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THE IRRADIATION OF A NOVEL FUEL ELEMENT USED FOR
U-233 PRODUCTION

R. W. WOODRUFF AND G. W. STUART

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THE IRRADIATION OF A NOVEL FUEL ELEMENT USED FOR
U-233 PRODUCTION

By

R. W. Woodruff and G. W. Stuart

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Physics Unit
Applied Research Sub-Section

September 10, 1954

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ABSTRACT

A new slug for U-233 production consisting of a solid thorium core externally jacketed by a thin Oralloid foil is described. Advantages of this system are pointed out. The bulk of this report concerns itself with the long-term changes of reactor parameters as the foil slug is irradiated at constant specific power (kilowatts per foot). Isotope yield curves are also presented.

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THE IRRADIATION OF A NOVEL FUEL ELEMENT USED FOR
U-233 PRODUCTION

Nomenclature

- ϕ - average flux
- W - specific power
- K - kw-sec/fission
- V - volume
- T - see equation (2)
- F - disadvantage factor
- R - radius of core
- d - thickness of film
- β - blackness to thermal neutrons
- λ - decay constant
- Σ - macroscopic cross section
- N - number of nuclei/cc
- k - reactivity
- f - thermal utilization
- P - resonance escape probability
- ν - number of fast neutrons per fission
- α - ratio of activation to fission cross section
- 02 - Th-232
- 03 - Th-233
- 04 - Th-234
- 13 - Pr-233
- 14 - Pr-234
- 23 - U-233
- 24 - U-234
- 25 - U-235
- 26 - U-236

Subscripts

- p. t. - process tube, dry
- p - poison
- c - core
- a - absorption
- f - fission
- s - scattering

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INTRODUCTION

A current Hanford method of producing uranium-233 employs separate fuel and target slugs. A process tube is loaded with six-inch thorium target slugs (Q slugs) alternating with eight-inch uranium-235-aluminum fuel slugs (J slugs, 7.8% uranium-235 by weight). The conversion ratio, i. e., nuclei of uranium-233 produced per nucleus of uranium-235 destroyed, for the J-Q loading is approximately 0.80. The small magnitude of the quantity is a reflection on the inefficiency of producing uranium-233 with a J-Q loading.

It is not difficult to isolate the undesirable characteristics of the J-Q scheme. Primarily there is just not enough thorium metal packed into the reactor. Only six-fourteenths of the process tube length is occupied by thorium. The situation is further aggravated by the smaller neutron absorbing properties of thorium compared to uranium in both the thermal and resonance energy ranges. This last statement must be correlated with the knowledge that Hanford lattices were designed to operate on uranium. In addition, parasitic neutron capture in the J-slug aluminum is appreciable since such material occupies eight-fourteenths of the process tube length.

We now seek to alleviate the insufficiencies of the J-Q loading by designing a new slug for uranium-233 production. In essence, we now attempt to crowd as much thorium as possible into the existing Hanford lattice. This can best be done by employing the most concentrated fissionable isotope available to satisfy nuclear fuel requirements. Such utilization of concentrated fissionable fuel will not only increase the volume of thorium in the pile, but will also mitigate parasitic capture in the diluent.

The most concentrated fissionable isotope which would seem to be available is Orallo, an enriched uranium having an isotopic composition of ca. 94% uranium-235. Accordingly, a composite fuel-target slug has

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been designed which would consist of a solid thorium core of standard Hanford radius (1.69 cm.) externally jacketed with an Oralloy foil. This slug would be used in a homogeneous process tube loading; i.e., the entire length of process tube would be loaded with one type slug in counter-distinction to the J-Q loading. Attendant upon the remark, one notes that the Oralloy film loading will generate heat along the entire length of process tube. Such is a marked advantage over the J-Q loading which generates heat in only eight-fourteenths of the process tube length and thus, for a given pile power, would generate a specific power per unit length of fuel fourteen-eighths greater than the Oralloy foil slug.

From the viewpoint of reactivity, the Oralloy foil slug has been designed to have $k_{\infty} = 1.045$. This is achieved with a foil thickness of 2.8 mils. The conversion ratio is 0.80.

There are several factors contributing to the selection of the particular geometrical disposition of Oralloy and thorium considered. Of prime importance are the thermal characteristics of the fuel element. By adopting an external film slug, one generates the great majority of power (all in the case of green metal) directly adjacent to the cooling water annulus. Obviously such an assembly will have superior heat transfer behavior.

Disposing the fuel material externally to the target material eliminates shielding of the former by the latter, reducing the required uranium-235 inventory. On the other hand this same effect reduces the conversion ratio somewhat. (A homogeneous blend of thorium and U-235 would have a higher conversion ratio than the foil type slug. Unfortunately, such an element would not be satisfactory for U-233 production due to the mixing of uranium-233 and uranium-235 in the product. In the event that such a mixture of isotopes is acceptable for weapons purposes then the homogeneous slug appears desirable.)

