LA-UR-96-3690

CONF-9610228--2

Title: **NEUTRON-BASED MEASUREMENTS FOR NONDESTRUCTIVE ASSAY OF MINOR ACTINIDES PRODUCED IN NUCLEAR POWER REACTORS**

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Submitted to: **Alternate Nuclear Material Conference (ANM)
Aiken, SC USA
October 15-17, 1996
(FULL PAPER)**

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NEUTRON-BASED MEASUREMENTS FOR NONDESTRUCTIVE ASSAY OF MINOR ACTINIDES PRODUCED IN NUCLEAR POWER REACTORS

by

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ABSTRACT

Because of their impacts on long-term storage of high-level radioactive waste and their value as nuclear fuels, measurement and accounting of the minor actinides produced in nuclear power reactors are becoming significant issues. This paper briefly reviews the commercial nuclear fuel cycle with emphasis on reprocessing plants and key measurement points therein. Neutron signatures and characteristics are compared and contrasted for special nuclear materials (SNMs) and minor actinides (MAs). The paper focuses on the application of neutron-based nondestructive analysis (NDA) methods that can be extended for verification of MAs. We describe current IAEA methods for NDA of SNMs and extension of these methods to satisfy accounting requirements for MAs in reprocessing plant dissolver solutions, separated products, and high-level waste. Recommendations for further systems studies and development of measurement methods are also included.

I. INTRODUCTION

Minor actinides (MAs) produced in nuclear power reactor fuel have received considerable attention because of (1) their potential energy production value in recycle fuels for thermal and breeder reactors, and (2) because of their negative environmental impact on the long term storage of spent fuels.[1] While the plutonium in spent light-water reactor (LWR) fuel amounts to about 1% of the total heavy metal, the sum of the neptunium and americium is in the range 13-16% of the plutonium produced. Worldwide annual production of neptunium is about 3.5 tonnes and that of americium is comparable. There are approximately 50 g of curium per tonne of spent fuel, or 0.5% of the plutonium.[2] Most MAs reside in spent fuels located in temporary storage.

In the past, when spent fuel was reprocessed, the primary goal was to recover the plutonium and uranium for reuse in power reactor fuel cycles. The value of the recovered fuel, as well as nuclear safeguards, provided ample motivation for the careful measurement of the quantities of plutonium and uranium recovered. The minor actinides such as neptunium, americium, and curium were usually passed through the process with the fission product waste streams. Consequently, measurement of these materials was unimportant except as needed for process control. The copious spontaneous-fission neutron emission of curium has proven useful for spent fuel verification measurements. On a very small scale, some neptunium was separated for use as a target material to produce ^{238}Pu, which is useful as a radioisotopic heat source. MAs occasionally cause interferences in measurements of uranium and plutonium.

Today, some countries reprocess spent fuel as a matter of national energy security while others, such as the United States, have decided not to reprocess because of the lack of economic incentive to recycle plutonium and because of the potential nuclear proliferation problems associated with separated plutonium. Japan and France are currently reprocessing and planning to extract and burn minor actinides, as well as

plutonium, in advanced reactor fuel cycles. MAs are also extracted at reprocessing plants in the Russian Federation.

A very strong motivation for the separation and burning of the minor actinides is to transform them into less hazardous, shorter-lived fission products, thereby greatly reducing the cost and complexity of long-term storage of spent fuel wastes. By far the heaviest environmental burden of a nuclear waste repository is the neptunium (^{237}Np), which has a half life of 2.14 million years. The predominant isotope of americium, ^{241}Am, decays to ^{237}Np. Fission products without plutonium, neptunium, and americium pose a negligible environmental hazard after about 300 years.

The Power Reactor and Nuclear Fuel Development Corporation, Japan, is evaluating several options for the separation and utilization of the minor actinides.[3] One of these options is to pass these actinides through processing with the plutonium and without separation. Another option would be to accumulate the separated minor actinide nitrate solution and mix it with plutonium nitrate to be used exclusively in fast reactor fuels. The Japanese have also proposed using the Np/Am/Cm/Pu mix (up to 10% actinides) to produce fuel that is proliferation-resistant or self-protecting. The European community has also experimented with the production of fuels containing minor actinides, and the Russians are experimenting with the burning of MAs in fast reactors.

In his recent paper, R. J. S. Harry mentioned actinide waste in the context of nuclear nonproliferation.[4] Referring to strong neutron sources other than nuclear reactors that have been proposed for nuclear transmutation to burn actinide waste, Harry states that "these sources can be used to irradiate fertile material (thorium or uranium) or actinide waste to create fissile nuclides, which are not covered by the definition of a special fissionable material in the IAEA Statute (e.g., ^{237}Np and some isotopes of Cm and Cf)." If this avenue for the minor actinides becomes a reality, new accounting measures for these materials might be needed.

