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Po²¹⁰ PRODUCTION CAPABILITIES IN N REACTOR

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PHYSICS STUDY OF
PO²¹⁰ PRODUCTION CAPABILITIES IN N REACTOR

By

R. A. Dieterich

Reactor Physics
Research and Engineering
N REACTOR DEPARTMENT

November 19, 1963

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PHYSICS STUDY OF
Po²¹⁰ PRODUCTION CAPABILITIES IN N REACTOR

I. INTRODUCTION

The production of Po²¹⁰ by irradiating Bi²⁰⁹ has been proposed as a possible use of N Reactor. The purpose of this study was to make an estimate of the production rates for various exposure times and flux levels. The production rate was calculated for the three flux levels of:

- a) 5.0×10^{13} neutrons/cm²/second, which is about the average flux level expected in the N Reactor graphite moderator;
- b) 6.74×10^{13} neutrons/cm²/second, which is the estimated flux level in the 68 empty process tubes on the top and bottom of the N Reactor core;
- c) 7.5×10^{13} neutrons/cm²/second, which is the estimated flux level present in a flux trap made by removing a column of fuel.

In addition to the above calculations, an estimate was made of the fuel enrichment needed to support a column of bismuth in the core of the reactor; and the percentage loss in the Pu production was calculated for the reactor.

II. CONCLUSIONS

Figure 1 shows the buildup of Po²¹⁰ for the three flux levels discussed in the introduction. The production is expressed as the ratio of Po²¹⁰ atoms present to the number of Bi²⁰⁹ atoms originally present at the beginning of the irradiation, $N(\text{Po}^{210})/N(\text{Bi}^{209})$. One assumption made in the calculations for these curves was that the thermal absorption cross section for bismuth (34 millibarns) is small enough so that no self shielding is present.

If the bismuth were placed in the core of N Reactor so that it would be surrounded by eight fuel elements (i.e., the bismuth would be a central target cell in a supercell of nine lattices), it would be exposed to a flux of about 7.5×10^{13} neutrons/cm²/second. As can be seen in Figure 1, 1.1×10^{-5} of the original Bi²⁰⁹ atoms present would be converted to Po²¹⁰ at the end of four month's irradiation. If the entire reactor were composed of the supercells described above, there would be 111 bismuth targets, and four month's irradiation would yield 370 grams of Po²¹⁰. No allowance is included here for decay of Po²¹⁰ after the irradiation ceases or for losses which will occur during chemical separations. Also, for this

case, uranium fuel enriched to ~ 1.02 w/o U^{235} would be required in the eight process tubes surrounding the target in each supercell, and there would be a 13.8 percent loss in the Pu production if the fuel tubes were run at the same power level as they would be normally. However, if the tube power is raised by a factor of 9/8, thus maintaining the normal reactor power level, the loss in Pu production would be only 2.9 percent.

On the other hand, if the bismuth were placed in the 68 empty process tubes at the top and bottom of the core, it would be exposed to a flux of 6.74×10^{13} neutrons/cm²/second. For this case, as can be seen from Figure 1, 1.0×10^{-5} of the original Bi^{209} atoms present would be converted to Po^{210} at the end of four month's irradiation. This would amount to 206 grams of Po^{210} for all 68 empty tubes, again assuming no decay or chemical separations loss following the four month's irradiation period. In this case, there would be no additional fuel enrichment needed and no loss in Pu production.

III. DISCUSSION

A. Detailed Nuclear Processes

Irradiation of stable Bi^{209} leads to production of the α emitter, Po^{210} .

The physics processes involved are the following:

Here the neutron bombardment of Bi^{209} leads to two isomeric states of Bi^{210} . The half life of the upper state (2.6×10^6 years) is extremely long in comparison to that (5.0 days) of the lower; and therefore its effect on the buildup of Po^{210} was neglected. Also, the absorption cross section for Bi^{209} is so small (34 millibarns) that its concentration was assumed to be constant throughout the exposure. In addition, neutron capture in Po^{210} was neglected because it represents a much smaller loss of Po^{210} than the α decay

of Po^{210} to Pb^{206} . One further assumption was that the Bi^{209} proceeds directly to Po^{210} without the intermediate formation of Bi^{210} . This was justified on the basis of the relatively short half life (5.0 days) of the Bi^{210} . These assumptions lead to the following simplified scheme.

