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^{236}Pu CONTAMINANT IN ^{238}Pu PRODUCED IN POWER REACTORS

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ABSTRACT

The utility of ^{238}Pu produced from ^{237}Np irradiation is dependent on the amount of ^{236}Pu present. Calculations of ^{236}Pu produced by (n,2n) reactions on ^{237}Np indicate that light water power reactors produce ^{238}Pu containing 20 to 40 times as much ^{236}Pu as that produced in the USAEC D_2O or graphite production reactors, and the liquid metal fast breeders produce 600 to 900 times as much ^{236}Pu as the production reactors.

SUMMARY

The projected availability of large amounts of ^{237}Np from power reactors by the mid-1970's indicates this material could be irradiated on a large scale to produce ^{238}Pu as an easily shielded α heat source for isotopic power sources, heart pumps, and heart pacers. The required irradiation capability exists in the USAEC production reactors and has been proposed for the power reactors.

In most applications the usefulness of the ^{238}Pu is dependent on the amount of ^{236}Pu present, since the ^{236}Pu decay daughters produce hard gamma radiation. The ^{236}Pu isotope is formed from (n,2n) and (γ,n) reactions during ^{237}Np irradiation. Calculations of ^{236}Pu contamination due to (n,2n) reactions indicate that light water reactors of current design produce ^{238}Pu containing 20 to 40 times as much ^{236}Pu as the D_2O or graphite production

reactors, and liquid metal fast breeders produce 600 to 900 times as much ^{236}Pu as the production reactors.

LIGHT WATER POWER REACTORS

The Savannah River THOR program was used to calculate the amount of ^{236}Pu. THOR is a burnup code incorporating HAMMER cell calculations (1). The (n,2n) reaction on ^{237}Np was treated by introducing into HAMMER an artificial isotope with absorption cross sections of 240 mb and 2.3 mb in MUFT groups 1 (10 to 7.79 MeV) and 2 (7.79 to 6.07 MeV), respectively. A typical light water power reactor model assumed UO_2 fuel of 0.3669 in. OD, 10.2 gm/cc density, 2.7 wt % ^{235}U enrichment, and clad OD of 0.422 in. These rods were assumed to be on a 0.563-in.-square pitch in H_2O containing 900 ppm boron. This configuration was transformed to a cylindrical model by considering a square array of nine rods where the central rod was replaced by a neptunium target, the four nearest neighbors were replaced by a cylindrical shell of fuel of equal area separated from the neptunium target by a moderator space, and the four next nearest neighbors formed another cylindrical fuel shell separated from the inner fuel region by moderator. The outer fuel shell was surrounded by a moderator region to complete the model.

The neptunium target was assumed to contain 3×10^{-3} neptunium atoms/(bn)(cm), but the results given here were found to be relatively insensitive to neptunium concentrations; the $^{236}Pu/^{238}Pu$ ratio increases slightly as the neptunium concentrations increase. The results were rather insensitive to changes in the cylindrical model, such as considering the eight surrounding rods to be replaced by a single fuel tube instead of two, or including a third fuel ring composed of the next 16 surrounding rods. The fuel enrichment was increased from a nominal design value of 2.7 wt % to 4.6 wt %

to approximately compensate for the loss in reactivity as one-ninth of the fuel rods are replaced by neptunium targets. As more fuel rods were assumed to be replaced by neptunium and the enrichments raised to compensate, the $^{236}Pu/^{238}Pu$ ratio increased.

Figure 1 gives the $^{236}Pu/^{238}Pu$ ratio, Np burnup, and ^{238}Pu/total Pu vs. fuel exposure at a moderator/fuel ratio of 1.267. Figure 2 gives the $^{236}Pu/^{238}Pu$ ratio vs. moderator/fuel ratio for a fuel exposure of 30,000 MWD/MTU. As seen in Fig. 2, no significant change in the $^{236}Pu/^{238}Pu$ ratio is made by increasing the moderator/fuel ratio to any credible value. Table I gives yields and production rates vs. exposure. In calculating the production rate (atoms ^{238}Pu/atoms Np charged per year), 60 days of discharge-ship time was added to the reactor time.

