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NUCLEAR DATA FOR ACTINIDE RECYCLE

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ABSTRACT

Large quantities of heavy actinides will be byproducts of burning plutonium and uranium fuel in nuclear power reactors. Chemical recovery and subsequent recycle in reactors will be for the production of ^{238}Pu and ^{244}Cm as heat sources and ^{252}Cf as a unique source of neutrons or for the ultimate conversion of heavy actinides to shorter-lived fission products to reduce long-term waste storage problems. Test measurements and production yields have provided data for developing a consistent set of multigroup cross sections which give excellent predictions of actinide concentrations in a variety of reactor environments and exposures. These multigroup cross sections are compatible with advanced reactor theory calculational codes.

(Key Words: transplutonium, cross section sets, actinide burnup, yield, waste management, neutron spectrum)

Introduction

The actinides found in sizable quantities in nature are thorium and the isotopes of uranium. Heavier actinides have become available in substantial quantities from the operation of fission reactors. Large-scale recycling of these actinides has occurred for about 15 years.[1] For example, current ERDA production programs have resulted in transmutation of significant quantities of actinides to ^{238}Pu, $^{241,243}Am$, ^{244}Cm, and ^{252}Cf, in addition to plutonium for reactor fuel and nuclear weapons uses. These production programs are continuing. They have been successful because there are efficient chemical separation and target reprocessing facilities that permit recycling of unique target material for further irradiation.

Four years ago at the last conference,[2] a self-consistent two-group set of cross sections for the isotopes of curium and californium was presented. It was based primarily on data obtained from the ^{244}Cm and ^{252}Cf production programs. An improved 84-group set has been reported at this conference.[3] It is based on the original data plus many additional detailed measurements and analyses. The cross sections are available for conversion to the pointwise ENDF/B format for use in advanced reactor calculational codes. This paper concentrates on the applicability of the new neutron cross section data to the evaluation of concepts that have been suggested for the management of recycling heavy transplutonium actinides.

In the future, the largest actinide recycle program will involve the recycle of uranium and plutonium fuel in power reactors. The recycle of these fuels is of economic importance to nuclear power. Proposed sodium-cooled fast-breeder reactors also require plutonium recycle to burn the bred plutonium as fuel. Nuclear data for these operations as well as recyclings involving thorium-^{233}U systems, such as for the gas-cooled reactors and the molten salt reactor, are available, but they are not covered in this paper. Recycle of $^{241,243}Am$ and ^{237}Np is discussed briefly.

One consequence of uranium and plutonium recycle in nuclear power reactors is the gradual buildup of the heavy actinides neptunium, americium, curium and californium. This prospect stimulated projections, six years ago, that very large quantities would become available for applications.[4] Applications include ^{238}Pu and ^{244}Cm as isotopic heat source material for space or terrestrial missions and ^{252}Cf as a unique portable source of neutrons, already useful for such widely diverse applications as the treatment of tumors and neutron radiography in industry. Recently others have suggested that large accumulations of byproduct heavy actinides in process waste or separate residues provide a serious problem for long-range storage in safe repositories because half-lives are long ($>10^3$ years). It has been suggested that transmutation and/or fission would reduce storage to the more manageable hazard of fission products.[5]

A study was made to determine if the current nuclear data are adequate for evaluating heavy actinide production and/or removal programs. The results show the data to be adequate for such purposes. Large-scale actinide burnup late in this century may require improved fission data because irradiation of the expected large quantities could alter reactor charge design characteristics and reactivity transient effects. Such data can certainly be provided within the required time span, particularly as more actinide material becomes available. There is no urgency for the improved data to begin to recycle the lesser amounts expected in the next decade or two.

