Review Paper A 7

Some Activities in the United States Concerning the Physics Aspects of Actinide Waste Recycling*

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Abstract

This review paper briefly discusses the reactor types being considered in the United States for the purpose of actinide waste recycling. The reactor types include thermal reactors operating on the 3.3% ^{235}U-^{238}U and the ^{233}U-^{232}Th fuel cycles, liquid metal fast breeder reactors, reactors fueled entirely by actinide wastes, gaseous fuel reactors and fusion reactors. This paper also discusses cross section measurements in progress or planned toward providing basic data for testing the recycle concept.

INTRODUCTION

One of the most urgent needs of the nuclear power industry today is a viable plan which is technically sound and has public acceptance, for the long-term management of radioactive waste. Many suggestions are being considered for dealing with these wastes in terms of disposal or storage such as retrievable storage in near surface vaults and deep underground mines, irretrievable disposal in salt domes, deep holes drilled in stable rock formations, in the sea, outer space, etc. Although it may be feasible to package and safely store wastes for periods as long as a thousand years, there is no way to ensure complete containment for the more than 250,000 years necessary for the actinides (and a few fission products) to decay to innocuous levels. One suggested method for alleviating this problem would involve removing nearly all of the long-lived actinides (thorium, protactinium, uranium, neptunium, plutonium, americium, and curium) contained in the wastes and disposing of this fraction by transmutation to fission products (which decay essentially to nonradioactive isotopes in less than 1000 years).

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Recycling requires a high-flux neutron source, which might be the same reactor producing the wastes or a specially designed burner reactor or even a fusion reactor. Transmutation calculations require neutron cross sections which are more complete and of higher accuracy than are now available. This review paper will focus attention on only two aspects of the actinide waste problem: (a) reactor types under consideration for recycling and (b) cross section measurements in progress or planned in support of the recycling concept. The specific cross section requirements have been outlined in a separate review paper [1]. It should be mentioned at the outset that recycle calculations carried out till now are preliminary in nature. They are in the form of feasibility studies carried out quickly with whatever cross section data were readily at hand. The computer codes used for these calculations contain no provisions for entering data uncertainties. Detailed sensitivity studies have not been carried out to the best of our knowledge.

RECYCLING IN THERMAL REACTORS

The first reported detailed study of actinide recycling was carried out by Claiborne [2] who considered recycling in light-water reactors (LWR) operating on the 3.3% ^{235}U-^{238}U fuel cycle. The reductions in the hazards due to actinides are brought about in two ways. U and Pu recoveries of up to 99.5% have been attained in the conventional reprocessing of LWR fuel; in some cases, 90 to 95% of neptunium has also been recovered, but Am and Cm have been routinely discharged in the high-level waste. Claiborne [2] showed that it is possible to reduce the hazard (at >1000 years) associated with high-level wastes to values comparable to those from naturally occurring uranium deposits provided that 99.99% of the Pu, 99.9% of the U, Am, Cm and ^{129}I, and 95% of the Np are recovered from LWR fuels. Except perhaps for U and Pu, the removals required are beyond present technology.

If essentially all of the actinides normally discharged in the waste are recovered and recycled in a reactor, the inventory becomes stabilized at a value several times the quantity produced in the first cycle. In other words, the inventory approaches a maximum value, an asymptote, rather than increasing linearly with time (see Figure 1). Claiborne [2] showed that through the combination of chemical separations and recycling, the hazards in the fission product wastes can be reduced by a factor of 50 to 200. Claiborne's calculations have been repeated and extended by the group at Battelle Pacific Northwest Laboratories [3] with similar results. The technical assessment of transmutation is only one of several topics discussed in the Battelle study [3] which is a comprehensive review of potential alternative methods for long-term management of high-level radioactive waste. This study identified continued recycling in thermal power reactors as the

most promising method of transmutation of actinides within the constraints of existing technology provided that the chemical separation problems can be solved.

