LA-13054-MS **C.3**

LA - 13054-MS
c. 3

Measurement and Accounting of the Minor Actinides Produced in Nuclear Power Reactors

Los Alamos
NATIONAL LABORATORY

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36.

Edited by Paul W. Henriksen, Group CIC-1
Prepared by Celina M. Ortiz, Group NIS-5

This work was supported by the U.S. Department of Energy, Office of Nonproliferation and National Security, Office of Safeguards and Security.

An Affirmative Action/Equal Opportunity Employer

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither The Regents of the University of California, the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by The Regents of the University of California, the United States Government, or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of The Regents of the University of California, the United States Government, or any agency thereof. The Los Alamos National Laboratory strongly supports academic freedom and a researcher's right to publish; therefore, the Laboratory as an institution does not endorse the viewpoint of a publication or guarantee its technical correctness.

LA-13054-MS

UC-700
Issued: January 1996

Measurement and Accounting of the Minor Actinides Produced in Nuclear Power Reactors

J. E. Stewart
R. B. Walton
J. R. Phillips
S.-T. Hsue
G. W. Eccleston
H. O. Menlove
J. M. Davidson
W. D. Stanbro

Los Alamos
NATIONAL LABORATORY
Los Alamos, New Mexico 87545

CONTENTS

MEASUREMENT AND ACCOUNTING FOR MINOR ACTINIDES PRODUCED IN NUCLEAR POWER REACTORS

by

J. E. Stewart, R. B. Walton, J. R. Phillips, S. -T. Hsue,
G. W. Eccleston, H. O. Menlove, J. M. Davidson, and W. D. Stanbro

ABSTRACT

Because of their value as nuclear fuels and their impact on long-term storage of high-level radioactive waste, measurement and accounting for minor actinides (MAs) produced in nuclear power reactors are becoming significant issues. This report attempts to put the issues in perspective by reviewing the commercial nuclear fuel cycle with emphasis on reprocessing plants and key measurement points therein. Radiation signatures and characteristics are compared and contrasted for special nuclear materials (SNMs) and MAs. Also, inventories and relative amounts of SNMs and MAs are generally described for irradiated nuclear fuel and reprocessing plants. The bulk of the report describes appropriate measurement technologies, capabilities, and development needs to satisfy material accounting requirements for MAs, with emphasis on adaptation of current technologies. Recommendations for future systems studies and development of measurement methods are also included.

I. INTRODUCTION

In this report, minor actinides (MAs) include neptunium, americium, and curium. The 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) 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 between 13 and 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 in temporary storage or in fuels undergoing irradiation.

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 MAs 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 ^{244}Cm 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 MAs, 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 MAs is to transform them into less hazardous, shorter-lived fission products or actinides, 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, alpha decays to ^{237}Np with a half-life of 434 years. Fission products without plutonium, neptunium, and americium pose a negligible environmental hazard after about 300 years.

The Power Reactor and Nuclear Fuel Development Corporation (PNC), Japan, is evaluating several options for the separation and utilization of the MAs.[3] One of these options is to pass them through processing with the plutonium without separation. Another option would be to accumulate the separated minor actinide nitrate solution and mix it with plutonium nitrate to be used exclusively for 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 MAs, and the Russians are experimenting with the burning of MAs in fast reactors.

In his recent paper, R.J.S. Harry discussed 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 MAs becomes a reality, new accounting measures for these materials might be needed.

From this perspective, it appears that the measurement and accounting for the more abundant MAs is becoming more important to 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 report is to suggest techniques that may be feasible 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. Partial estimates of the effort needed to adapt and test these techniques and the equipment costs are also included.

II. MEASUREMENT AND ACCOUNTING SYSTEMS

A. Overview

The introduction of measurement and accounting for the MAs would impose some additional costs primarily at reprocessing plants and fuel fabrication facilities producing mixes of these materials with 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 for the MAs 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 SNM 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 materials accounting. However, as an expedient for this study, we borrow from the language and structure of 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.