The various parameters involved are:

Bi^{209} : $\sigma_{\text{abs}} = 0.019 \text{ b}$ (for formation of the lower Bi^{210} isomer which proceeds to Po^{210}).

Po^{210} : $t_{1/2} = 138.4 \text{ days}$ or $\lambda = 5.795 \times 10^{-8} \text{ sec}^{-1}$

B. Production Rate of Po^{210}

The following equation was used to calculate the Po^{210} production rate.

$$\frac{dN(\text{Po}^{210})}{dt} = \sigma_{\text{abs}} \phi N_{209} - \lambda_{\alpha 210} N_{210},$$

where N_{210} equals the Po^{210} concentration, and N_{209} is the Bi^{209} concentration which is assumed to be a constant.

The solution of this equation is:

$$\frac{N(\text{Po}^{210})}{N(\text{Bi}^{209})} = \frac{\sigma_{\text{abs}} \phi}{\lambda_{\alpha 210}} (1 - e^{-\lambda_{\alpha 210} t})$$

Figure 1 shows this solution as a function of time for three different flux levels. The lower curve ($\phi = 5.0 \times 10^{13}$) represents about the average flux expected in the N Reactor graphite moderator. The middle curve ($\phi = 6.74 \times 10^{13}$) represents the flux assumed to be present in the 68 empty process tubes on the top and bottom of the reactor core. The flux is rapidly varying at this location and there is considerable uncertainty in this assignment of a flux level. This assumption was made with the aid of Figure 44 of the 100-N Technical Manual (HW-69000, Vol I), which shows the relative flux in N Reactor as a function of the distance from the pile center. The upper curve ($\phi = 7.5 \times 10^{13}$) represents the average flux assumed in a bismuth target tube in the

core of the reactor. This is a factor of three increase over the average flux in the fuel and results from a flux trap effect. There is also considerable uncertainty in the factor of three in flux enhancement, but it is perhaps a reasonable estimate. The macroscopic absorption cross section for a bismuth element clad with zirconium-2 and cooled with a 1/8" thick water annulus in a process tube was approximately 0.001 cm^{-1} while that for a standard fuel element was approximately 0.125 cm^{-1} .

Since the half life of Po^{210} is 138.4 days, it appears that an irradiation of approximately four months would be a reasonable selection for purposes of illustration. As can be seen from Figure 1, $N(\text{Po}^{210})/N(\text{Bi}^{209})$ for four months (1.03×10^7 seconds) is 1.1×10^{-5} for the target in a supercell, and 1.0×10^{-5} for a target in an empty process tube on the fringe. Assuming the bismuth target to be cylindrical with a radius of 3.05 cm (this leaves room for a 50 mil Zircaloy-2 cladding and a 1/8" thick water annulus within the process tube), the weight of bismuth per tube is 303 kg. Therefore, four month's irradiation would produce a total of 370 grams of Po^{210} in the 111 supercell target tubes, or a total of 206 grams in the 68 empty process tubes.

C. Estimated Enrichment Required to Support a Bi^{209} Column

To make rough estimates of the parameters associated with a Bi^{209} column within the core of N Reactor, a supercell consisting of a 3×3 lattice structure was used. In this supercell, the center cell was filled with a Bi^{209} element, while the eight surrounding cells were filled with standard uranium fuel elements.

1. Estimate of f' for the Supercell

The thermal utilization is given by:

$$f = \frac{1/v \text{ absorption rate in the fuel}}{1/v \text{ absorption rate in the entire cell}}$$

or

$$f = \frac{A_F}{A_F + A_M} = \frac{1}{1 + \frac{A_M}{A_F}}$$

where

$A_M = 1/v$ absorption rate in the other cell components

and

$A_F = 1/v$ absorption rate in the fuel.