The $^{236}Pu/^{238}Pu$ ratios given above do not include any contribution due to the (γ,n) reaction on ^{237}Np or any $(n,2n)$ events that occur above 10 MeV. The latter effect could increase the $^{236}Pu/^{238}Pu$ ratio approximately 25%. The numbers given roughly agree with ^{236}Pu measurements in Yankee fuel, where at 24,600 MWD/MTU the measured $^{236}Pu/^{238}Pu$ ratio was 24.7 ppm, but where it is estimated that about half the ^{238}Pu present came from α decay of ^{242}Cm, giving an effective $^{236}Pu/^{238}Pu$ ratio of about 50 ppm for the ^{238}Pu produced from captures in ^{237}Np.

LIQUID METAL FAST BREEDER REACTORS

Since the fast breeder reactors are usually considered to be the ultimate successors to the PWR's and BWR's, it is of interest to consider the properties of ^{238}Pu that is made from neptunium irradiation in these reactors. In order to get effective capture and fission cross sections for the isotopes of interest to use

in a burnup calculation, a typical fast reactor spectrum was mocked up in HAMMER by introducing various isotopes (some with fictitious cross sections) into a two-region problem until the desired spectrum was obtained. The inner region was considered the neptunium target region, and the dimensions and neptunium concentration in this region were chosen to approximate an actual neptunium target. The spectrum was mocked up for the Westinghouse Modular Core Liquid Metal Fast Breeder (normal core, WAPD-F1 library) (2), but the results given here should be in the correct range for any such reactor with a very hard spectrum. Relative reaction rates obtained from HAMMER were normalized to the actual core power density to obtain absolute reaction rates per atom for the isotopes of interest.

Figure 3 gives the $^{236}Pu/^{238}Pu$ ratio, Np burnup, and ^{238}Pu/total Pu vs. fuel exposure for the model described above. Table II gives yields and production rates vs. exposure. In addition to the high ^{236}Pu content, the fast reactor has a low ^{238}Pu yield for the neptunium burned because of the large fraction of neptunium fissions in this spectrum. As for the light water reactors, these calculations do not include ^{236}Pu produced from (γ,n) reactions on ^{237}Np or the contribution due to neutrons above 10 MeV.

OTHER CONSIDERATIONS

The $^{236}Pu/^{238}Pu$ ratios given above for the power reactors are to be compared with the 0.8 → 1.0 ppm made in the production reactors. In addition, the production reactors have a high yield and a much higher rate of ^{238}Pu production for the neptunium charged to the reactor because of the combination of higher flux levels and higher cross sections in such reactors. Table III gives yields and production rates for four charges designed to irradiate neptunium in

the D_2O production reactors at Savannah River. Recent measurements made at Hanford indicate that a large contribution to the ^{236}Pu contaminant for neptunium irradiated in the graphite production reactors comes from the (γ,n) reaction on ^{237}Np. The possibility of reducing this effect with proper shielding and target matrix material is being studied.

Current requirements for ^{238}Pu power sources for space applications have an upper limit on ^{236}Pu content of about 2 ppm, and for biomedical applications, no more than about 0.1 ppm. The ^{238}Pu made in production reactors meets the space power specification, and can meet the medical specification by letting the mixture decay a few years before use ($T^{1/2}$ for ^{236}Pu = 2.85 yr). Another possibility is some form of isotopic separation or production of ^{238}Pu by irradiation of ^{241}Am and decay of ^{242}Cm. Blanket irradiation in the power reactors would offer some improvement in ^{236}Pu content, but the rate of production of ^{238}Pu would be prohibitively small.

ACKNOWLEDGMENT

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REFERENCES

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Table I

^{238}Pu Production Rates and Yields of PWR Model

Reactor Time, yr	Exposure, MWD/MTU	$\frac{^{238}Pu}{Np\ Burned}$	$\frac{^{238}Pu}{Np\ Charged}$	$\frac{^{238}Pu}{Np\ Chg\text{-}Yr}$	1-Np BU	$\frac{^{238}Pu}{Tot\ Pu}$
.792	10,700	.87	.11	.11	.88	.94
1.50	20,400	.81	.18	.11	.77	.90
2.21	30,000	.75	.24	.10	.68	.87
2.93	39,700	.69	.29	.092	.59	.84

NOTE: $\frac{^{238}Pu}{Np\ Chg\text{-}Yr}$ includes 60 days discharge-ship time

Table II

^{238}Pu Production Rates and Yields of Fast Reactor Model

Reactor Time, yr	Exposure, MWD/MTU	$\frac{^{238}Pu}{Np\ Burned}$	$\frac{^{238}Pu}{Np\ Charged}$	$\frac{^{238}Pu}{Np\ Chg\text{-}Yr}$	1-Np BU	$\frac{^{238}Pu}{Tot\ Pu}$
.782	34,000	.43	.017	.018	.96	.99
1.56	68,000	.41	.032	.018	.92	.98
2.31	100,300	.39	.044	.017	.89	.97
2.76	120,000	.38	.051	.017	.87	.97