The low flux level of 3 x 10^{13} n/cm^2 sec used by Claiborne in his calculations results in long irradiation times of approximately 50 to 60 years. Even then, sustained recycling of actinides reduces the overall inventory of the actinides by only a factor of 3. The calculations carried out by Raman, Nestor and Dabbs [4] showed that further reductions in actinide inventories, especially those due to transuranium isotopes, should be possible through recycling in a ^{233}U-^{232}Th reactor. In such a reactor, the capture of neutrons by the ^{232}Th fertile material leads to the replenishment of the original ^{233}U fuel. Successive neutron captures by ^{233}U result in higher U isotopes until ^{237}U is reached, where a higher Z isotope, ^{237}Np, is produced. It is further obvious that plutonium and transplutonium isotopes are generated to a far lesser extent in a ^{233}U-^{232}Th reactor when compared to a ^{235}U-^{238}U reactor. For nuclides above ^{238}Pu, the typical reduction factor is >10^6 because in the ^{233}U case, five additional neutron captures are required to reach the same mass. Therefore, due to the absence of substantial new production of these elements, the ^{233}U-^{232}Th reactor can be effectively employed to reduce the Np, Pu, Am and Cm inventories.

RECYCLING IN FAST REACTORS

The ultimate aim in recycling actinides is to induce fission because transmutation of one actinide to another probably has little potential for decreasing toxicity. It was recognized early in the recycling studies that the use of fast reactors should result in faster burnup than in thermal reactors. The fission to capture ratio is generally higher for fast reactor neutron spectra. The combination of flux and cross section results in higher fission rates and lower capture rates as shown in Table I. Preliminary calculations for the burnup of actinides in a Liquid Metal Fast Breeder Reactor (LMFBR) have been carried out by Breen [5] and by Beaman [6]. The equilibration of actinides recycled in an LMFBR is qualitatively similar to that shown in Figure 1 except that equilibrium is approached in a shorter time and the equilibrium values are lower suggesting a more efficient approach for the removal of the actinides. Kubo and Rose [7] have pointed out that recycling of the actinides in an LMFBR has several additional advantages over recycling in a thermal reactor. Extreme separations of chemical groups may not be required because a less pure actinide product should be recyclable without degrading the neutron economy in an LMFBR. The incorporation in the fuel element of the actinides to be recycled should not be too difficult since the fabrication of LMFBR fuel