From this perspective, it appears that the measurement and accounting of the more abundant minor actinides could become important in the international community, as well as in the relevant advanced fuel operations. There will be a need to measure these actinides as separate products in solid or solution blends with plutonium and uranium and in process waste. The effects of these materials when mixed with plutonium, on the accuracy of the measurement of the plutonium should also be determined for both safeguards and process control.

The purpose of this paper is to identify neutron-based measurement techniques that may be used for measuring some of the MAs recovered from spent nuclear reactor fuel. Possible nondestructive techniques for use in a reprocessing facility are emphasized because they generally involve less expense than conventional chemical analyses.

II. THE POWER REACTOR FUEL CYCLE

Introduction of measurement and accounting of the minor actinides would impose some additional costs primarily at reprocessing plants and fuel fabrication facilities producing mixes of these materials with mixed, uranium-plutonium (MOX). Accounting for actinides at reactor facilities would already be covered by the same procedures used for safeguards accounting of spent fuel contents, namely, item accounting of the spent fuel assemblies coupled with burnup code calculations of isotope production and limited, nondestructive, verification measurements of burnup and cooling time.

The quantitative performance criteria for systems of measurements and accounting of the minor actinides will be strongly influenced by their reactor fuel worth and processing costs, and the cost of their permanent storage if they are not recovered. Because none of the MAs of interest here, i.e., neptunium, americium, and curium, are currently included in the special nuclear materials category of safeguards, they have no assigned "significant quantity," which is a value used for safeguards criteria. Consequently, without considerably more information on the separation and use of these materials in the fuel cycle, it is premature to attempt to establish performance criteria for the accounting of these materials. However, as an expedient for this study, we borrow from the language and structure used in existing safeguards systems used for the control and accounting of plutonium and uranium in the fuel cycle. Moreover, as will be shown below, several of the techniques and instruments currently used to measure plutonium and uranium can be adapted to the accounting needs for the MAs.

Safeguards at reprocessing plants are currently based on measurements of plutonium and uranium in the input and product streams. In addition, measurements are made to ensure that large quantities of material are not removed (gross defects) from a facility in waste streams. The 1991-95 IAEA Safeguards Criteria call for measurements of in-process materials on a monthly basis to fulfill timeliness criteria. Whether analogous measurements for the minor actinides will be needed depends on their relative "values," which are yet to be established. Whatever choices are made, the arguments presented here will still apply. As in the case of reactor facilities, the accounting of spent fuel in the input section of a reprocessing facility is done by item identification and control. Increased containment and surveillance methods will probably be emphasized for future reprocessing plants. The first actual measurements of plutonium for input materials accountability are made on samples taken from the input accountability tank after the fuel is dissolved. Solid residues remaining in leached hulls (from zirconium-alloy-clad fuels) may be measured nondestructively to ensure that large quantities of plutonium are not lost via this route. A sample of each batch of dissolver solution is taken and the uranium and plutonium contents are analyzed accurately to establish the process input. The dissolved material then moves to the separations area of the reprocessing plant where the plutonium and uranium are separated from the fission products and each other. At this point, the MAs could also be separated from the fission products, or some or all could be allowed to pass through with the fission products. After separation, the amounts of the purified plutonium and uranium products, either nitrate solutions or oxide powders, are then determined by destructive (DA) and/or nondestructive analysis (NDA) techniques. With the product measurements, the materials balance can then be calculated. Waste streams are also monitored to ensure that large quantities of plutonium are not removed.

In considering how standard reprocessing plant safeguards should be modified to allow for accounting of the minor actinides, two issues are important. The first is a reasonable choice for a unit of "material value" for the MAs. Because values have not yet been established for MAs, we assume them to be at least as large as the IAEA significant quantity (SQ) of 25 kg for enriched uranium. The second point is that the quantities of MAs present in a facility are typically 10 to 20 times lower than the quantity of plutonium. Thus, it is logical that the level of effort expended in accounting for these materials should be considerably less than that expended for plutonium accountability. Therefore, in general, it should not be necessary to make measurements of the MAs to accuracies and detection limits better than those for plutonium or on streams that are not now measured for plutonium unless some details of the process indicate a special need for their accountability. Hence, it is likely that no streams would have to be sampled in a reprocessing plant other than those already sampled for plutonium, except for actual MA product streams. There would obviously have to be extra analyses done to measure the MA contents, and this could require slightly larger samples. However, the important point in estimating resource requirements is that there would not be substantial investments of time to acquire samples from additional streams. The costs of MA accounting are not likely to add substantially to the current cost of materials accounting in a reprocessing plant. To make the additional analyses of the samples and the MA products as rapid and efficient as possible, development activities should be undertaken to improve the ability to measure the MA concentrations in relevant matrices. A detailed analysis of an operating reprocessing plant should be made to confirm the conclusions made above.