NOTE: $\frac{^{238}Pu}{Np\ Chg\text{-}Yr}$ includes 60 days discharge-ship time

Table III

^{238}Pu Production Rates and Yields of D_2O Model

Type	Reactor Time, yr	$\frac{^{238}Pu}{Np\ Burned}$	$\frac{^{238}Pu}{Np\ Charged}$	$\frac{^{238}Pu}{Np\ Chg\text{-}Yr}$	1-Np BU	$\frac{^{238}Pu}{Tot\ Pu}$
1	.068	.79	.088	.37	.89	.90
2	.137	.81	.095	.33	.88	.90
3	.274	.90	.182	.41	.80	.92
4	.821	.72	.359	.36	.50	.80

NOTE: $\frac{^{238}Pu}{Np\ Chg\text{-}Yr}$ includes 60 days discharge-ship time

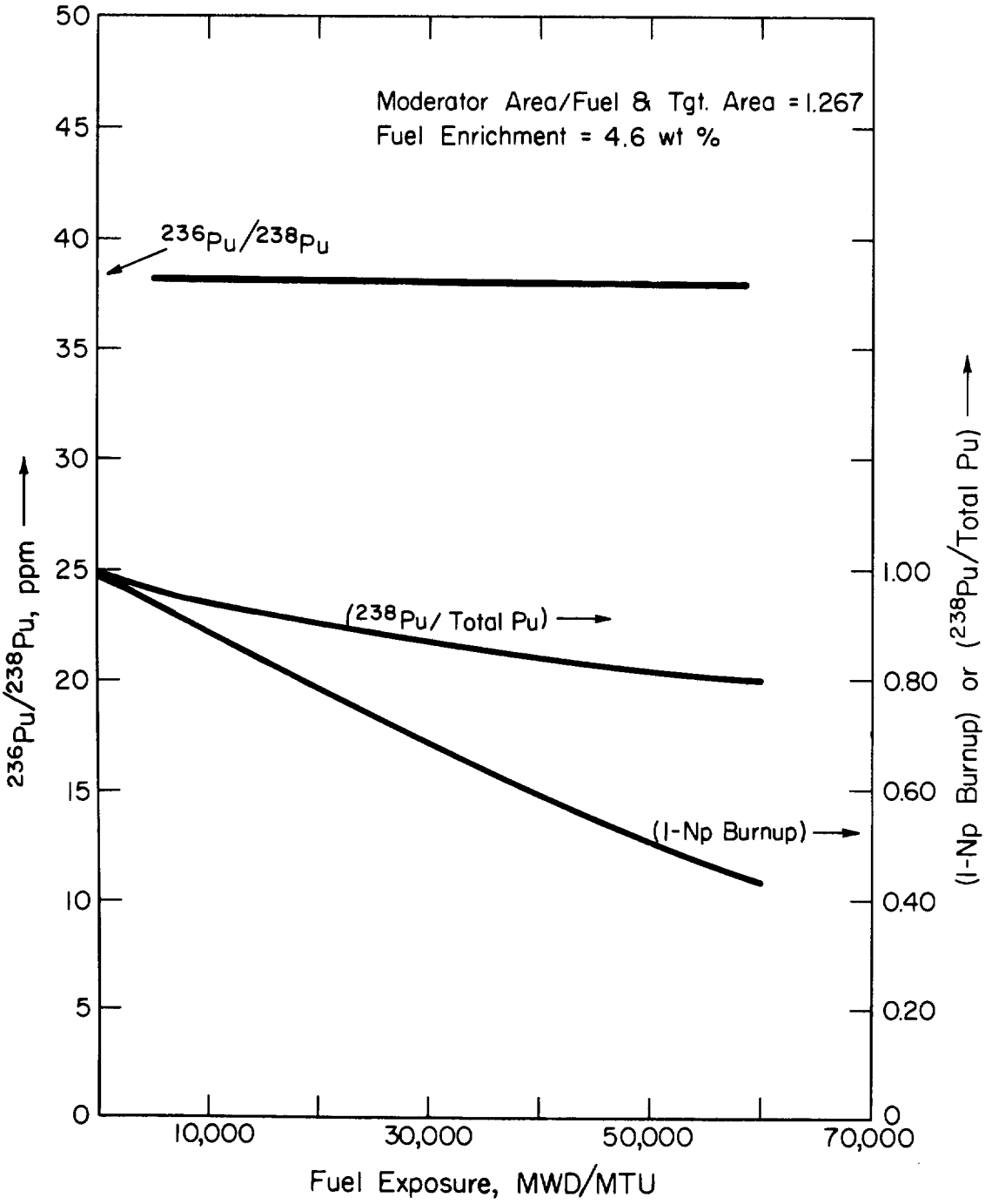

FIG. 1 ^{236}Pu CONTAMINATION IN ^{238}Pu PRODUCED IN PRESSURIZED LIGHT WATER REACTORS (Exposure Effects)

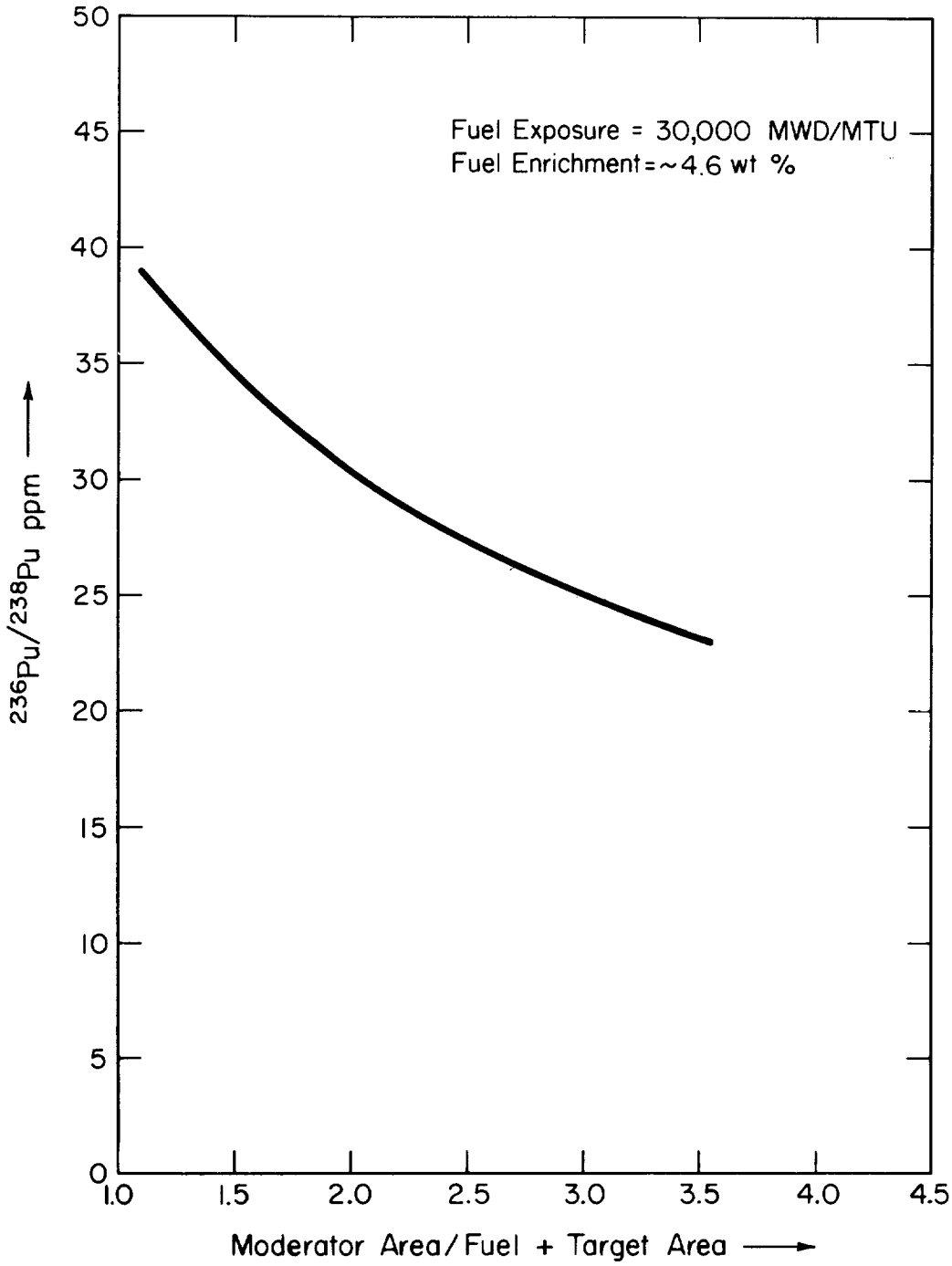

FIG. 2 ^{236}Pu CONTAMINATION IN ^{238}Pu PRODUCED IN PRESSURIZED LIGHT WATER POWER REACTORS (Spectrum Effects)

