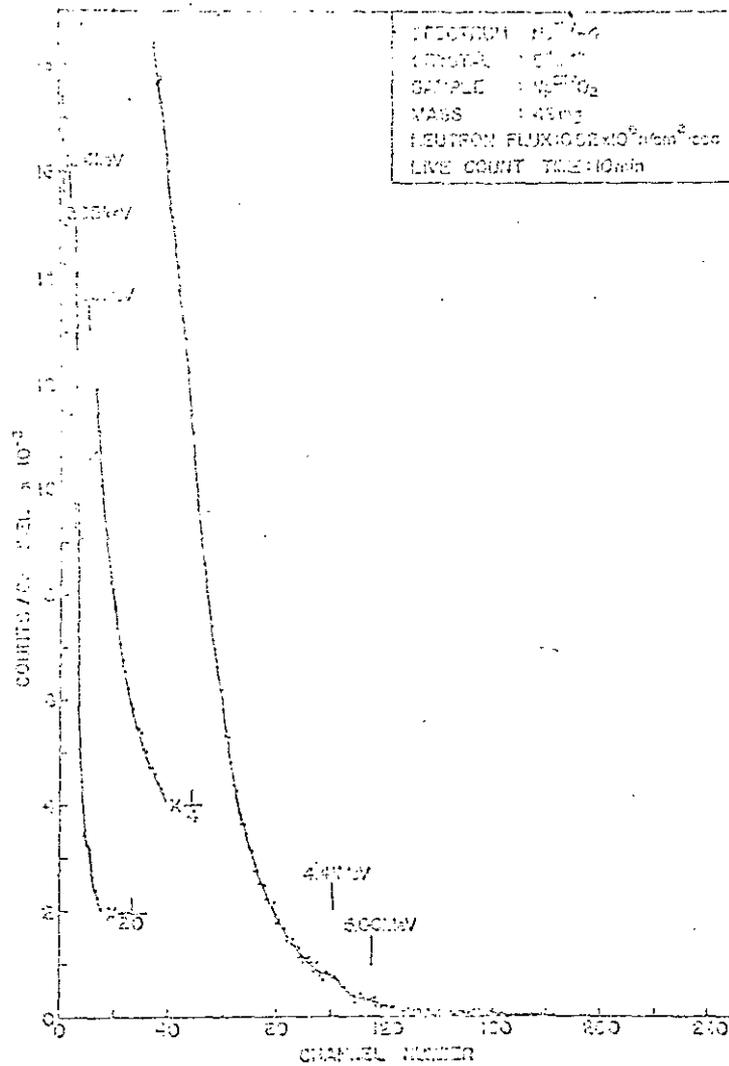


FIG 2. PROMPT GAMMA RAYS FROM THERMAL NEUTRON CAPTURE BY  $^{237}\text{Np}$

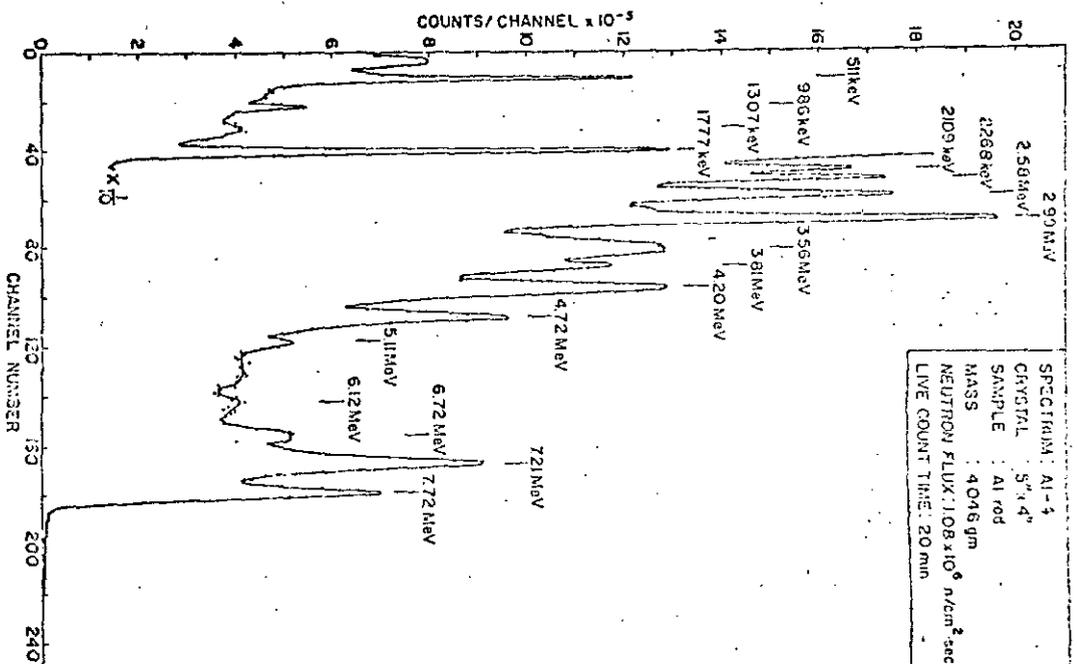


FIG 3. PROMPT GAMMA RAYS FROM CAPTURE OF THERMAL NEUTRONS BY 27Al