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There are two additional benefits inherent in the film slug. Gamma ray heating of graphite will be reduced below the level generated by J slugs due to Compton scattering in the thorium core. Criticality hazards in a separations facility will be mild, due to the comparatively small mass of enriched material (Oralloy) which must be handled.

The Oralloy foil would be removed from the thorium core either mechanically or chemically as the first step in a separations process.

CALCULATION

A most attractive feature of the thorium-uranium-233 manufacturing cycle is the possibility of attaining large exposures of process metal. This statement is predicated on the absence of appreciable spontaneous fission in the uranium-234 isotope along with the comparatively small value of α for uranium-233. We have, therefore, formulated the problem of isotope buildup for the Oralloy foil element described above.

Instead of the usual unrealistic condition of isotope buildup in constant flux, we have instead accepted the model of constant specific power per unit length of process tube (kilowatts per ft.) since a production reactor attempts to maintain a flat power distribution during operation. This statement requires flux to vary with irradiation according to

$$(1) \phi_{25} = \frac{W}{K(V_{25}\Sigma_{f25} + V_c\Sigma_{f23}T)}$$

where

$$(2) \phi_c = T \phi_{25}$$

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Adopting slab geometry for the Oralloy film and using⁽¹⁾

(1) H. Neumann, HW-32349, p. 34 et seq.

$$(3) F = \frac{\Sigma_{ac} R(2-\beta_c)}{\mu_c}$$

we may express

$$(4) T = \frac{\frac{\sqrt{\pi}}{2} (\Sigma_p + \Sigma_{a25}) + \Sigma_{s25}}{\frac{\sqrt{\pi}}{2} \Sigma_{ac}} \left(\frac{d}{R} \right) \left(\frac{\beta_c}{2-\beta_c} \right) \frac{1}{\exp d \frac{\sqrt{\pi}}{2} (\Sigma_p + \Sigma_{a25}) - 1}$$

where the factor $\frac{\sqrt{\pi}}{2}$ averages $1/v$ cross sections over a Maxwellian neutron distribution and

$$(5) \Sigma_p = 50 \text{ barns} \left(\frac{\sigma_{f25}}{\sigma_{a25}} \right) [N_{25}(0) - N_{25}(t)]$$

accounts for fission product buildup in the film. Fission product poisoning due to uranium-233 fissions in the core is neglected throughout as a higher order effect. (2) Core blackness is calculated by the techniques developed in HW-29992 (3) Thermal instead of effective cross sections are used in this direction. The primary result of such an exchange substitutes $\sigma_{a02} = 7^b$ for the $\sigma_{a02} = 9^b$ used below.)

The fluxes yielded by equations (1) and (2) are then substituted into the isotope buildup relations for the chain.

- (2) The errors introduced by this omission will be discussed below.
- (3) R. W. Woodruff and G. W. Stuart, HW-29992. Scattering cross sections used to calculate β_c are taken from BNL-170.

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which are

$$\begin{aligned}
 \dot{N}_{03} &= N_{02} \sigma_{02} \phi_c - N_{03} \lambda_{03} - N_{03} \sigma_{03} \phi_c \\
 \dot{N}_{13} &= \lambda_{03} N_{03} - \lambda_{13} N_{13} - N_{13} \sigma_{13} \phi_c \\
 \dot{N}_{23} &= \lambda_{13} N_{13} - N_{23} \sigma_{23} \phi_c \\
 (6) \quad \dot{N}_{04} &= N_{03} \sigma_{03} \phi_c - \lambda_{04} N_{04} \\
 \dot{N}_{14} &= N_{04} \lambda_{04} + N_{13} \sigma_{13} \phi_c - N_{14} \lambda_{14} \\
 \dot{N}_{24} &= N_{23} \sigma_{23} \phi_c + N_{14} \lambda_{14} \\
 \dot{N}_{25} &= -N_{25} \sigma_{25} \phi_c
 \end{aligned}$$

Equations (6) are incorrect by the use of average fluxes in core and film instead of the actual fluxes at each point. We have not estimated the magnitude of this error.

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RESULTS

The above formalism has been applied to constant uninterrupted irradiation under specific powers of 20, 40, 60, and 80 kilowatts per foot. In actual practice, reactor down time would allow Pa-233 and other isotopes to decay, introducing deviations from our calculation.

The results of this calculation may be succinctly revealed through a review of the attached graphs. All plots have exposure in days as abscissa.

Figures 1 through 6 are the usual type of isotope concentration graph.