To provide perspective and to organize later information contained, we have considered a generic power-reactor fuel-cycle model. A simplified diagram of such a cycle is shown in Fig. 1. The model assumes a light-water reactor (LWR), a reprocessing plant, and an LWR MOX fuel fabrication plant. The MOX plant could provide fuel for an LWR or a fast-breeder reactor. One or many cycles are possible.

Figure 1 depicts material flows and key measurement points (KMPs) for the model LWR-MOX fuel cycle. Material flows are part of facility and process design information required for designing materials control and accountability (MC&A) systems for domestic and international safeguards. Material flows and facility design help determine key points where measurements are required to satisfy inspection criteria. These criteria stem from definitions of SQs and timeliness goals. According to IAEA 1991-95 Safeguards Criteria 7:9.1, 7:9.4, and Annex E, when 1 SQ or more of material is present at a facility, verifications of in-process inventory (IPI) for timely detection are carried out 12 times per year according to approved procedures. The purpose is to detect anomalies that could indicate abrupt diversion of 1 SQ during the period. Table I shows values presently used by the IAEA for special nuclear material (SNM). Similar values must

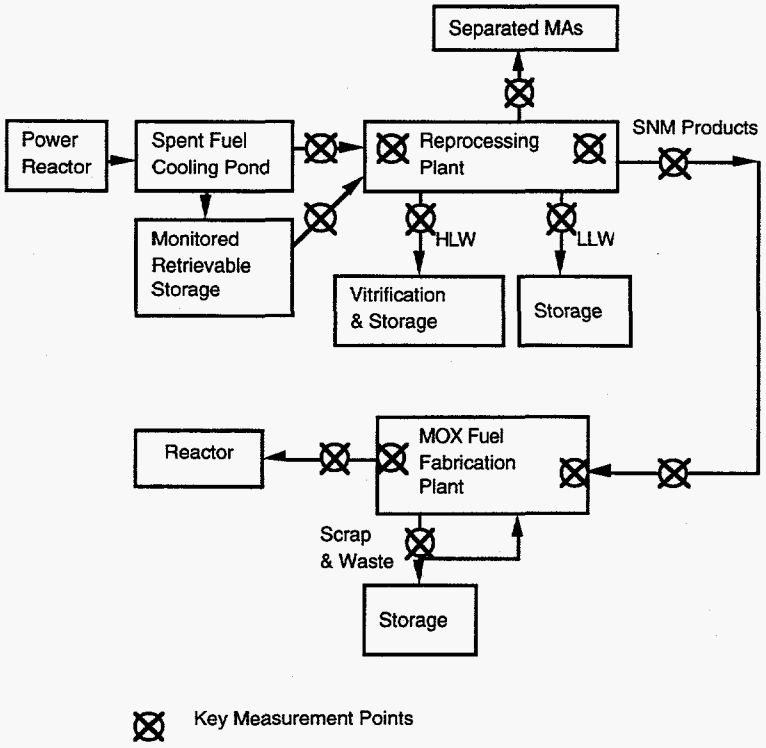

Fig. 1. Simplified model of LWR-MOX fuel cycle.

TABLE I. Special Nuclear Material (SNM) Significant Quantities and Timeliness Goals (IAEA)[5]

Material Category	Material Type	Significant Quantity (kg)	Timeliness Goal (months)
Direct Use Material	Plutonium[a] (Separated)	8	1
	High-Enriched Uranium (20% ^{235}U)	25	1 (unirradiated) 3 (irradiated)
	Plutonium in Spent Fuel	8	3
	^{233}U	8	1
Indirect Use Material	Low-Enriched[b] Uranium (20% ^{235}U)		12
	Thorium	20 t Th	12

[a]For plutonium containing less than 80% ^{238}Pu.

[b]Including natural and depleted uranium.

be established for MAs either for economic reasons or if they are ever placed under IAEA safeguards. Following establishment of such criteria for MAs, facility-specific measurement criteria would be defined via systems studies.

III. IAEA INSTRUMENTS IN ROUTINE USE

In anticipation of the possibility that some MAs will become subject to international safeguards, it is natural to first consider NDA methods currently in routine use by the IAEA at storage and reprocessing facilities. Some of these methods are described in Table II.

IV. SNM AND MA NEUTRON SIGNATURES

Table III lists neutron emission data for thorium, uranium, neptunium, plutonium, americium, curium, and californium isotopes.

Table III can be used to indicate the feasibility of passive neutron counting for quantitative assay of the isotopes listed, either in metal or oxide form.

Neutrons emitted from the spontaneous fission of ^{237}Np are too few to use as a passive assay signature. Active neutron interrogation is therefore required for pure metal. For bulk oxide, however, it may be possible to use a multiplication-corrected count of all the (α,n) neutrons from ^{237}Np as a signature. Alternatively, passive multiplicity counting may be used to assay the ^{240}Pu-effective, and infer the MA content using high-resolution gamma spectrometry. For ^{241}Am and ^{243}Am, the emission rates for spontaneous-fission neutrons are also low but probably sufficient for signatures of bulk metal items. For americium oxides, a multiplication-corrected counting of the total (α,n) neutrons looks promising as a signature. In practice, combinations of SNM and MAs could be encountered. These points are discussed in Sec. IV.B.2.