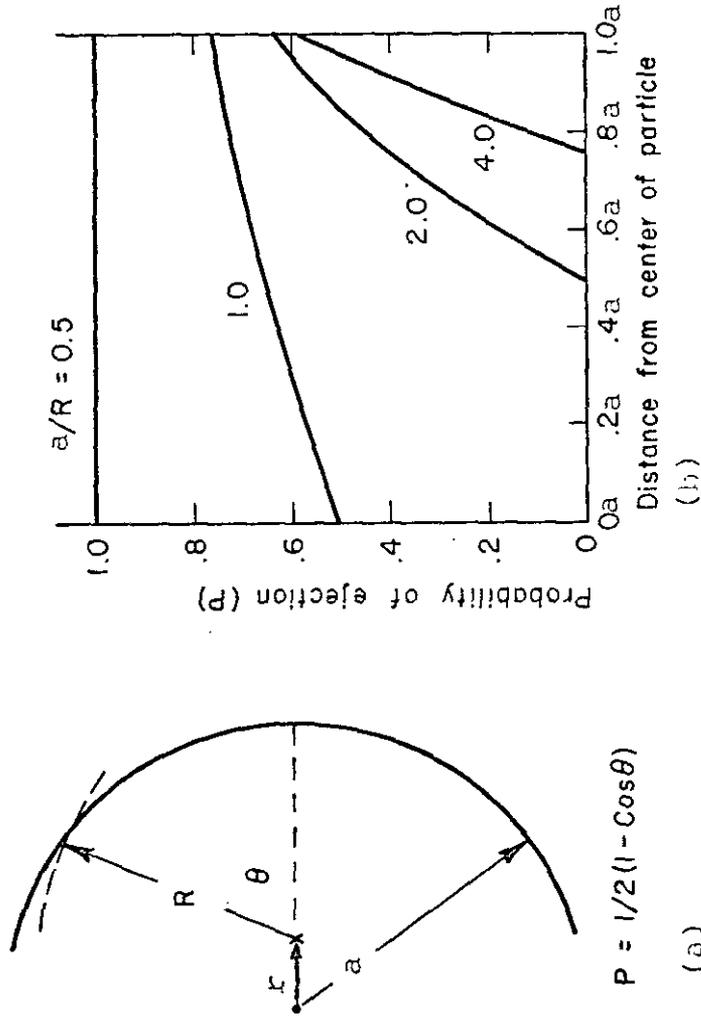


FIG 4. PROBABILITY OF RECOIL EJECTION FROM A SPHERICAL PARTICLE (a) RANGE DIAGRAM: (b) VARIATION WITH RATIO OF PARTICLE RADIUS (a) TO RECOIL RANGE (R)