Actinide Production

Actinide production calculated on the basis of the 84-group set of cross sections[3] agreed very well with actual transplutonium production programs. To apply these new cross sections to other studies, users must realize the sensitivity of calculations to possible errors from uncertainties in neutron exposure and neutron spectrum. Possible errors are comparable to experimental errors in the data used to derive the cross section set originally (Figures 1-3). Figure 1 shows the calculated-to-experimental yield ratio for three of the data sets used to verify the cross section set. Figure 2 shows the effect of 10% lower neutron exposures on nuclide yields for three typical exposures. Figure 3 shows the sensitivity of neutron spectrum to nuclide yields at two typical exposures for 10% reductions in the fast-to-thermal flux ratio (ϕ_f/ϕ_{th}). In these calculated

cases, ^{244}Cm is assumed as the starting target material. The plots in Figures 2 and 3 show how important correct determination of irradiation conditions is in making initial projections of transplutonium production. The production calculations were made using two-group cross sections derived from the new 84-group cross section set. The consistency of the cross sections and most of the experimental data for a variety of irradiation conditions suggests that experimental values in Figure 1 lying outside ±10% are possibly suspect and that any data trends similar to those calculated for Figures 2 and 3 indicate that possible errors in the estimates of irradiation conditions should be investigated. Equivalent overestimates of these variables would give mirror image plots rotated about the yield ratio of unity.

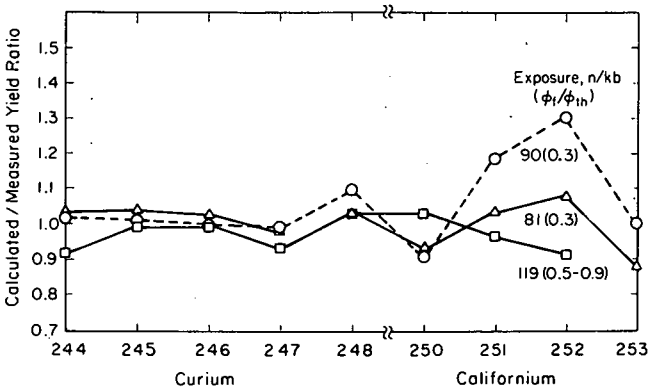

FIG. 1 Transplutonium Burnup: Comparison of Calc/Meas Yield Ratio for Typical Test Samples

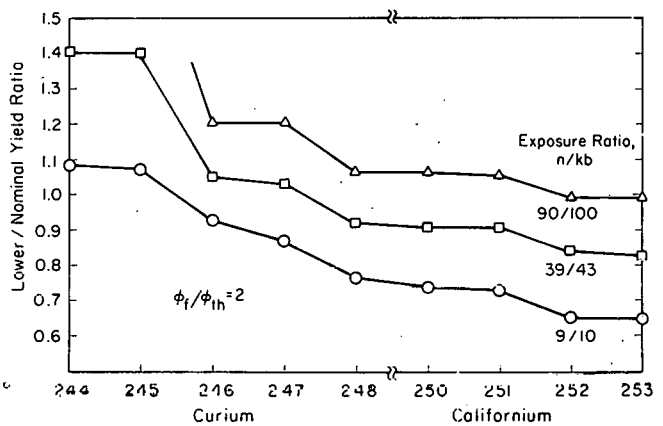

FIG. 2 Calculated ^{244}Cm Burnup: Effect of 10% Lower Exposure on Yield Estimates

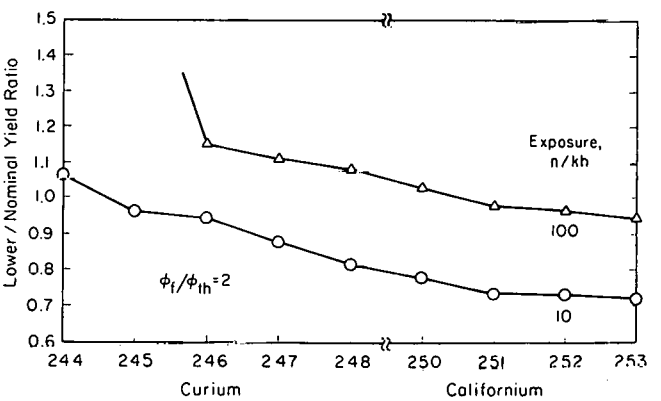

FIG. 3 Calculated ^{244}Cm Burnup: Effect of 10% Lower ϕ_f/ϕ_{th} Ratio on Yield Estimates