TABLE II. Routinely Used IAEA NDA Instruments

Measurement	Method	Instrument (acronym)
U, Pu radiation	•Low-resolution gamma spectrometry	•Portable MCA - NaI (PMCN)
^{235}U mass	•Active neutron coincidence counting	•Active Well Coincidence Counter (AWCC)
^{235}U enrichment	•Low-resolution gamma spectrometry	•Portable MCA - NaI (PMCN)
	•High-resolution gamma spectrometry	•Portable MCA - HPGe (PMCG)
^{240}Pu-effective mass	•Passive neutron coincidence counting	•High Level Neutron Coincidence Counter (HLNC)
		•Inventory Sample Counter (INVS)
	•Passive neutron multiplicity counting	•Plutonium Scrap Multiplicity Counter (PSMC)
^{240}Pu-effective fraction	•High-resolution gamma spectrometry	•Medium Count Rate System (MCRS)

TABLE III. Spontaneous Fission and (α,n) Neutron Yields of Selected Isotopes

Isotope	Total Half Life[a] (yr)	Spontaneous Fission (SF)[b] SF Half-Life (yr)	Neutrons per SF	Neutron Yield (n/g-s)	(α,n) Reaction in Oxide[b] α-decay Half-Life (yr)	Neutron Yield (n/g-s)
^{232}Th	1.41×10^{10}	$>1 \times 10^{21}$	2.14	$>6 \times 10^{-5}$	1.41×10^{10}	2.2×10^{-5}
^{232}U	71.7	8×10^{13}	1.71	1.3	71.7	1.49×10^{4}
^{233}U	1.59×10^{5}	1.2×10^{17}	1.76	8.6×10^{-4}	1.59×10^{5}	4.8
^{234}U	2.45×10^{5}	2.1×10^{16}	1.81	5.02×10^{-3}	2.45×10^{5}	3.0
^{235}U	7.04×10^{8}	3.5×10^{17}	1.86	2.99×10^{-4}	7.04×10^{8}	7.1×10^{-4}
^{236}U	2.34×10^{7}	1.95×10^{16}	1.91	5.49×10^{-3}	2.34×10^{7}	2.4×10^{-2}
^{238}U	4.47×10^{9}	8.2×10^{15}	2.01	1.36×10^{-2}	4.47×10^{9}	8.3×10^{-5}
^{237}Np	2.14×10^{6}	1.0×10^{18}	2.05	1.14×10^{-4}	2.14×10^{6}	0.34
^{238}Pu	87.74	4.77×10^{10}	2.22	2.59×10^{3}	87.74	1.34×10^{4}
^{239}Pu	2.41×10^{4}	5.48×10^{15}	2.16	2.18×10^{-2}	2.41×10^{4}	38.1
^{240}Pu	6.56×10^{3}	1.16×10^{11}	2.16	1.02×10^{3}	6.56×10^{3}	1.41×10^{2}
^{241}Pu	14.35	(2.5×10^{15})	2.25	(4.94×10^{-2})	5.90×10^{5}	1.3
^{242}Pu	3.76×10^{5}	6.84×10^{10}	2.15	1.72×10^{3}	3.76×10^{5}	2.0
^{241}Am	433.6	1.05×10^{14}	2.27	1.18	433.6	2.69×10^{3}
^{242m}Am	152	9.5×10^{11}	2.34	1.35×10^{2}	152	33.1
^{243}Am	7.38×10^{3}	3.35×10^{13}	2.42	3.93	7.38×10^{3}	1.34×10^{2}
^{240}Cm	26.8 days	1.9×10^{6}	2.39	6.93×10^{7}	26.8 days	2.53×10^{7}
^{241}Cm	32.4 days	(1.6×10^{12})	(2.50)	(8.57×10^{1})	32.4 days	1.72×10^{5}
^{242}Cm	163 days	6.56×10^{6}	2.52	2.1×10^{7}	163 days	3.76×10^{6}
^{243}Cm	28.5	(1.2×10^{11})	(2.69)	(1.22×10^{3})	28.5	5.00×10^{4}
^{244}Cm	18.1	1.35×10^{7}	2.69	1.08×10^{7}	18.1	7.73×10^{4}
^{245}Cm	8.48×10^{3}	(4.0×10^{12})	(2.87)	(3.87×10^{1})	8.48×10^{3}	1.24×10^{2}
^{246}Cm	4.73×10^{3}	1.81×10^{7}	3.18	9.45×10^{6}	4.73×10^{3}	2.24×10^{2}
^{252}Cf	2.646	85.5	3.757	2.34×10^{12}	2.731	6.0×10^{5}

[a]Ref. 6

[b]Ref. 7. Values in ( ) are from Ref. 8, from which half-lives and yields have estimated accuracies of 2 orders of magnitude. ^{240}Pu spontaneous fission rate is taken from Ref. 9.