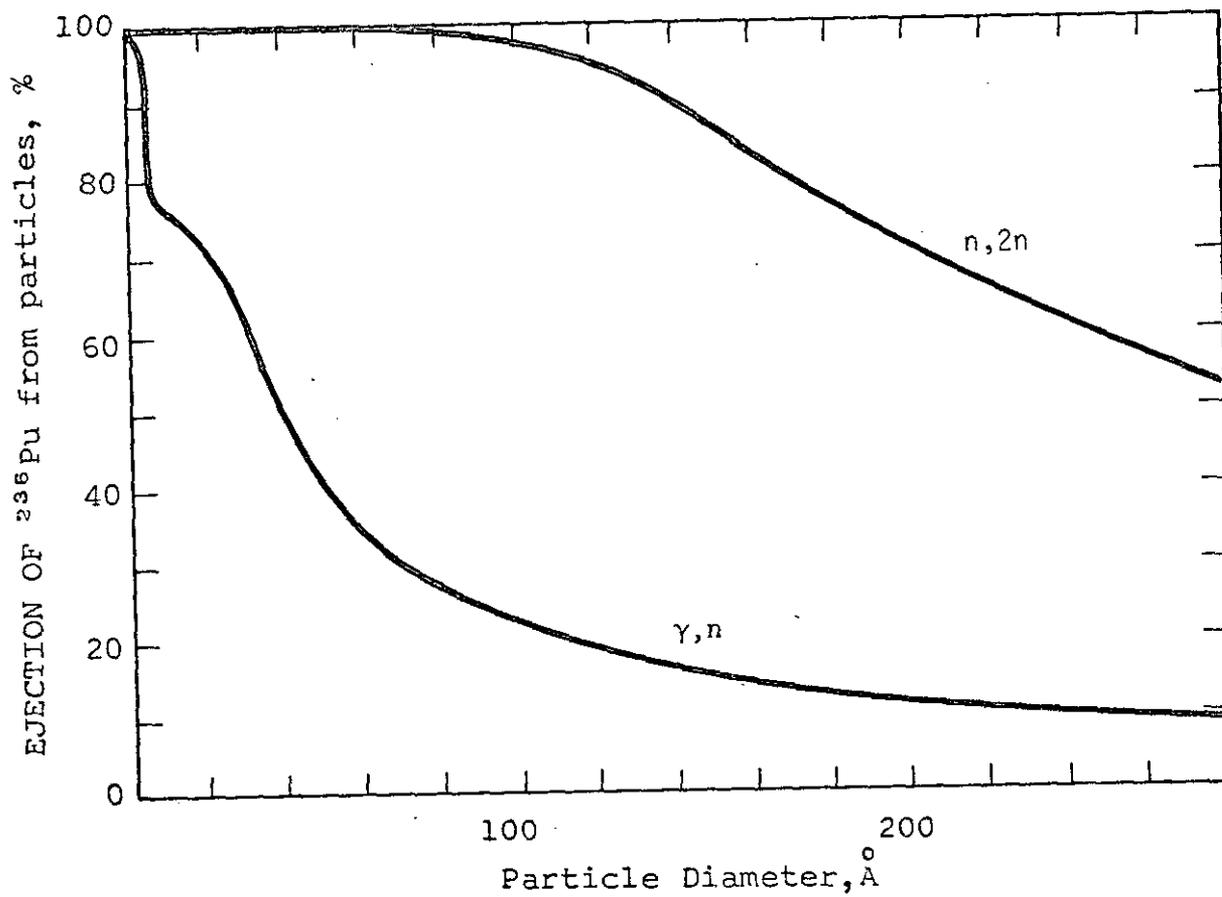


FIG 5. RECOIL EJECTION OF  $^{236}\text{Pu}$  PRODUCED BY  $(\gamma, n)$  AND  $(n, 2n)$  REACTIONS AS A FUNCTION OF  $\text{NpO}_2$  PARTICLE DIAMETER