Actinide Burnup

Official criteria for setting goals for actinide burnup have not been established. The goal for decay of fission products has been set nominally to be ∿600 years of storage[5], equivalent to about 20 half-lives of ^{137}Cs and ^{90}Sr or a 10^6 reduction. Using similar criteria, the goal for the burnup of transplutonium actinides could be set to be ∿40 years, equivalent to a professional career or to the approximate lifetime of the nuclear plant in which the actinides were generated. An effective burnup half-life of about two years to give the 20 half-lives or the 10^6 reduction, seems to be a reasonable burnup rate and will be used as a goal in this recycle study.

Two important aspects of any burnup program are yield (Table 1) and rate (Table 2). The heavy actinides can be fissioned; transmutation to an inactive nuclide is not feasible. The amount fissioned in present day reactors on a single irradiation is 99.44% through ^{252}Cf and subsequent burnup can reduce the actinide reduction to the goal of ∿10^6. Use of another consistent two-group cross section set developed at HFIR[6] shows a nearly identical fission yield of 99.42%. The ORNL set provides similar but not identical cross sections. Thus, two independently developed and tested production cross section sets give nearly identical high fission yields for the transplutonium chain.

TABLE 1

ACTINIDE BURNUP YIELDS

Nuclide	Fiss/Abs	Fissioned*
^{243}Am	0	0
^{244}Cm	0.05	5
^{245}Cm	0.86	87
^{246}Cm	0.09	88
^{247}Cm	0.59	95
^{248}Cm	0.05	95
^{250}Cf	0	95
^{251}Cf	0.65	98.4
^{252}Cf	0.65	99.44

* Cummulative percent of original mass that is fissioned.

TABLE 2

ACTINIDE REMOVAL RATE

Nuclide	Half-Life, years Decay	Burnup PWR	Burnup D_2O	Burnup HPD
^{243}Am	737C	1.5	1.3	0.14
^{244}Cm	18	4.9	4.9	0.48
^{245}Cm	8500	0.75	0.12	0.04
^{246}Cm	4760	23	24	2.2
^{247}Cm	15 x 10^6	2.1	1.2	0.18
^{248}Cm	35 x 10^4	10.7	11.6	1.05
^{249}Bk	0.85	0.6	0.1	0.03
^{249}Cf	352	0.5	0.1	0.03
^{250}Cf	13	0.2	0.1	0.02
^{251}Cf	900	0.17	0.04	0.01
^{252}Cf	2.6	13	4.6	0.92
^{253}Cf	0.05	0.9	0.2	0.05

The rate can be evaluated by simple two-group calculation of burnup half-lives, i.e., the time it takes for a neutron exposure to reduce a nuclide concentration by 50%. Burnup half-life, years = $2.17 \times 10^{16}/\phi\sigma$, where ϕ_{th} = n/(cm²-sec) and σ = cross section, barns. Table 2 compares calculated actinide burnup half-lives to decay half-lives for three nominal irradiations. The column labeled PWR is for a nominal pressurized water reactor ($\bar{\phi}_{th} = 2 \times 10^{13}$ n/(cm²-sec); $\phi_f/\phi_{th} = 6$). The column labeled D_2O is for a nominal D_2O reactor ($\phi_{th} = 10^{14}$ n/(cm²-sec); $\phi_f/\phi_{th} = 2$). The column labeled HPD is for a special high power density reactor charge ($\bar{\phi}_{th} = 4 \times 10^{14}$ n/(cm²-sec); $\phi_f/\phi_{th} = 3$). This latter concept is similar to a modified Savannah River high flux charge.[7] It was created, calculationally, because the other two reactor types gave unacceptably long burnup half-lives for the rate-limiting isotope ^{246}Cm. The net effective calculated half-life for ^{244}Cm burnup to 10^{-6} total actinides is ~42 years for the HPD charge, a reasonable expectation. The burnup and the resulting actinide residues are shown in Figure 4. The effective burnup half-life in a PWR for the precursor plutonium isotope chain is short (<4 years) and is accomplished to improve fuel cycle efficiency.