Table IV lists cross-sections (probabilities) for the induced fission ($\sigma_{fission}$) and radiative capture ($\sigma_{n,\gamma}$) reactions and for all neutron-isotope reactions (σ_{total}). Values are cited for thermal neutron energies, 1 MeV, and 14 MeV. These data indicate the feasibility of active neutron interrogation of the isotopes listed. Monte Carlo simulations using the entire energy range (0-20 MeV) of neutron cross-sections will be useful in evaluating feasibility.

For ^{237}Np and ^{243}Am, only fast neutrons generate an induced-fission signature. Thermal neutrons could be used for a neutron-capture gamma-ray assay. Americium-241 has a small thermal-neutron fission cross-section, perhaps enabling an induced-fission signature.

Figure 2 is a plot of neutron-induced fission cross-sections for ^{235}U, ^{238}U, ^{237}Np, and ^{239}Pu. The figure shows that above 0.7 MeV, the fission cross-section of ^{237}Np exceeds those of ^{235}U and ^{238}U, and above 7 MeV, the ^{237}Np cross-section is comparable to that of ^{239}Pu.

TABLE IV. Neutron Cross-Sections for Selected Isotopes[10]

Isotope	Thermal			1 MeV			14 MeV		
	σ_{total} (barns)	$\sigma_{fission}$ (barns)	$\sigma_{n,\gamma}$ (barns)	σ_{total} (barns)	$\sigma_{fission}$ (barns)	$\sigma_{n,\gamma}$ (barns)	σ_{total} (barns)	$\sigma_{fission}$ (barns)	$\sigma_{n,\gamma}$ (barns)
^{232}Th	20.4	-	7.4	7.0	-	0.13	5.6	0.3	-
^{233}U	587.1	529.0	45.8	6.8	1.9	0.1	5.8	2.3	-
^{234}U	116.1	0.5	103.3	8.0	1.1	0.4	5.5	2.1	-
^{235}U	697.1	584.1	98.3	6.8	1.2	0.1	5.8	2.1	-
^{236}U	13.3	0.1	5.2	7.7	0.4	0.4	5.7	1.6	-
^{238}U	11.6	-	2.7	7.1	-	0.1	5.9	1.1	-
^{237}Np	196.0	-	181.3	6.8	1.5	0.2	5.6	2.3	-
^{238}Pu	599.3	16.8	562.2	6.7	2.1	0.2	7.1	2.7	-
^{239}Pu	1020.8	742.1	270.5	7.1	1.7	-	6.0	2.5	-
^{240}Pu	292.1	-	290.3	7.2	1.5	0.1	6.1	2.3	-
^{241}Pu	1389.9	1017.4	361.4	8.0	1.6	0.1	5.4	2.2	-
^{242}Pu	27.0	-	19.3	7.3	1.4	0.1	6.0	2.0	-
^{241}Am	592.0	3.6	578.4	7.1	1.4	0.3	6.0	2.7	-
^{242m}Am	7985.8	6636.2	1341.6	6.0	2.4	-	6.2	2.6	-
^{243}Am	82.0	-	75.0	7.3	1.2	0.1	5.8	2.5	-
^{242}Cm	30.8	3.0	17.2	6.8	0.5	-	6.0	2.6	-
^{243}Cm	1093.2	690.9	391.4	8.5	2.0	-	5.7	2.2	-
^{244}Cm	18.0	0.6	10.4	7.1	2.0	0.1	6.7	3.2	-
^{245}Cm	2431.6	2020.6	391.5	8.1	1.5	-	6.1	2.6	-
^{246}Cm	12.7	0.2	1.2	8.3	1.6	-	5.6	2.1	-

Table IV lists cross-sections (probabilities) for the induced fission ($\sigma_{fission}$) and radiative capture ($\sigma_{n,\gamma}$) reactions and for all neutron-isotope reactions (σ_{total}). Values are cited for thermal neutron energies, 1 MeV, and 14 MeV. These data indicate the feasibility of active neutron interrogation of the isotopes listed. Monte Carlo simulations using the entire energy range (0-20 MeV) of neutron cross-sections will be useful in evaluating feasibility.

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V. REPROCESSING PLANT DISSOLVER SOLUTION

Referring to Fig. 1, calibrated accountability tanks are required for volume measurements at the KMPs indicated, or where significant flows of SNM or MA or both are found. For concentrations, both chemical and NDA methods are used, some of which require development for MAs. Applicability of existing NDA methods depends quite strongly on concentrations of MAs, fission products, uranium, and matrix in the various solution streams.