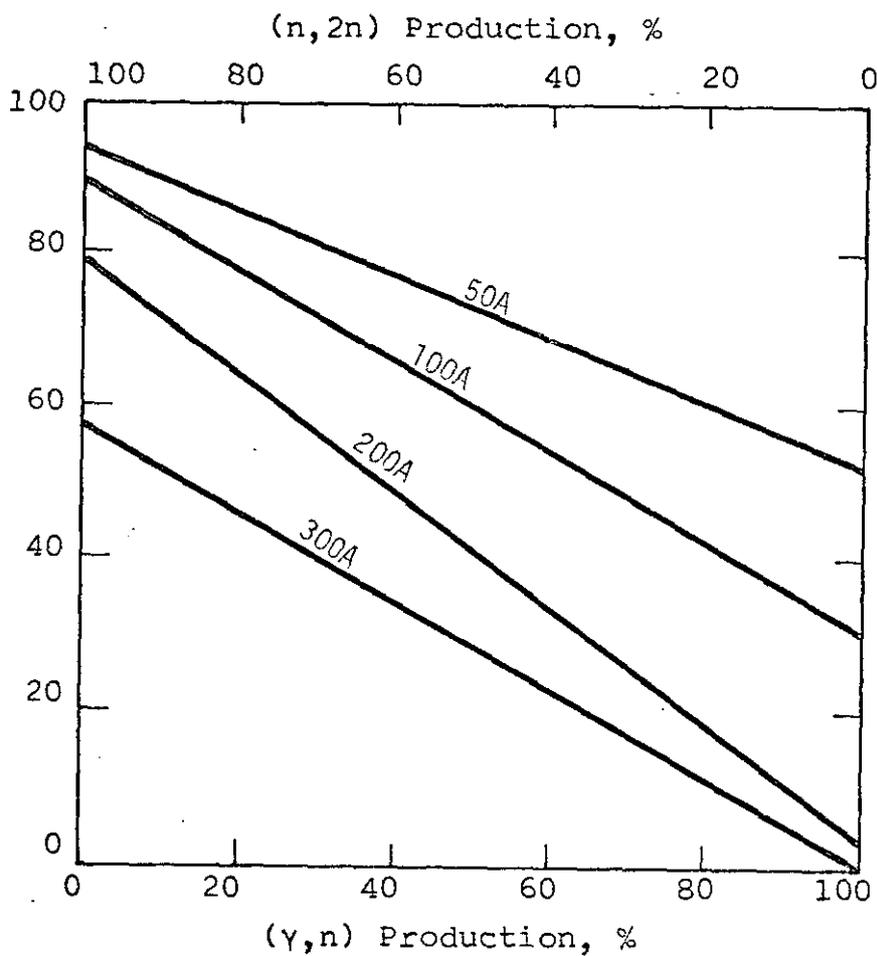
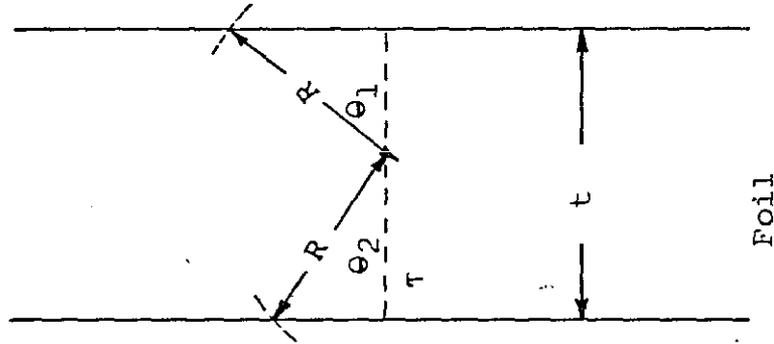
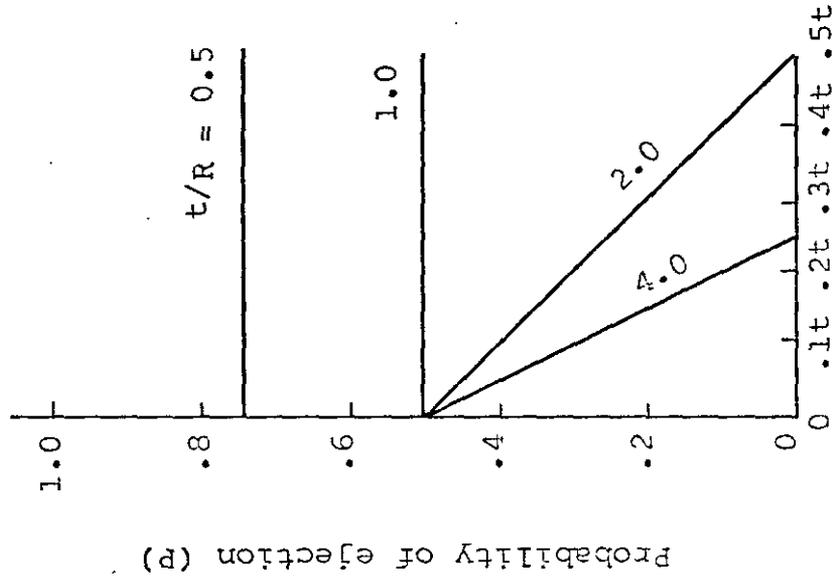