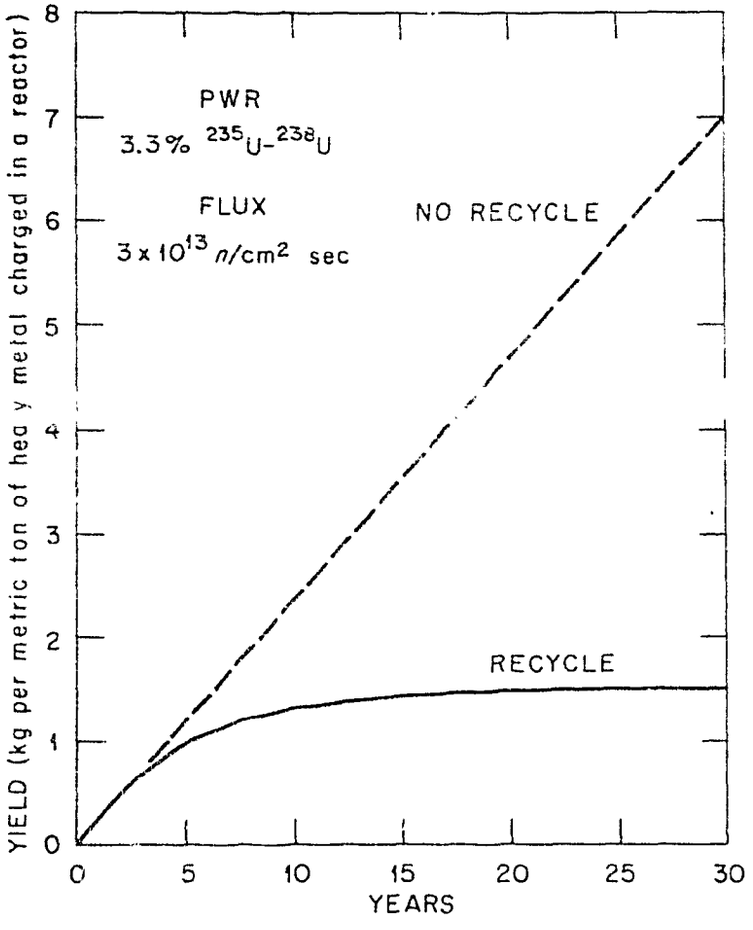

Figure 1. Influence of recycling in the yield of selected actinides (^{237}Np + ^{241}Am + ^{242}Am + ^{243}Am + ^{242}Cm + ^{243}Cm + ^{244}Cm + ^{245}Cm). Figure based on calculations given in Reference 3.

TABLE I

ACTINIDE REACTION RATES IN FAST AND THERMAL REACTORS[†]
(Reactions/sec/Atom)

		Fast Spectrum*		Thermal Spectrum**	
Isotope	Half-Life, Years	Fission Reaction Rate	Capture Reaction Rate	Fission Reaction Rate	Capture Reaction Rate
Np^{237}	2.14×10^6	2.2×10^{-9}	1.03×10^{-8}	6.18×10^{-12}	4.9×10^{-8}
Am^{241}	433	2.7×10^{-9}	2.35×10^{-8}	6.18×10^{-10}	1.38×10^{-7}
Am^{242m}	152	4.7×10^{-8}	9.69×10^{-9}	1.49×10^{-7}	1.33×10^{-7}
Am^{243}	7370	1.39×10^{-9}	4.5×10^{-9}	1.55×10^{-11}	3.18×10^{-8}
Cm^{244}	17.9	3.47×10^{-9}	2.77×10^{-9}	4.02×10^{-10}	5.9×10^{-9}

[†]From Reference 6
*Average Total Flux = 6.93×10^{15} in Core Zone 1
**Average Total Flux = 3.09×10^{14}

elements would already include provisions for handling the highly toxic Pu isotopes.

RECYCLING IN OTHER TYPES OF REACTORS

A group of fifteen senior and graduate students working with Binney and Spinrad [8] of Oregon State University (Nuclear Engineering Department) have carried out interesting studies on the use of fast reactors for the destruction of actinide wastes. Initially, they considered liquid metal fast breeder and gas cooled fast breeder reactors. They found that the actinide destruction rates in such reactors was better but not by what they [illegible] significant factor over rates to be expected in thermal reactors. These reactors are not really "fast" but intermediate and fail to take full advantage of the excellent neutron economy of the fast chain reaction. The follow-up study focused attention on hard-spectrum (>100 keV) fast reactors of three types: sodium-cooled, metal fueled; sodium-cooled, carbide fueled; and helium-cooled, carbide fueled. The fuel, and this is the interesting part, consisted of all the higher actinides (Np, Am, Cm, and 0.5% of Pu from twenty 1000 MWe LWRs). It was found that a reactor with a core of the size of the Idaho Experimental Breeder Reactor II (EBR-II) or smaller would go critical on essentially pure actinide fuels with a margin of reactivity sufficient to permit some considerable dilution. A key question was the extent to which dilution with inert materials such as Zr and ZrC could be tolerated and still maintain very high specific powers. To answer this question and to explore fully the possibilities of fast reactors operating on actinide fuels, extensive data (σ_f, ν, η, etc.) for E_n > 100 keV would be needed. In the absence of such data for ^{241}Am, ^{243}Am and ^{244}Cm, Binney and Spinrad and their coworkers were forced to employ the known cross sections for ^{240}Pu for all three of these materials.