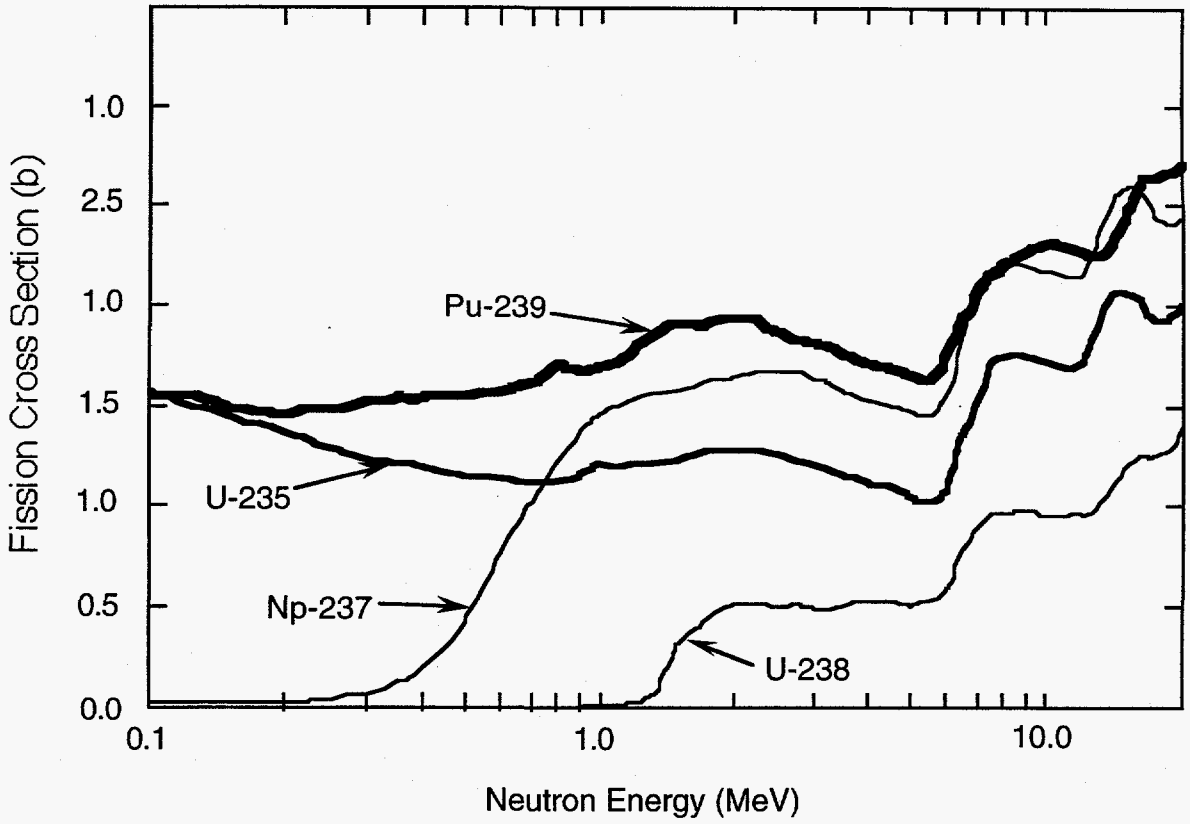

Fig. 2. Fast-neutron-induced fission cross-sections for ^{239}Pu, ^{235}U, ^{237}Np, and ^{238}U.

References 2 and 11 show that for exposures greater than 10 GWd/tU, the curium isotopes are the dominant neutron producers in spent fuel. For burnups greater than 25 GWd/tU and after approximately 2 year's cooling time, ^{242}Cm neutron production decays to insignificance, compared to ^{244}Cm. For lower burnups, the relative ^{242}Cm concentration is greater. Because of the very high rate of spontaneous-fission neutron emission (see Table III), the absolute concentration of ^{244}Cm can be determined by counting the neutrons from a small sample of known volume from the dissolver solution. The inventory sample neutron coincidence counter (INVS),[12] appropriately shielded and calibrated, is ideally suited for this measurement. The ratios of the concentrations of ^{244}Cm to elements important for safeguards accounting, i.e., plutonium, neptunium, and americium, can then be used with absolute neutron counting to determine the content of these elements in some of the materials downstream from the headend dissolver tank. The plutonium, neptunium, and americium concentrations in the dissolver solution sample, which can be used to form "tagging" ratios, would be determined by the Hybrid XRF K-edge Densitometer. A similar approach has been inferred in reference 13.

This technique can be applied only if chemical processes do not change the tag ratio, i.e., there is no partitioning. There may be at least two such cases. The first is in determining the levels of plutonium, neptunium, and americium in leached spent-fuel hulls, where fractionation of the suite of transuranics is expected to be small. The second potential application is to all process and waste streams in which the Cm/Am ratio has not changed during chemical partitioning, e.g., high-level liquid waste containing fission products, americium, and curium. In this case the objective could be to draw a curium balance. The feasibility of this method can be determined only if details of facility-specific processes are known. The "curium balance method" appears to possess significant promise for verifying SNM and MA in waste streams.