Material Control and Accountability (MC&A) at reprocessing plants is 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 monthly measurements of in-process materials to fulfill timeliness criteria. Whether analogous measurements for the MAs 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, accounting for 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 and/or nondestructive analysis 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 MAs, 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 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 minor actinide product streams. Clearly, extra analyses would have to be done to determine the minor actinide 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 minor actinide 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 minor actinide 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.

B. Fuel Cycles

To provide perspective and organize information contained in Sec. III, 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) power reactor, a reprocessing plant, and an LWR mixed uranium-plutonium (MOX) fuel fabrication plant. The MOX plant could provide fuel for an LWR or a fast breeder reactor (FBR). One or many cycles are possible.

C. Material Flows and Key Measurement Points

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 (SC) 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 SNM. Similar values must 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.

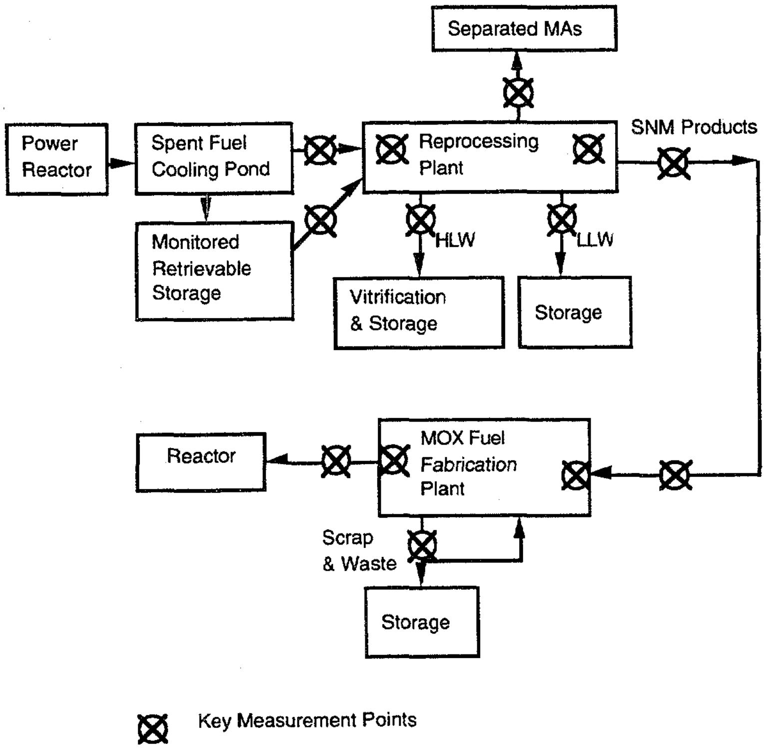

Fig. 1. Simplified model of LWR-MOX fuel cycle.

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	12

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

[b] Including natural and depleted uranium.

III. SPECIAL NUCLEAR MATERIAL (SNM) AND MINOR ACTINIDE (MA) RADIATION SIGNATURES AND CHARACTERISTICS

Before describing measurement technologies possible for the KMPs of Fig. 1, we summarize important nuclear and atomic parameters of SNM and other isotopes, several of which are MAs.

A. Gamma Rays

Table II lists the gamma rays most commonly used (signatures) for the nondestructive assay (NDA) of uranium and plutonium. The table also lists gamma-ray signatures for candidate MA isotopes.

Figure 2 shows part of the decay chain for ^{237}Np. Gamma rays from the daughter ^{233}Pa are much more abundant and therefore more easily detected than those from the direct alpha decay of ^{237}Np, which has a half-life of 2.14 million years. Because the half-life for alpha decay of ^{233}Pa is 27 days, ^{237}Np and ^{233}Pa reach 94% of equilibrium after 108 days.