Paternoster, Ohanian, Schneider and Thom [9] have considered the use of a conceptual gaseous core reactor for transmutation of fission product (^{129}I and ^{99}Tc) and actinide wastes (Am and Cm). The main feature of such a reactor is the use of fissile fuel in a gaseous or plasma state thereby breaking the barrier of temperature imposed by solid-fuel elements. The fuel is UF_6 enriched to 6 percent ^{235}U. A reactor with a continuous fuel cycle (whether of the gaseous type or the molten salt type) requires a much lower inventory of fissile and fertile material. With continuous fuel addition and removal of xenon and other volatile fission product poisons, higher fuel burnup rates can be achieved. For these reasons, the UF_6 gas core reactor generates less actinide waste than the conventional LWR. The rates of transmutation were found to depend strongly on the ratio of resonance-to-thermal neutron fluxes. Since the neutron energy spectrum of a gas core

reactor is not known with certainty, Paternoster et al considered three representative values 0.058, 0.29 and 0.58 for the resonance/thermal ratio. A five-year transmutation of Am and Cm wastes resulted in an order-of-magnitude decrease in the overall hazard potential of actinide wastes generated in 60 reactor-years of LWR operation. The calculations assumed an average thermal neutron flux of 6.4×10^{14} n/cm^2 sec.

RECYCLING IN FUSION REACTORS

In 1973, Wolkenhauer, Leonard and Gore [10] made a survey study of actinide transmutation in the blanket of a conceptual thermonuclear reactor (CTR). Calculations were made for neutrons from both a D-D and a D-T plasma. The authors assumed two wall loadings, 1.0 MW/m^2 and 10.0 MW/m^2, in order to determine the neutron flux. With Be in the blanket by which the 14-MeV neutrons from the D-T reaction could be multiplied by a factor of 2.5, thermal neutron fluxes of 3×10^{15} and 3×10^{16} were obtained with the two wall loadings. These flux levels are 100 to 1000 times higher than in an LWR. Therefore, it was possible to consider not only the rapid transmutation of actinides but also the transmutation of problem fission products such as ^{85}Kr, ^{90}Sr, ^{137}Cs and ^{129}I. In addition to the fission and capture cross-sections, the CTR studies established the need for (n,2n) and (n,3n) cross section data for many nuclides, especially for ^{237}Np, ^{239}Pu and ^{243}Am. Draper and Parish [11] have undertaken a more detailed evaluation of a CTR device for transmutation of high-level wastes. With an array of computer programs, they plan to study plasma conditions, blanket designs, heat removal systems and waste configurations that will optimize waste disposal.

Quite recently, Rose, Olhoeft, Kellman, et al [12] have suggested that fission product and actinide waste burning can be accomplished with high energy neutrons from a device similar to the proposed Tokamak Fusion Test Reactor at Princeton which is a device of intermediate size wherein the conditions for D-T burning are achieved by the injection of high energy neutral beams into a relatively cool plasma. Further neutron multiplication is envisaged through the use of a subcritical assembly of fissile fuel in the blanket walls -- a fusion-driven fission reactor. The blankets considered will include fast fission liquid cooled blanket, beryllium-graphite moderated helium cooled thermal blanket, molten salt blanket and aqueous homogeneous blanket.

CROSS-SECTION NEEDS AND MEASUREMENTS

The quantitative prediction of the various nuclei produced, transmuted and fissioned in reactors is necessary for systematic actinide management.

These quantitative predictions are made with special computer programs which require as input values all relevant cross sections of all the nuclides in the region of interest. At present, most codes utilize effective values at thermal, resonance and fast neutron energies. These are obtained from suitable weighted averages over the reactor spectrum in question, and require detailed data. Benjamin [13] has surveyed neutron cross-section measurements of the actinides with emphasis on those desired for an LWR for ^{252}Cf and ^{238}Pu production. In many isotopic cases, an adjusted set of cross sections has been used because of the urgent need to perform the calculations in spite of gaps in our knowledge (see Table II) of these cross sections. Such an "adjusted set" of cross sections has limited applicability. Therefore, it will be necessary to make neutron cross-section measurements to obtain better predictions. There are certain obvious deficiencies in the data which can be attacked immediately. There is a total lack of differential fission cross section data below 15-eV neutron energy from underground nuclear explosions on several fissile nuclides of Cm and Cf. The deficiencies in capture cross sections are numerous especially over the fast-reactor spectrum region. In the determination of fission and capture widths in the resolved resonance region, a knowledge of total cross sections over the same neutron energy region can supplement fission and capture measurements. For the fast reactor calculations, some determinations of (n,2n), (n,3n) and (n,charged particle) cross sections will likely be needed. Pulsed neutron sources and the techniques for the measurement of many of the needed cross sections are in existence. Underground nuclear explosions [14] have already provided a wealth of fission cross-section measurements on the actinides. We discuss briefly some measurement programs on the actinides currently proposed or underway in the United States, spurred mainly by the urgency of the actinide waste problem.