Determination of the feasibility of the Cm/MA ratio method using INVS requires development and in-field experiments.

VI. REPROCESSING PLANT SEPARATED MA AND SNM PRODUCTS AND RESIDUES

For separated, pure plutonium oxide, the standard NDA methods are neutron coincidence counting and high-resolution gamma-ray spectroscopy (HRGS). The High-Level Neutron Coincidence Counter[14] and related instruments are used routinely by international and domestic inspectors to verify the effective ^{240}Pu mass in plutonium oxide items. HRGS systems using FRAM[15] or MGA[16] analysis routines are used to determine plutonium and americium isotopic ratios. Combining the neutron and gamma-ray methods yields total plutonium. A modification to MGA has been developed that contains an analysis of ^{237}Np/Pu.[17] This algorithm assumes that protactinium and americium are both removed during chemical processing. With this assumption, the code calculates the state of equilibrium based on the observed in-growth of ^{241}Am and makes necessary corrections to the ^{237}Np - ^{233}Pa equilibrium. Detection sensitivity for ^{237}Np is quoted at 50 parts per million of plutonium.

For separated, pure neptunium oxide, total neutron counting could be used to directly determine the ^{237}Np mass from (α,n) neutron emission. However, it is possible that plutonium would be present at levels of 0.1 to 1.0%. This being the case, neutron multiplicity counting[18] could be employed to assay the effective ^{240}Pu mass from the emission of spontaneous-fission neutrons. This application would be similar to that of impure plutonium oxide and residues. HRGS methods could be used to determine the ^{237}Np/Pu ratio. Development would be required to extend existing spectroscopy physics methods to this new case, i.e., ^{237}Np/Pu ratios of 100/1 or 1000/1.

Table V shows the neutron source production for 1 kg of ^{237}Np and Pu as oxide, for three Np/Pu ratios: 1000, 100, and 20. In all cases, the spontaneous fission production from ^{237}Np is inconsequential. The (α,n) production is dominated by ^{237}Np only for the first case (0.1% Pu). For Pu concentrations of 1% and 5%, the (α,n) production is dominated by Pu.

These cases and others were simulated using a "figure-of-merit" code that gives ^{240}Pu-effective mass measurement precision based on detector and sample parameters. Results of application of this code to PSMC measurements of 1 kg of ^{237}Np and Pu as oxide, for a wide range of Np/Pu ratios, are shown in Figure 3.

From Figure 2, it is seen that passive neutron multiplicity assay precisions for ^{240}Pu-effective mass of 1% or less are achievable for Np/Pu ratios between ~3 and ~1000.

While implementation of passive neutron techniques is preferable to active methods from the standpoints of cost, complexity, and reliability, active methods may have to be applied, e.g., the case of pure ^{237}Np. Table VI compares candidate methods for active, high-energy-neutron interrogation.

For separated uranium oxide, a combination of neutron multiplicity counting and HRGS could yield SNM and MA masses. Depending on concentrations, active neutron interrogation could be required, possibly including isotopic neutron sources emitting fast and intermediate neutrons. Development is required to determine feasibility.

Extending an HRGS system to MAs would require new analysis algorithms to cover a wide range of ^{237}Np/Pu and U/Pu ratios.

Sampling and DA could also be used for measurements of homogeneous product oxides for a cost of ~$10K per sample. For international safeguards, this method is complementary to NDA methods.

The case of residues case is similar to that of neptunium oxide with trace amounts of plutonium. Neutron multiplicity counting and enhanced HRGS methods would be applicable for homogeneous and slightly inhomogeneous residues with neutron outputs not completely dominated by (α,n) emission. DA is applicable to homogeneous residues. For very high (α,n) neutron-emitting residues, calorimetry would also be applicable.

TABLE V. Neutron Production (n/s) for 1 kg of ^{237}Np with 0.1, 1, and 5% Pu								
Np/Pu	^{237}Np SF	Pu SF	Total SF	^{237}Np (α,n)	Pu (α,n)	Total (α,n)	α	Total
1000	0.1	353.4	353.5	339.7	159.2	498.9	1.41	852.4
100	0.1	3533.8	3533.9	336.6	1592	1928.6	0.55	5462.5
20	0.1	17668.9	17669	323	7959.9	8282.9	0.47	25951.9