Thus for the standard cell we have:

$$f_o = \frac{1}{1 + \frac{A_{M_o}}{A_{F_o}}}$$

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The 100-N Technical Manual (HW-69000, Vol I, p. 3.2-23) gives a value for

$$f_o = 0.8803.$$

Therefore

$$\frac{A_{M_o}}{A_{F_o}} = 0.1360$$

For the supercell containing the Bi^{209} target cell, we have:

$$f' = \frac{1}{1 + \frac{A'_M + A'_T}{A'_F}}$$

where

$A_T = 1/v$ absorption rate in the target material,

and

A'_F and $A'_M = 1/v$ absorption rates in the fuel and other cell components in the supercell.

The Bi^{209} absorption cross section is so small that it was assumed that A_T was negligible (i.e., $A_T = 0$) and it was also assumed that the average flux in the target cell would be higher by a factor of approximately three than the flux in the other components of a regular cell.

Therefore,

$$A'_M = [(8 \times 1) + (1 \times 3)] A_{M_o} = 11 A_{M_o}$$

and

$$A'_F = [(8 \times 1) + (1 \times 0)] A_{F_0} = 8 A_{F_0}.$$

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Therefore,

$$\frac{A'_M}{A'_F} = \frac{11}{8} \frac{A_{M_0}}{A_{F_0}} = 0.1870$$

Thus the new value of f is:

$$f' = \frac{1}{1 + .1870 + 0} = 0.8425 = 0.9571 f_0.$$

2. Calculation of p' for the Supercell

The resonance escape probability is given by:

$$p = \exp \left[- \frac{N_F V_F \bar{\phi}_F}{N_M V_M \bar{\phi}_M} \times \frac{R.I.}{\xi \Sigma_{S_M}} \right],$$

where

N = number of atoms per cc of fuel or moderator,

V = volume of fuel or moderator,

$R.I.$ = resonance integral of U^{238} ,

ξ = average logarithmic energy decrement,

Σ_{S_M} = macroscopic scattering cross section of the moderator,

and

$\bar{\phi}$ = epithermal flux in fuel or moderator

Due to the uncertainty in the change in the epithermal flux, two values were assumed for the moderator flux in the central target cell:

$$\text{Case 1) } \bar{\phi}'_M = \bar{\phi}_{M_0},$$

and

$$\text{Case 2) } \bar{\phi}'_M = 1/2 \bar{\phi}_{M_0}.$$

In both cases, it was assumed that no changes occur in the epithermal flux of the moderator or fuel of a regular fuel cell, and again, $V'_F = \frac{8}{9} V_{F_0}$.

Using a value for $p_o = 0.8291$ from the 100-N Technical Manual (HW-69000, Vol I, p. 3.2-23), values for p' are given as follows:

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Case 1) $(V_F' = \frac{8}{9} V_{F_o})$:

$$p_1' = (0.8291) \frac{8}{9} = 0.8465 = 1.0210 p_o$$

Case 2) $(V_F' = \frac{8}{9} V_{F_o} \text{ and } \phi_M' = 1/2 \phi_{M_o} \text{ for the target cell})$

Here $\phi_M' = \frac{(8 \times 1 + 1 \times 1/2)}{9} \phi_{M_o} = \frac{8.5}{9} \phi_{M_o}$

Therefore,

$$p_2' = (0.8291) \frac{8}{9} \times \frac{9}{8.5} = (0.8291) \frac{8}{8.5} = 0.8383 = 1.0111 p_o$$

These two values of p' will probably bracket the actual value of p' .

3. Resultant value of k_∞ for the Supercell

No changes were assumed to occur in η and ϵ in the four factor formula. On this basis, new values for k_∞ for both cases 1 and 2 were calculated.