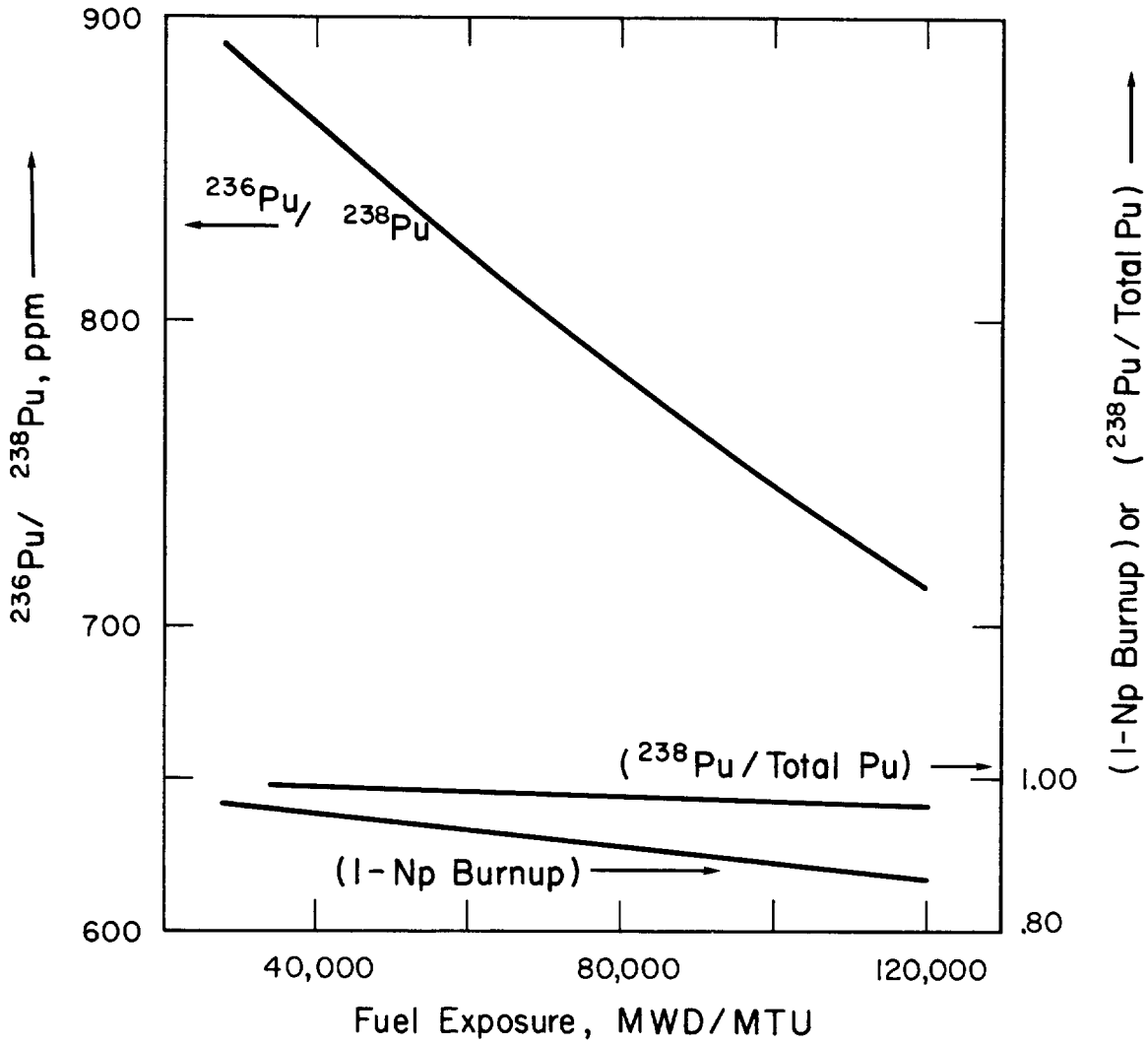

FIG. 3 ^{236}Pu CONTAMINATION IN ^{238}Pu PRODUCED IN FAST BREEDER REACTORS (Exposure Effects)