FIG 6. EJECTION OF  $^{238}\text{Pu}$  BY RECOIL FROM  $\text{NpO}_2$  PARTICLES OF INDICATED DIAMETERS AS A FUNCTION OF RELATIVE AMOUNTS OF PRODUCTION BY  $(\gamma,n)$  AND  $(n,2n)$  REACTIONS



$$P = \frac{1}{2}(1 - \cos \theta_1) + \frac{1}{2}(1 - \cos \theta_2)$$

(a)



Distance from surface of foil  
(b)

FIG 7. PROBABILITY OF RECOIL EJECTION FROM A FOIL; (a), RANGE DIAGRAM; (b), VARIATION WITH RATIO OF FOIL THICKNESS(t) TO RECOIL RANGE (R)

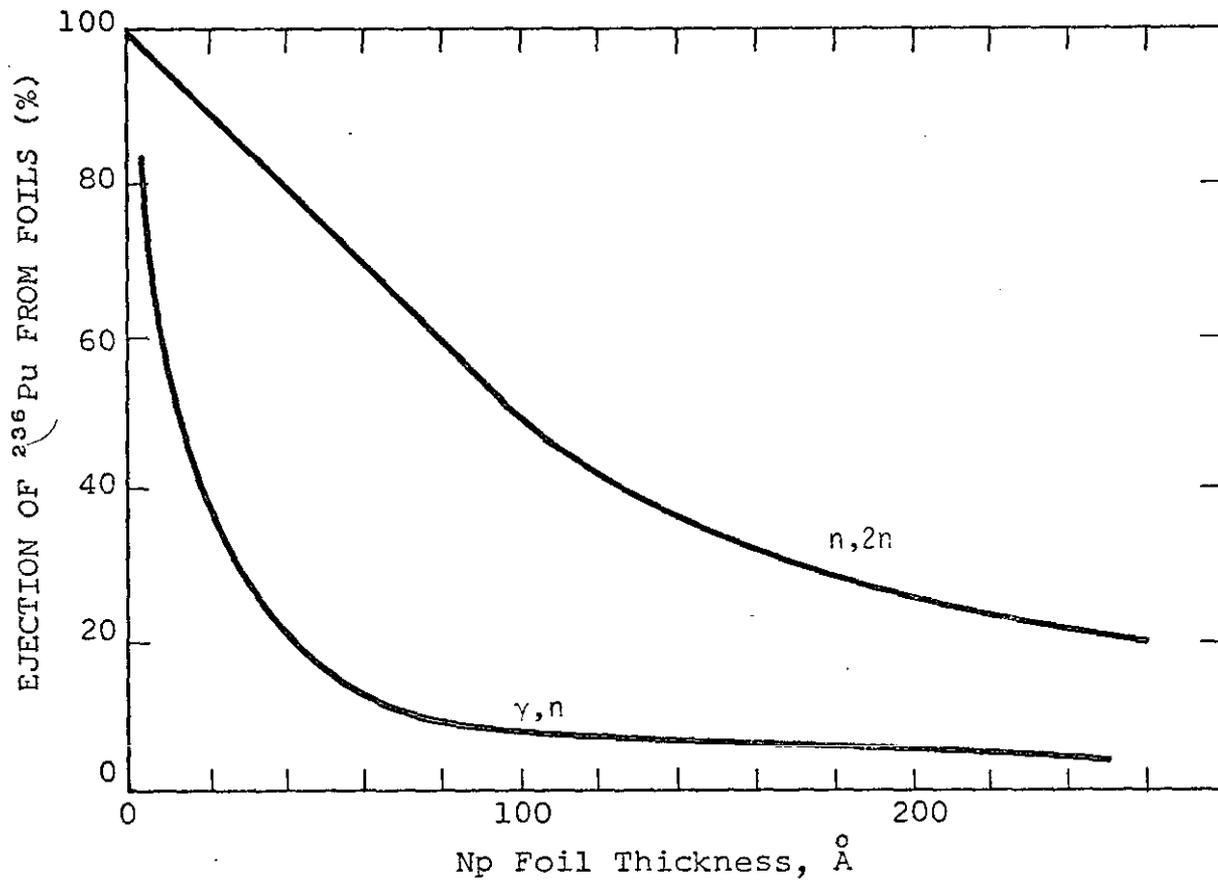


FIG 8. RECOIL EJECTION OF  $^{236}\text{Pu}$  PRODUCED BY  $(\gamma, n)$  AND  $(n, 2n)$  REACTIONS AS A FUNCTION OF  $\text{Np}$  FOIL THICKNESS