Case 1) $(k_\infty)_1 = \frac{f' p_1' \eta_o \epsilon_o}{f_o p_o \eta_o \epsilon_o} k_{\infty_o} = 0.9571 \times 1.0210 k_{\infty_o} = 0.9772 k_{\infty_o}$

Case 2) $(k_\infty)_2 = \frac{f' p_2' \eta_o \epsilon_o}{f_o p_o \eta_o \epsilon_o} k_{\infty_o} = 0.9571 \times 1.0111 k_{\infty_o} = 0.9677 k_{\infty_o}$

4. Required Fuel Enrichment

The values calculated for k_∞' above assume no change in the fuel enrichment. It is now desired to find that enrichment which will bring the value of k_∞ for the supercell up to k_{∞_o} . Since p and ϵ are relatively insensitive to changes in the enrichment, the following formulation was used:

$$\frac{k_\infty''}{k_\infty'} = \frac{\eta'' f''}{\eta' f'} = \frac{k_\infty}{k_\infty'}$$

Here,

$$\eta f = \frac{\sum a_F}{\sum f_F} \frac{1}{1 + \frac{A_M}{A_F}}$$

Since

$$A_T \approx 0$$

and

ν = average number of neutrons released per thermal fission,

Σ_{f_F} = macroscopic fission cross section of the fuel,

Σ_{a_F} = macroscopic absorption cross section of the fuel.

In a new enrichment, E'' , the ratio of the new value $\eta'' f''$ to the old, $\eta' f'$, at enrichment E' is given by:

$$\frac{k_{\infty}''}{k_{\infty}'} = \frac{\eta'' f''}{\eta' f'} = \frac{\Sigma_{f_F}'' \Sigma_{a_F}' (1 + \frac{A_M'}{A_F'})}{\Sigma_{f_F}' \Sigma_{a_F}'' (1 + \frac{A_M''}{A_F''})}$$

Here,

$$\frac{\frac{A_M''}{A_F''}}{\frac{A_M'}{A_F'}} \approx \frac{\Sigma_{a_F}'}{\Sigma_{a_F}''}, \text{ where } \frac{A_M'}{A_F'} = 0.1870$$

(see Section III, C.1.)

Substitution of these quantities into the equation for k_{∞}''/k_{∞}' and algebraic manipulation yields:

$$\frac{k_{\infty}'}{k_{\infty}''} = \frac{\Sigma_{f_F}' \Sigma_{a_F}''}{1.1870 \Sigma_{f_F}'' \Sigma_{a_F}'} + \frac{0.1870 \Sigma_{f_F}'}{1.1870 \Sigma_{f_F}''}$$

where

$$k_{\infty}' = 0.9772 k_{\infty}'' \text{ (Case 1)}$$

$$= 0.9677 k_{\infty}'' \text{ (Case 2),}$$

$$\Sigma_{f_F} \propto E \sigma_f (235),$$

$$\Sigma_{a_F} \propto E \sigma_a (235) + (1 - E) \sigma_a (238),$$

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and

$$E' = 0.00947.$$

The solution of the above equation indicates that the enrichment needed in the supercell is 1.003 w/o U^{235} for the case where there is no change in the epithermal flux in the moderator of the target cell, and 1.029 w/o U^{235} for the case where the epithermal flux in the target cell is equal to one half that in the other cells. A reasonable estimate of that enrichment actually required is ~ 1.02 w/o U^{235} .

D. Estimation of Pu Production Losses

One further factor of importance when considering the irradiation of bismuth is the percent loss of plutonium production. Assuming N Reactor to be completely loaded with the supercells described above, i.e., containing eight regular fuel cells and a central target cell, calculations were made to give an estimate of the percent loss in the conversion ratio. If the tube powers are kept at the same level as they are in the case of a full reactor loading of 0.947 w/o U^{235} , and if the uranium enrichment is changed to 1.02 w/o U^{235} as was calculated above, the conversion ratio (expressed in grams Pu/Mwd) will suffer a 13.8 percent loss. On the other hand, if the tube power is raised from 4000 kw to 4500 kw, i.e., keeping the reactor power at the same level, and again raising the enrichment to 1.02 w/o U^{235} , the conversion ratio will suffer a loss of only 2.9 percent.

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