INTEGRAL MEASUREMENTS

Recycling studies in the LMFBR are seriously handicapped by the non-availability of differential cross section data. In the interim, the Argonne group composed of Fields, Fried, Friedman and Unik [15] propose that gross burnout measurements made at selected locations in the Idaho Experimental Breeder Reactor-II (EBR-II) will provide integral data which can be employed for feasibility studies. In fact, forty samples of separated nuclides ranging in mass from ^{232}Th to ^{241}Pu have been already irradiated for four years in EBR-II receiving an integrated dose of 10^{23} fast neutrons. These samples have been cooling for over four years. The Argonne group intends to carry out mass spectrometric and radiometric analyses of the purified actinide and selected fission product fractions. It is planned that addi-

TABLE II[†]

Actinide Cross Sections in Barns

(Uncertainties given as standard deviation)

	$\sigma_{n\gamma}^{2200}$	σ_{nf}^{2200}	Resonance Integrals $I_{n\gamma}$	Resonance Integrals I_{nf}
92-U-236	5.3 ± 0.2	a	358 ± 20	a
92-U-237	478 ± 160	a	b	a
93-Np-237	172 ± 3	a	819[c]	a
93-Np-238	b	2200 ± 200	b	1500 ± 500
94-Pu-238	559 ± 20	17.3 ± 0.4	164 ± 15	25 ± 5
95-Am-241	832 ± 21	3.14 ± 0.10	1538 ± 118	21 ± 2
95-Am-242	b	2100 ± 200	b	< 300
95-Am-243	77 ± 3	a	1813 ± 70	b
96-Cm-242	b	5	b	150 ± 40
96-Cm-243	b	960 ± 50	b	1860 ± 400
96-Cm-244	14 ± 4	1.1 ± 0.5	606 ± 23	18 ± 1
96-Cm-245	342 ± 20	2018 ± 37	102 ± 7	772 ± 30
96-Cm-246	1.3 ± 0.5	0.17 ± 0.10	121 ± 7	10 ± 0.4
96-Cm-247	60 ± 30	82 ± 5	800 ± 400	778 ± 50
96-Cm-248	4 ± 2	0.34 ± 0.07	300 ± 75	13.2 ± 0.8

[†]From Reference 13
a - unknown, but probably quite small
b - unknown, but probably quite appreciable
c - probably too high

The cross sections for Bk and Cf are either unknown or very poorly known.

tional samples of ^{242}Pu, ^{241}Am and ^{243}Am will be inserted in EBR-II for irradiation. A computer model will be developed to calculate the yields of various actinides and fission products which will be compared with measured yields.

The Los Alamos Radiochemistry Group [16] has also made several integral cross section measurements in critical assemblies utilizing activation and fission chamber techniques. The samples studied include ^{235}U, ^{236}U, ^{237}U, ^{238}U, ^{237}Np, ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{241}Am. The data have not been fully analyzed but with sufficient impetus and interest generated

by the actinide recycle concept, the cross section data should become available.

MEASUREMENTS WITH ELECTRON LINEAR ACCELERATORS

The electron linear accelerators provide a versatile pulsed neutron source for detailed differential cross section measurements. Sample acquisition, purification and preparation constitute a major part of any cross section measurement program. The minimum quantity of separated isotope needed for cross section measurements is about 50 μg, 10 mg and 100 mg for fission, total and capture cross sections, respectively. The availability of samples is discussed at length in another review paper[1].