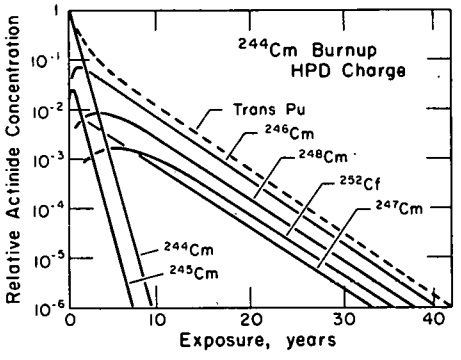

FIG. 4 ^{244}Cm Burnup in HPD Charge

Burnup halflives computed with ORIGEN[8] built-in two-group cross sections would be similar to the PWR results in Table 2, differing only in detail for the heavier curium isotopes. However, the 84-group, new cross sections can be used with modern reactor codes to give more precise calculations and to account correctly for self shielding of the even numbered curium isotopes in practical irradiation environments.

The data in Table 2 show that burnup can be achieved in a reasonable time by using a reactor with high power density but not with conventional thermal reactors. The cross section set is sufficiently precise so that this important conclusion will not be modified by future refinements in the data. If few applications are developed for ^{244}Cm and if large quantities of transplutonium actinides are to be burned up, it may be necessary to operate in two stages. The first would be in conventional thermal reactors to burn up large quantities of ^{243}Am-^{244}Cm mixtures and to remove the substantial heat from ^{245}Cm fission (87% of the transplutonium fissions occur at this isotope of curium). The second stage at high power density would be to burn up the remaining actinide residues, predominately ^{246}Cm, the burnup rate-limiting nuclide. The ultimate goal could be either a ^{252}Cf product or fission products. Both goals, production or burnup, are served better in a high power density reactor charge.

Fission product yield data for the actinides are available and show that no additional unwanted fission product is produced in excess of that already made by conventional fissions in ^{235}U and Pu.[9] From the projected fissile plutonium inventory, world wide, of about 680 tons[10] by the year 2000, we could expect the accumulated transplutonium actinides to approach at least 100 tons at that time. Either burnup or production techniques would need to displace substantial portions of thermal reactor charges selected for such operations. Actinide irradiation in LMFBR's might also be possible, at that time, and cross section data now available can be used to estimate the feasibility if the permissible LMFBR irradiation conditions of exposure and spectrum could be defined for the large quantities involved. The potential adverse effect of actinide burnup on the breeding of plutonium fuel would be a major concern to LMFBR irradiations of actinides. However, large-scale LMFBR operation is expected to reduce actinide residue output substantial because of increased plutonium fissioning.

Large-scale transplutonium actinide recycle will of course involve substantial development work. Targets to contain the actinides would require special handling, would generate heat from curium alpha decay, and would operate as fuel during early burnup stages. Concentration of actinides in target assemblies would provide self-shielding that would reduce rate-limiting burnup rates of ^{246}Cm and ^{248}Cm, thus extending actinide burnup times. Large-scale chemical processing facilities would also have to be built to separate the actinides for initial irradiation and for recycle to new targets as irradiation proceeds. In the forecast of 1968, it was projected that there would be "Tons of Curium and Pounds of Californium."[4] The magnitude of an actual recycle late in this century, for whatever purpose, could approach a "Ton of Californium."