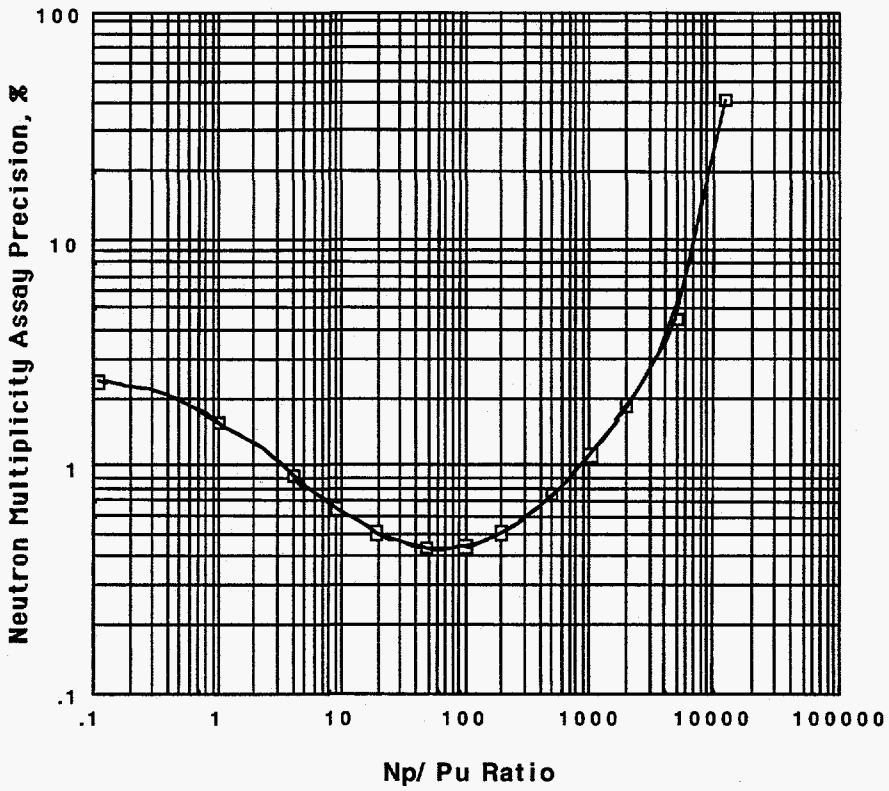

Fig. 3. Precision of 1000 s passive neutron multiplicity assay of 1 kg of ^{237}Np plus Pu (as Oxide) versus Np/Pu ratio.

TABLE VI. Comparison of Active Neutron NDA Methods for Separated Np Oxide					
Type of Source	Half-Life	Availability of Intense Sources	γ-Ray Back-ground	Neutron Spectrum	Cost
(α,n) on Be, B or F	long (+)	$<5x10^7$ - 10^8 (+)	low, 4.43 MeV in Be (α,n) (+)	hard ~5 MeV (+)	Reasonable for low and medium intensity (+) high for high intensities (-)
(α,n) on Li	long (+)	$5x10^4$ - $5x10^5$ (+,-)	very low (+)	soft, ~0.3 MeV (-)	high (-)
^{252}Cf	relatively short (2.6 yr) (-,+)	$<10^{12}$ (+)	high, soft and medium (-,+)	fission spectrum ~2 MeV avg. (-,+)	lowest cost/ neutron for isotopic sources (+)
14 MeV neutron (D,T) generator	limited target life when full beam is used (-)	10^9 - 10^{12} (+)	low (+)	very hard 14 MeV (+)	lowest cost/ neutron, highest capital cost (-,+)

VII. RECOMMENDED NEXT STEPS

In the course of preparing this paper and examining the application of neutron NDA methods to quantitative assay of MAs, either alone as separated pure products, or mixed with U and Pu and/or fission products, several tasks have been identified as essential to further progress. These are identified below.

- Establishing Loss Detection Criteria for MAs

Significant quantities and detection times must be established for MAs before meaningful systems studies can be performed. These must be based on material attractiveness for diversion.

- Systems Studies of Operating or Planned Reprocessing Plants (small, large)

In addition to identification of the key measurement points, which are obvious in most cases, such studies can quantify the measurement accuracies, precisions, and throughputs required to meet or exceed loss detection criteria using NDA and DA methods.

- Development of Characteristic NDA Standards

Successful application and extension of neutron NDA methods for assay of MAs depends on development of standards characteristic of the process materials and items. This is particularly true for mixtures of SNMs and MAs. Neutron coincidence counting is quite sensitive to trace amounts of Pu and even more so, Cm.

- Development of Nuclear Data and Analysis Algorithms for Application of PSMC and HRGS to MAs

Spontaneous and induced fission neutron multiplicity data are required for some MAs. Analysis algorithms may require reformulation for application of neutron multiplicity counting to MAs. The required compilations of decay gamma-ray intensities must be added to current libraries supporting HRGS analysis routines. HRGS analysis algorithms must be modified to cover a wide dynamic range of MA and SNM concentrations. HRGS and/or DA isotopic ratio data are essential for proper interpretation of both neutron multiplicity counting and calorimetry data.

- Laboratory Measurements Using Existing NDA Instruments Adapted for MAs

Where possible, laboratory measurements should be made to calibrate NDA instruments and benchmark performance. As far as practicable, these measurements should be made on well-characterized materials.

- Further Simulations of NDA Instrument Performance for MAs

Simulations must be used to "fill in the gaps" made necessary by the unavailability of adequate quantities of well-characterized reference materials.

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