The ratios of the fission cross sections of ^{234}U and ^{236}U relative to the ^{235}U fission cross section have recently been measured [17] at the Lawrence Livermore Laboratory (LLL) Linear Accelerator in the neutron energy range 0.1 to 30 MeV. Typical energy resolution was 5% at 20 MeV and 1.5% at 1 MeV. Counting uncertainties were less than 4%; overall systematic uncertainties were estimated to be less than 5%. These data were presented recently [18]. Future plans include the measurement of the fission cross section ratios of ^{238}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu and ^{244}Pu relative to ^{235}U over this same energy range.

Preliminary results for the fission cross section of ^{245}Cm have also been obtained at the LLL linac in the neutron energy range 0.006 eV to 20 eV [17]. Similar measurements covering the neutron energy range from 0.01 eV to 14 MeV are planned with ^{242m}Am, ^{243}Cm, ^{245}Cm, ^{247}Cm and ^{249}Cf. The LLL facility also possesses the capability of measuring $\bar{\nu}$, the number of prompt fission neutrons, as a function of incident neutron energy from 0.01 eV to 14 MeV. Future plans for these measurements will be based on need and interest for various actinides.

A detailed program for the measurement of actinide cross sections has been recently formulated at the Oak Ridge Electron Linear Accelerator (ORELA). The initial set of nuclides to be investigated is given in Table III. Preliminary results for the fission cross section of ^{245}Cm and total cross section of ^{249}Bk are discussed in another review paper [1]. Capture studies on ^{240}Pu, ^{241}Pu, and ^{241}Am are currently being carried out at ORELA [19]. Some short-lived isotopes have enough gamma ray and spontaneous fission activity to preclude normal differential capture cross section measurements. Some of these measurements may possibly be done with underground nuclear explosions [14]. Total cross section measurements can give good estimates of the capture cross section at low energies in the well-resolved region if appreciable fission is not present. Such measurements also are needed to give accurate resonance fission widths. Total cross section measure-

TABLE III

Proposed Five-Year Program at the Oak Ridge National Laboratory for Carrying out Needed Cross-Section Measurements in the Actinide Region

Isotope	Type	Proposed Energy Range	Measurement Techniques Required	Sample Availability
^{237}Np	σ_c	Thermal - 0.5 MeV	Current	Available
^{242m}Am	σ_c	Thermal - 0.5 MeV	Current	Very difficult[a]
^{242m}Am	σ_f	Thermal - 1.5 MeV	Current	Available
^{243}Am	σ_c	Thermal - 0.5 MeV	Current	Available
^{243}Am	σ_f	0.3 eV - 1.5 MeV	Some development required	Available[b]
^{243}Cm	σ_f	0.3 eV - 1.5 MeV	Some development required	Available[b]
^{245}Cm	σ_c	Thermal - 0.5 MeV	Current	Very difficult[c]
^{245}Cm	σ_f	0.3 eV - 1.5 MeV	In progress	12 μg available
^{247}Cm	σ_c	Thermal - 0.5 MeV	Current	Extremely difficult[d]
^{247}Cm	σ_f	0.3 eV - 1.5 MeV	Current	Very difficult[e]
^{251}Cf	σ_f	0.3 eV - 1.5 MeV	Current	Very difficult[f]

a. Irradiation of feed material and calutron separation necessary.
b. Feed material available. TRL isotopic separation required.
c. Calutron separation necessary. Available facilities require additional shielding and containment.
d. Preparation and irradiation of feed material very difficult. Calutron separation necessary. Available facilities require additional shielding and containment.
e. Feed material available. Difficult TRL isotopic separation required.
f. Small quantity of feed material available. Very difficult TRL separation required.

(TRL - Transuranium Laboratory (ORNL))

ments with samples as small as 10 mg have been carried out at ORELA. It should be noted that total cross section measurements are not practical with underground nuclear explosions.