^{238}Pu Production

Both ^{237}Np and ^{241}Am will accumulate in large quantities as a consequence of nuclear power operation and growth.[10] The ^{237}Np can be recycled to produce large amounts of ^{238}Pu, a product that has been useful as a power source in space exploration, and that is also intended for heart pacemakers and heart assist devices. ^{241}Am is separated from power reactor fuel along with a companion isotope, ^{243}Am. If recycled and burned up, it can be a source of very high-purity ^{238}Pu via alpha decay of the separated irradiation product ^{242}Cm.[11] The eventual burnup of the other isotope, ^{243}Am, has already been discussed. Nuclear data for these types of irradiation are well established and have been applied to plant-type operations.

In the burnup of ^{241}Am, some minor uncertainties seem to exist in the choice of a branching ratio for ^{242}Am, which leads to the simultaneous formation of of ^{242}Cm and ^{242}Pu. ^{242}Am is formed by neutron capture in ^{241}Am. Resolution of the uncertainties presented by differing data would be achieved promptly if a large-scale irradiation program were ever to become a reality. In short, for recycle of ^{237}Np and $^{241,243}Am$, current ENDF/B data appear to be quite adequate for recycle estimation and planning. ^{236}Pu, an unwanted impurity in ^{238}Pu and in recycled plutonium fuel,* is formed by high energy (n,2n) and (γ,n) reactions on ^{237}Np. The nuclear data for these reactions are also well understood and readily available.[12]

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* ^{236}Pu daughters emit hard gammas that make handling plutonium more difficult as ^{236}Pu decays.

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References

1. D. Perlman. "The Atomic Archeologist: Dr. Glenn T. Seaborg." *Saturday Review*, p 52 (March 5, 1960).

2. E. J. Hennelly. "The Californium Cross Section Story." p 494 in *Third Conference on Neutron Cross Sections and Technology*. CONF-710302, Vol II (1971).

3. R. W. Benjamin, et al. "A Consistent Set of Transplutonium Multigroup Cross Sections." Presented at the *Conference on Nuclear Cross Sections and Technology*, Washington, DC, March 3-7, 1974.

4. J. L. Crandall. "Tons of Curium and Pounds of Californium." p 193, *Proceedings of International Conference of Constructive Uses of Atomic Energy*. R. Franks (Ed.), American Nuclear Society, Hinsdale, Ill. (1968).

5. *Atomic Energy Clearing House*, Vol. 21, No. 1, p 7, Congressional Information Bureau, Inc., Washington, DC (January 6, 1975). "High Level Radioactive Waste Management Alternatives - Vol. 4. Sec. 9, BNWL-1900 (May 1974).

6. J. L. King, J. E. Bigelow, and E. D. Collins. *Transuranium Processing Plant Semiannual Report of Production, Status, and Plans for Period Ending December 31, 1973*. USAEC Report ORNL-4965 p 24, Oak Ridge National Laboratory, Oak Ridge, TN (1974).

7. E. J. Hennelly. "Large-Scale Production of Radioisotopes." Ch. 2, p 86 in *Radioisotope Engineering*, (G. G. Eichholz, ed.), Marcel Dekker, Inc., NY (1972).

8. M. J. Bell, *ORIGEN*. USAEC Report ORNL-4628, Oak Ridge National Laboratory, Oak Ridge, TN (1973).

9. K. W. MacMurdo and R. M. Harbour. "Mass Yields from Thermal-Neutron Induced Fission of ^{245}Cm." *J. Inorg. Nucl. Chem 34*, 2109 (1972).

10. *Nuclear Power Growth: 1974-2000*, USAEC Report WASH-1139, p 33-34, U. S. Government Printing Office, Washington, DC (1974).

11. C. J. Banick and M. H. Goosey. *Improved Methods for Determination of ^{236}Pu in ^{238}Pu*. USAEC Report DP-1291, Savannah River Laboratory, E. I. du Pont de Nemours & Co., Aiken, SC (1972).

12. C. K. Paulson and E. J. Hennelly. "Cross-Section Measurement of Plutonium-236 Formation in Plutonium-238 by ^{237}Np (n,2n) Reactions." *Nucl. Sci. Eng. 55*, 24 (1974).