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TITLE

FRINGE ISOTOPE PRODUCTION

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FRINGE ISOTOPE PRODUCTIONINTRODUCTION

The purpose of this work has been to determine experimentally the rate of production of tritium in fringe lithium-aluminum alloy loadings with the degree of precision necessary for economic analyses of such a method of isotope production (1,2). These results are provided for use in such an analysis.

SUMMARY

The production rate of tritium in the outermost fringe tubes is related to the heat generation in the enriched material in the adjacent lattice by the equation:

$$T = 0.0021 M_{adj}$$

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SUMMARY (Continued)

where T = grams of tritium per full length charge (64 four-inch pieces)
of lithium-aluminum alloy material (concentration 3.5 per cent
natural lithium)

M_{adj} = MWD of heat generated in the adjacent enriched column
normalized on a per ton basis

The experimental constant (0.0021) is estimated to be accurate to plus or minus ten per cent. The constant should apply also to fringe loads utilizing somewhat greater or smaller quantities of enrichment than that employed in the test, that is for isotope production loadings which are slightly over or under-compensated from a reactivity standpoint.

A numerical evaluation of the production in the test loading plus the central natural uranium columns indicates that 80 atoms of T and 2300 atoms of Pu would be produced while 2826 atoms of U-235 would be consumed. For a natural uranium loading in the test zone it is calculated that 2410 atoms of Pu would have been produced and 2909 atoms of U-235 consumed in the same columns during a similar operating interval. It appears that the atom conversion ratio of the reactor could be increased by about one per cent by the use of complete fringe poison loadings.

The accuracy of this experimental evaluation is limited by the large variation in the radioactivity of the cobalt pieces used to measure the neutron flux in various parts of the loading. In the present analysis it was assumed that the average found in the test load would be representative of any large scale reactor loading. It is felt that actual processing of significant quantities of material is necessary to obtain a really good evaluation of conversion efficiency.

DESCRIPTION OF TEST

Isotope production rates in materials placed within a reactor are governed by the neutron flux in which the material resides. In the case of fissionable materials the neutron flux can be evaluated from the heat generation rate in the column of metal. In full length isotope production columns the heat generation is small and some other method must be used to determine the flux. The radioactivity of cobalt samples irradiated in experimental metal charges has been used to relate the flux level in the fringe mint columns to the flux level (as indicated by heat generation rate) in the adjacent E metal columns.

Cobalt has a long half-life (5.2 years) compared to the length of the irradiation used in this test (approximately six months); hence, the radioactivity of the cobalt pieces is almost directly proportional to the integrated exposure (ϕt). The cobalt-aluminum mixture which was used in the test was designed to absorb neutrons at the same rate as either E-metal or mint material. Therefore, the volume average flux in an isolated cobalt-aluminum piece should be representative of the volume average flux in the E-metal or mint piece which it displaced.

Four pairs of mint and E-metal columns were used in the fringe poison load at the DR Reactor. Cobalt pieces were centered with mint in tubes 1896-DR and 2296-DR; matched cobalt pieces were centered with E-metal in tubes 1894-DR and 2294-DR. Likewise, cobalt pieces were placed one-quarter of the way from the downstream end of the active charge in tubes 2096-DR and 2596-DR (with mint) and in tubes 2094-DR and 2594-DR (with E-metal). Daily readings of the flow and temperature rise on these E-metal charges

DESCRIPTION OF TEST (Continued)

and also on the E-metal charges on the corresponding rows in column 95 were taken. After discharge the numbered E metal pieces were weaseled to obtain the front-to-rear flux distribution, and the cobalt pieces were weaseled to obtain the flux ratio between the E-metal charges and the mint charges.

A. Derivation of Production Relations

It is assumed that the amount of tritium produced in a full length column of mint slugs is given by the equation:

$$(1) \quad T = \bar{\phi}_p t N_6 \sigma_a w$$

where T = grams of tritium per tube of mint

$\bar{\phi}_p$ = volume average flux in the mint, n/cm^2 sec

t = exposure time, seconds

N_6 = number of atoms of Li-6 in a tube of mint

σ_a = absorption cross section for n, α process, cm^2

w = grams of tritium per atom of tritium

Likewise the integrated exposure of the charge of E-metal in the tube immediately adjacent to the mint column is:

$$(2) \quad M_{adj} = \bar{\phi}_{adj} t H_f N_{25} \sigma_f c (1 + \delta)$$

where M_{adj} = MWD per adjacent ton of E-metal (MWD/AT)

H_f = heat of fission, assumed to be 200 Mev

$(1 + \delta)$ = fast effect, taken as 1.057

N_{25} = atoms of U-235 per ton of E-metal

σ_f = fission cross section of U-235, cm^2

c = energy conversion constant, 1.854×10^{-24} MWD/Mev

Because the cobalt-aluminum pieces were not irradiated in the adjacent columns it is necessary to include also the experimental columns:

$$(3) \quad M_{exp} = \bar{\phi}_{exp} t H_f (1 + \delta) N_{25} \sigma_f c$$

where the meanings are as before and the subscript "exp" stands for experimental.