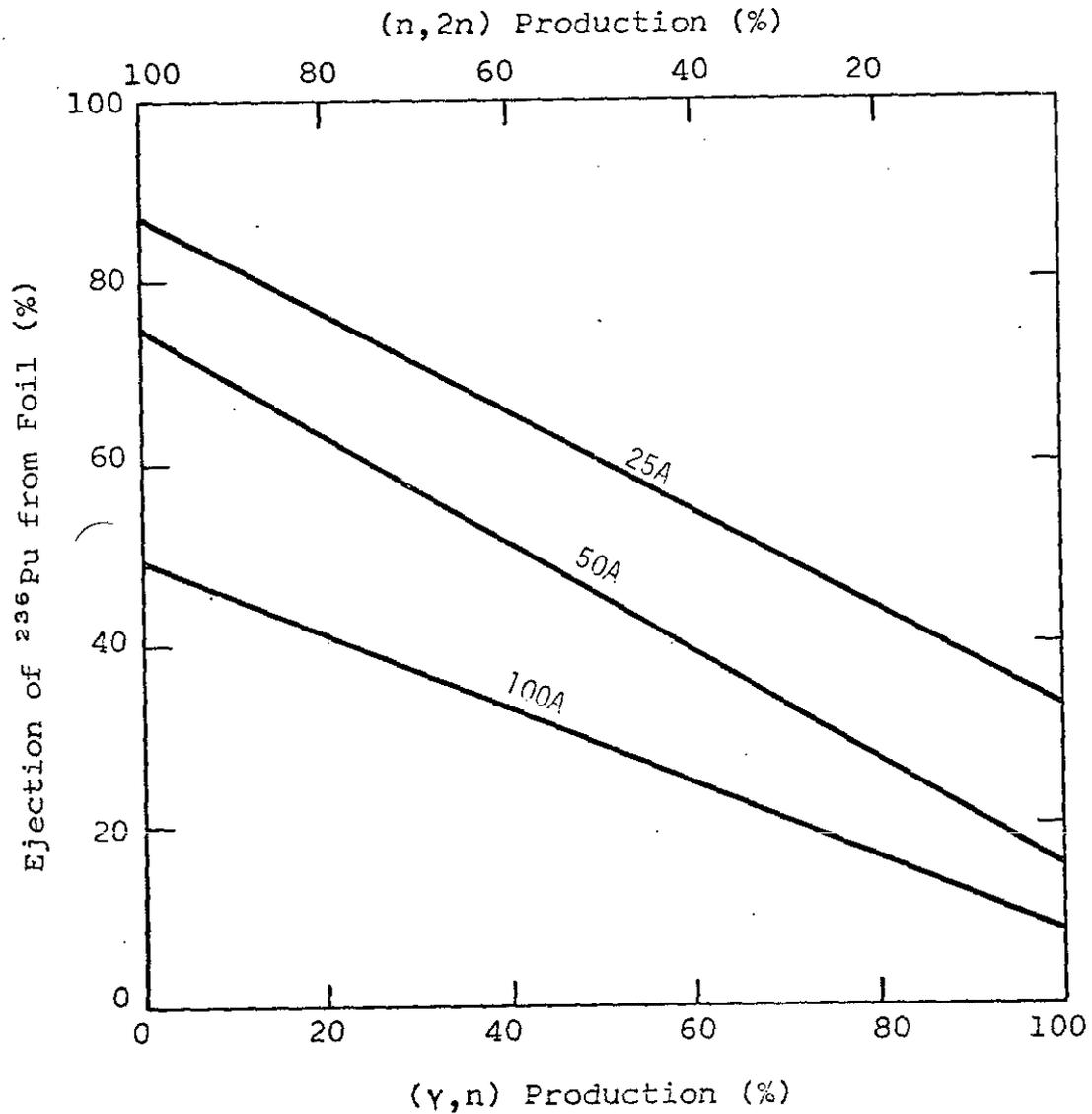


FIG. 9. EJECTION OF  $^{236}\text{Pu}$  BY RECOIL FROM Np METAL FOILS OF INDICATED THICKNESS AS A FUNCTION OF RELATIVE AMOUNTS OF PRODUCTION BY  $(\gamma,n)$  AND  $(n,2n)$  REACTIONS