(n,f) CROSS SECTIONS FOR EXOTIC ACTINIDES

It is highly desirable to have reliable (n,f) cross sections for as many actinides as possible. If for various reasons direct neutron measurements become impractical, Wilhelmy, Britt, Gavron, Konecny and Weber [20] have proposed an indirect method of extracting equivalent (n,f) cross sections from experimentally measured charged-particle induced fission probabilities (P_f) via the expression

$$\sigma_{n,f}(E_n) = P_f(E_n+B_n) \times \sigma_{CN}(E_n)$$

where σ_{CN} stands for the total compound-nuclear neutron cross section calculated with the optical model and E_n and B_n represent the incident neutron energy and the neutron binding energy, respectively. The assumption made is that the fission probabilities are independent of the reaction mechanism which the authors have verified to within ≈10% by producing the same compound nucleus with different reactions on appropriate targets. The charged-particle reactions employed were the (^{3}He,df) and (^{3}He,tf) reactions with future plans for ^{6}Li and ^{7}Li induced reactions. Wilhelmy et al have carried out DWBA calculations which show that the angular momentum transfer in the (^{3}He,d) reaction is quite similar to that expected for incident neutron energies in the few MeV range. However, the charged-particle simulation technique is not expected to be reliable for neutron energies below 2 MeV. The compound nuclei produced with the (^{3}He,d) and (^{3}He,t) reactions and the maximum equivalent neutron bombarding energy studied are given in Table IV. These measurements would be equivalent to carrying out (n,f) measurements on targets listed in the "Neutron Target" column. Future plans for these types of measurements being carried out at the Los Alamos Scientific Laboratory would include ≈50 isotopes at excitation energies ranging up to ≈24 MeV.

The preceding four sections show that a substantial start has been made in developing programs for neutron cross section measurements in the actinide region. The individual measurements would be guided by sensitivity studies which have also begun. Many important decisions regarding waste utilization, management and storage of waste depend on the knowledge to be acquired through these measurement programs.

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TABLE IV

Charged-Particle Induced Fission Probability Measurements Carried Out at Los Alamos Scientific Laboratory to Simulate (n,f) Measurements[†]

Neutron Target	$t_{1/2}$	Max Energy Neutron (MeV)	Compound Nucleus
^{229}Pa	1.4 d	5.2	^{230}Pa
^{230}Pa	17.4 d	3.7	^{231}Pa
^{231}Pa	3.25×10^4 y	6.3	^{232}Pa
^{232}Pa	1.32 d	4.4	^{233}Pa
^{230}U	20.8 d	7.0	^{231}U
^{231}U	4.2 d	5.2	^{232}U
^{232}Np	14.7 m	3.0	^{233}Np
^{233}Np	35 m	3.9	^{234}Np
^{234}Np	4.4 d	6.2	^{235}Np
^{235}Np	396 d	6.6	^{236}Np
^{236}Np	1.3×10^6 y	4.3	^{237}Np
^{237}Np	2.1×10^6 y	7.7	^{238}Np
^{238}Np	2.12 d	5.2	^{239}Np
^{236}Pu	2.85 y	7.2	^{237}Pu
^{237}Pu	45.6 d	5.6	^{238}Pu
^{238}Am	1.63 h	5.1	^{239}Am
^{239}Am	11.9 h	5.5	^{240}Am
^{240}Am	51 h	3.9	^{241}Am
^{241}Am	433 y	6.8	^{242}Am
^{242}Am	152 y	4.6	^{243}Am
^{240}Cm	26.8 d	6.2	^{241}Cm
^{241}Cm	36 d	4.6	^{242}Cm
^{242}Cm	163 d	7.1	^{243}Cm
^{243}Cm	28 y	5.3	^{244}Cm
^{247}Bk	1.4×10^3 y	5.9	^{248}Bk
^{248}Bk	18 h	4.2	^{249}Bk

[†]Reference 20

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