As stated before, the radioactivity of the cobalt pieces is a measure of the integrated flux at that point. Assuming the same front-to-rear flux to exist in both the E-column and mint column the following relation should hold for pieces irradiated at the same relative location in the charge:

DESCRIPTION OF TEST (Continued)

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$$\frac{\bar{\phi}_p^t}{A_p} = \frac{\bar{\phi}_{exp}^t}{A_{exp}}$$

where A represents the cobalt-aluminum activity in any arbitrary unit. Substituting this value of $\bar{\phi}_p^t$ in equation (1) yields:

$$T = \frac{\bar{\phi}_{exp}^t}{A_{exp}} A_p N_6 \sigma_a w.$$

Dividing by the exposure of the adjacent metal:

$$\frac{T}{M_{adj}} = \frac{A_p}{A_{exp}} \frac{\bar{\phi}_{exp}^t}{M_{adj}} N_6 \sigma_a w.$$

Using equation (3) to evaluate $\bar{\phi}_{exp}^t$:

$$(4) \quad \frac{T}{M_{adj}} = \left(\frac{A_p}{A_{exp}} \right) \left(\frac{M_{exp}}{M_{adj}} \right) \left(\frac{N_6 \sigma_a}{N_{25} \sigma_c H_f (1 + \delta)_c} w \right)$$

B. Evaluation of Production Relation

The production relation (equation 4) is the product of three ratios. The first two of these were determined experimentally in this production test; the other can be calculated using accepted values of material composition, cross sections, etc. The evaluation of these ratios will be discussed briefly.

1. A_p/A_{exp} - The ratio A_p/A_{exp} was obtained in four separate cases - two each for the central location of the charge and two each for the downstream location. Unfortunately, a large variation was found to exist in these ratios. The central values were found to be 0.550 (18 row) and 0.516 (22 row), whereas the downstream values were 0.481 (20 row) and 0.454 (25 row). A definite systematic variation can be seen in these ratios: the ratios increase from the top to the bottom of the load and increase from the downstream end to the center of the load. The reason for this variation is not apparent. The weasel data of the E-metal gave a reasonably smooth cosine distribution for all the charges; there is no reason to suspect that the distribution in the mint should be materially different. For lack of a better method, a simple average of the four experimental ratios was assumed to be realistic (0.50). An estimated accuracy of plus or minus 10 per cent is associated with this value.
2. M_{exp}/M_{adj} - Over four hundred determinations of the tube power of the experimental E-metal columns and the adjacent E-metal columns were obtained during the test period to evaluate the exposure ratio M_{exp}/M_{adj} . With an estimated accuracy of plus or minus four per cent on the individual determinations, the resulting ratio of 1.377 is calculated to be within plus or minus 0.2 per cent. No significant variation in this ratio was noted except for the

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DESCRIPTION OF TEST (Continued)

charges in the 18 row which gave a consistently lower ratio. This is attributed to the fact that all rows below 18 were filled with natural uranium (instead of the enrichment-poison loading); hence, the buckling was less. This "edge" effect would be associated with any small fringe poison load short of a complete blanket.

3. Ratio of Material Constants - The evaluation of the ratio of the macroscopic cross sections of the two materials requires simply a knowledge of material composition. It was assumed that mint was 3.5 per cent natural lithium; that is, 3.5 grams of lithium plus 96.5 grams of aluminum equal 100 grams of mint. On a volume basis this means there are 0.1549 cm^3 of lithium per cm^3 of mint. A full length charge of mint consists of 67 pieces with each piece having a core length of 3.80 inches. Thus,

$$N_6 = 67 \left[\pi (0.682)^2 (3.80)(2.54)^3 \right] (0.1549)(0.075)(0.0463 \cdot 10^{24})$$

$$= 3.278 \cdot 10^{24} \text{ atoms of Li-6 per tube of mint}$$

where natural lithium is assumed to be 7.5 per cent isotope 6 and there are $0.0463 \cdot 10^{24}$ atoms of lithium per cm^3 of lithium. The value of σ_a is 940 barns and the weight of tritium is $5.008 \cdot 10^{-24}$ grams per atom. Thus, the numerator becomes:

$$N_6 \sigma_a w = (3081)(5.008 \cdot 10^{-24}) = 15,430 \cdot 10^{-24}$$

The E-metal is assumed to be 0.95 per cent U-235; therefore,

$$N_{25} = 9.072 \cdot 10^5 \left(\frac{\text{gm of E}}{\text{ton of E}} \right) 6.023 \cdot 10^{23} \left(\frac{\text{atom of E}}{\text{gm at wt}} \right) \frac{1}{238} \left(\frac{\text{gm at wt}}{\text{gm of E}} \right) 0.0095$$

$$\left(\frac{\text{atom of 25}}{\text{atom of E}} \right)$$

$$= 22.08 \cdot 10^{24} \text{ atoms 25 per ton of E.}$$

Also

$$\sigma_E = 584 \cdot 10^{-24} \text{ cm}^2$$

$$H_F = 200 \text{ Mev/fission}$$

$$(1 + \delta) = 1.057$$

$$c = 1.854 \cdot 10^{-24} \text{ MWD/Mev.}$$

The denominator of the ratio becomes:

$$N_{25} \sigma_F H_F (1 + \delta) c = (22.08 \cdot 10^{24})(584 \cdot 10^{-24})(200)(1.057)(1.854 \cdot 10^{-24})$$

$$= 5,053,900 \cdot 10^{-24}$$

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DESCRIPTION OF TEST (Continued)

The ratio

$$\frac{N_6 \sigma_a w}{N_{25} \sigma_f H_f (1 + \delta) c} = \frac{15,430 \cdot 10^{-24}}{5,053,900 \cdot 10^{-24}} = 0.00305$$

4. T/M_{adj} - The numerical evaluation of equation 4 is:

$$\frac{T}{M_{adj}} = (0.50)(1.377)(0.00305) = 0.0021 \frac{\text{gms of T per tube of mint}}{\text{MWD per adjacent ton of E}}$$

An evaluation of the uncertainties indicates the accuracy of this conversion constant to be about ten per cent. The primary source of the uncertainty is introduced by the wide spread in cobalt slug activities which was obtained.

C. Numerical Example

To calculate the relative production efficiency of the mint loading the flux distribution in the fringe region before and after placing the poison-enrichment is used (see reference 2). The production along one row of tubes from the center to the far side of the reactor is considered as the basis of comparison. This should be conservatively realistic of a complete blanket loading. Using an arbitrary central power of one MW per tube in the flattened zone the heat generation before the test load is 20.95 MW. After the poison-enrichment load the power is 20.30 MW where 18.84 MW is generated in natural uranium and 1.46 MW is generated in the E-metal. The E-metal adjacent to the mint is operating at 0.43 MW per tube or 3.5 MW per ton. In 136 full power operating days the mint column will produce one gram of T:

$$T = 136 \text{ days } 3.5 \frac{\text{MW}}{\text{Ton}} 0.0021 \frac{\text{gm of T/tube}}{\text{MWD/Ton}} = 1 \text{ gm of T}$$

During this same period of time the natural uranium generates $(136)(18.84) = 2562$ MWD and the E generates $(136)(1.46) = 199$ MWD. If the load had not been present the heat generation would have been $(136)(20.95) = 2849$ MWD.

To calculate plutonium production and U-235 burnout, average factors for I & E natural and for solid E irradiated to 800 MWD/T were used. These values are:

	Plutonium Made g/MWD	U-235 Consumed g/MWD	Reference
I and E Natural	0.846	1.021	(3)
Solid E	0.666	1.057	(4)

The production and burnout in grams becomes:

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DESCRIPTION OF TEST (Continued)

	Pu.	T		U-235	Atom Conversion Efficiency
		Actual	Pu-equivalent		
Before	2410	0	0	2909	0.828
After	2300	1	80	2826	0.842

where the conversion efficiency is on a per atom basis. It is apparent that the overall pile conversion efficiency would be increased by the irradiation of a fringe isotope production load. This remains true even if the tritium production is considerably less than that determined by the experiment. However, the advisability of utilizing fringe isotope production depends on the relative economic value of the products, on the operational handling and separations problems involved, and on other side effects such as shield protection, rather than simply on the conversion efficiency.

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Irradiation Testing Sub-Section
Research and Engineering Section
(Work performed in Operational
Physics Sub-Section)

WL Bunch:mwe

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