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AMERICIUM, CURIUM, AND PLUTONIUM YIELDS IN SRP HIGH FLUX IRRADIATIONS

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Chemistry
(TID-4500)

AMERICIUM, CURIUM, AND PLUTONIUM YIELDS IN SRP HIGH FLUX IRRADIATIONS

by

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ABSTRACT

Analyses of sections of four $^{242}\text{PuO}_2$ targets irradiated at high flux in SRP reactors indicate that the ^{244}Cm and ^{245}Cm yields are near equilibrium between exposures of $5 \times 10^{22} \text{ n/cm}^2$ and $9 \times 10^{22} \text{ n/cm}^2$. High fission losses reduce yields of higher mass isotopes at these exposures. The yields of ^{246}Cm , ^{247}Cm , and ^{248}Cm are still increasing at $9 \times 10^{22} \text{ n/cm}^2$. Fission products correspond to the consumption of ~29% of the initial plutonium after an exposure of $9 \times 10^{22} \text{ n/cm}^2$.

AMERICIUM, CURIUM, AND PLUTONIUM YIELDS IN SRP HIGH FLUX IRRADIATIONS

A total of 520 g of ^{242}Pu , including eight $^{242}\text{PuO}_2$ slugs, were irradiated at high flux [$\sim 3 \times 10^{15}$ n/(cm²)(sec)] in SRP reactors to produce higher mass isotopes to serve as target material for the High Flux Isotopes Reactor (HFIR) at Oak Ridge.¹ Production rates of the transplutonium isotopes were needed to develop effective cross sections for extended high flux irradiation; for this purpose, four of the $^{242}\text{PuO}_2$ slugs were discharged and analyzed after exposures ranging from 3.7×10^{22} to 9.3×10^{22} n/cm². Nuclides through ^{248}Cm have been determined from the first of these slugs (P-3).² Similar determinations on slugs P-4, P-5, and P-6 are summarized in this report. Yields of transcurium nuclides in all four slugs have been reported by Holcomb.³

All four target slugs were of identical size, shape, and ^{242}Pu content. Duplicate wafers representing about one tenth of the total core weight were cut from the center of each slug. The wafers were then digested in NaOH. The reaction mass was acidified with HNO₃-HF, and then diluted, sampled, and analyzed as described previously.² Exposures were computed from power estimates and also by calculations in which a 17.5-barn burnup cross section for ^{242}Pu was used.

Americium, curium, and plutonium determinations in all four slugs are summarized in the table. These results are fitted to yield-exposure curves in the figure with a computer program for multiple neutron captures.⁴ In this method, key cross sections are varied, and an iterative approach is used until the observed actinide yields are obtained. The resulting set of cross sections can be used for calculating yields from other irradiations of this type. These calculations have been reported in detail.⁴

Mass spectrometric analyses of the curium fraction indicate that ^{247}Cm and ^{248}Cm have equal yields at about 6×10^{22} n/cm². Yields of ^{246}Cm , ^{247}Cm , and ^{248}Cm continue to increase even after exposures

of 9×10^{22} . ^{245}Cm with its rather high fission cross section is roughly in equilibrium with the ^{244}Cm throughout this range. Fission products, which account for less than 9% of initial plutonium after an exposure of 3.7×10^{22} , correspond to as much as 29% of initial plutonium after an exposure of $9 \times 10^{22} \text{ n/cm}^2$.

FIG. 1 YIELDS OF ACTINIDES CALCULATED WITH DEDUCED CROSS SECTIONS

Analyses of ^{242}Pu Irradiated at High Flux

Target	<u>P-3</u>	<u>P-4</u>	<u>P-5</u>	<u>P-6</u>	
Exposure, 10 ²² n/cm ²					
Estimated ^a	3.7	5.3	7	9	
Calculated ^b	3.8	5.2	7.3	9.3	
Actinide, g per initial g ²⁴² Pu					
²⁴² Pu	0.511	0.402	0.276	0.194	
²⁴³ Am	0.128 ^c	0.122	0.091	0.065	
²⁴⁴ Cm	0.276	0.374	0.417	0.430	
Fission Products, g per initial g plutonium irradiated ^d					
	0.088	0.147	0.214	0.29	
Total Products, g per g plutonium irradiated					
	0.991	1.05	1.02	1.03	
Isotopic Composition, wt %					
	<u>Initial</u>				
²³⁸ Pu	0.44	<5 x 10 ⁻³	<2 x 10 ⁻³	-	<3 x 10 ⁻⁵
²³⁹ Pu	0.35	0.017	0.003	0.008	<3 x 10 ⁻⁵
²⁴⁰ Pu	1.71	0.112	0.11	0.401	0.460
²⁴¹ Pu	0.62	0.023	0.040	0.134	0.137
²⁴² Pu	96.89	99.53	99.35	98.71	98.29
²⁴⁴ Pu	~0.05	0.321	0.50	0.733	1.12
²⁴⁴ Cm		96.63	95.33	93.58	91.47
²⁴⁵ Cm		0.580	0.535	0.504	0.487
²⁴⁶ Cm		2.71	3.97	5.70	7.69
²⁴⁷ Cm		0.044	0.072	0.104	0.141
²⁴⁸ Cm		0.022	0.06	0.116	0.208

^aFrom power data

^bUsing 17.5-barn burnup cross section for ^{242}Pu

^cCorrected from value in DP-1046

^dAssumes that the mean effective mass of fissioning nuclei is ^{242}Pu (P-3), ^{243}Pu (P-4), ^{245}Pu (P-5), and ^{245}Pu (P-6) with ^{137}Cs fission yields of 6.0% for mass 242 and 243 and 7.9% for mass 245. ^{137}Cs was determined by the tetraphenylboron method.

REFERENCES

1. J. L. Crandall. *The Savannah River High Flux Demonstration*. USAEC Report DP-999, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1965).
2. E. K. Dukes, R. L. Folger, C. J. Banick, G. A. Carothers, M. Y. Donnan. *Yield of Curium-244 in the SRP High Flux Reactor*. USAEC Report DP-1046, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1966).
3. H. P. Holcomb. *Yields of Transcurium Nuclides in the SRP High Flux Reactor*. USAEC Report DP-1137, E. I. du Pont de Nemours and Co., Savannah River Laboratory, Aiken, S. C. (1967).
4. J. A. Smith, C. J. Banick, R. L. Folger, H. P. Holcomb, and I. B. Richter. *Reactor Cross Sections for ^{242}Pu - ^{252}Cf* . Paper presented at the Conference on Neutron Cross Sections and Technology, Washington, March 1968. To be published in the Proceedings of the Meeting.