Figure 7 has as ordinate the quantity, $\Sigma_{03} + \Sigma_{13} + \Sigma_{23}$.

Figure 8 has as ordinate the quantity, $\frac{\Sigma_{03} + \Sigma_{13} + \Sigma_{23}}{\Sigma_{04} + \Sigma_{14} + \Sigma_{24}}$.

Figure 9 exhibits U-235 burnout as a function of exposure.

Figure 10 has as ordinate, $(\Sigma_{02} - \Sigma_{03} - \Sigma_{13} - \Sigma_{23}) \phi_c V_c$, i.e., the net number of nuclei of product per foot of slug (upon infinite decay) formed in the final second of irradiation. This will portray the economical point to terminate irradiation due to increasing burnout of final product.

Figure 11 portrays the variations of reactivity

$$(7) k_{\infty} \approx f p \frac{\nu_{25} \Sigma_{f25} V_{25} + \nu_{23} \Sigma_{f23} V_c T}{(\Sigma_p + \Sigma_{a25}) V_{25} + \Sigma_{ac} V_c T}$$

with exposure. The resonance escape probability is kept constant at 0.91 throughout exposure. The method of calculating f is discussed in Appendix

- A. Neglect of fission product buildup in the core, mentioned above, lowers k_{∞} in the extreme case of 300 days irradiation under 80 kw/ft by approximately one per cent. For exposures under the lower specific powers

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considered this effect is negligible. Any future calculation should include this term for completeness.

Figure 12 plots the ratio of power generated in uranium-233 to that generated in uranium-235.

Figure 13 plots the percentage blacknesses of core and slug where slug blackness is yielded by

$$(8) \beta = (1 - e^{-\sum_{25d}}) + e^{-\sum_{25d}} \beta_c + e^{-\sum_{25d}} (1 - \beta_c) (1 - e^{-\sum_{25d}})$$

and β_c is calculated as before (see footnote 3).

Figure 14 and 15 plot average foil and core flux, respectively. Perusal of these curves reveals the divergence between constant flux and constant power operation.

Figure 16 and 17 examine more deeply the differences between constant flux and constant power conditions. Figure 16 reveals the difference in k_{∞} calculated both ways for a low and a high specific power irradiation while Figure 17 discharges the same function for product formation.

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G. W. Stuart

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APPENDIX A - Calculation of f

Mr. David Lanning has performed a small source type analysis⁽⁴⁾ which relates process tube blackness to dry thermal utilization, i.e., f without water in the cooling annulus. Subtracting 0.02 for the effect of water we modify Lanning's results to read

$$(A.1) f = \left(\frac{0.0423}{P_{p.t.}} + 0.0095 \right)^{-1} - 0.0200$$

for the Hanford dimensioned lattice. Process tube blackness is calculated from equation (8) and the recipe given in HW-29992 (see footnote 3). As an example of this formalism the following chart plots f as a function of exposure for an exposure rate of 60 kw/ft.

• Days	f
0	0.8917
50	0.8907
100	0.8904
150	0.8904
200	0.8903
250	0.8903
300	0.8904

(4) D. Lanning, HW-30508, p. 21.

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Fig. 1 Thorium-233 Count
Versus Exposure Time For
Constant Specific Power

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FIG 5. PROTACTINIUM-234 CONTENT
VERSUS EXPOSURE TIME FOR
CONSTANT SPECIFIC POWER

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FIG. 7 URANIUM-233 CONTENT
AFTER INFINITE DECAY VERSUS EXPOSURE TIME
FOR CONSTANT POWER

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SPECIFIC POWER (KW/FT)

FIG. 10, COMBINED PRODUCTION RATES OF THORIUM 233, PROTACTIONIUM 233, & URANIUM 233 VERSUS EXPOSURE TIME FOR CONSTANT SPECIFIC POWER

Fig. 11, Iunura Multiplication Factor
Versus Exposure Time for Constant
Specific Power

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FIG. 13 Percent Blackness vs. Core
And Sun Versus Exposure Time
For Constant Specific Power

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FIG. 16, PERCENT DIFFERENCE
BETWEEN INFINITE MUL-
TIPLICATION FACTOR, K, FOUND
FROM CONSTANT POWER, C.P.,
& CONST. FLUX, CF, CALCULATION
VERSUS EXPOSURE TIME FOR
CONSTANT SPECIFIC POWER

SPECIFIC POWER (KW/FT)

20

300

250

200

150

100

50

0

EXPOSURE TIME (DAYS)

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SPECIFIC POWER (KW/FT)

20

80

300

250

200

150

100

50

0

EXPOSURE TIME (Days)

$N_{cp} - N_{inf} (\%)$

20

10

0

0

0

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