For safeguards applications, gamma rays from ^{233}Pa are the most useful for NDA of ^{237}Np, provided equilibrium has been reached or the time since separation can be determined. The situation is comparable to that for ^{238}U, where gamma rays from the daughter ^{234m}Pa are used.

TABLE II. Gamma-Ray Signatures of Selected Isotopes

Isotope	Energy[a] (keV)	Activity[a] (γ/g-s)
^{234}U	120.9	9.35×10^4
^{235}U	143.8	8.4×10^3
	185.7	4.32×10^4
^{238}U	766.4[b]	2.57×10^1
	1001.0[b]	7.34×10^1
^{237}Np	86.486	3.29×10^6
	143.26	1.13×10^5
	300.12[c]	1.68×10^6
	311.98[c]	9.82×10^6
	340.5[c]	1.13×10^6
^{238}Pu	152.7	5.90×10^6
	766.4	1.387×10^5
^{239}Pu	129.3	1.436×10^5
	413.7	3.416×10^4
^{240}Pu	45.2	3.80×10^6
	160.3	3.37×10^4
	642.5	1.044×10^3
^{241}Pu	148.6	7.15×10^6
	208.0[d]	2.041×10^7
^{241}Am	59.5	4.54×10^{10}
	125.3	5.16×10^6
^{243}Am	43.6	6.42×10^8
	74.8	4.87×10^9
	106.1[e]	1.72×10^9
	209.7[e]	2.45×10^8
	228.1[e]	8.05×10^8
	277.6[e]	1.06×10^9
	315.8[e]	1.20×10^8
	334.3[e]	1.52×10^8
^{245}Cm	133.0	4.11×10^8
	174.0	4.19×10^8

[a] Data for uranium isotopes are from Ref. 6; data for plutonium isotopes are from Ref. 7 (energy and branching ratio) and Ref. 8 (half-life). Data for ^{237}Np, ^{243}Am, and ^{245}Cm were compiled by Los Alamos NIS-5.

[b] From the ^{238}U daughter ^{234m}Pa. Equilibrium assumed (~ 3.5 months).

[c] From the ^{237}Np daughter ^{233}Pa. Equilibrium assumed (~ 4 months).

[d] From the ^{241}Pu daughter ^{237}U. Equilibrium assumed. (~ 4 weeks).

[e] From the ^{243}Am daughter ^{239}Np. Equilibrium assumed. (~10 days).

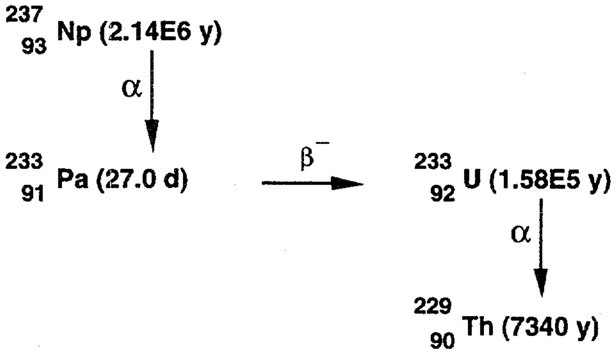

Fig. 2. Partial decay chain of ^{237}Np.

B. Neutrons

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. 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 IV lists cross-sections (probabilities) for the induced fission (σ_{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 3 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.

IV. MEASUREMENT TECHNOLOGIES

A. Safeguards for Minor Actinides at Reactor Facilities

Because the fuel in a nuclear reactor is packaged in discrete assemblies, a reactor is classified for safeguards as an "item facility." The material content of each assembly is very accurately

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

		Spontaneous	Fission	(SF)[b]	(α,n) in	Reaction Oxide[b]
Isotope	Total Half Life[a] (yr)	SF Half-Life (yr)	Neutrons per SF	Neutron Yield (n/g-s)	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. 9. Values in ( ) are from Ref. 10, from which half-lives and yields have estimated accuracies of 2 orders of magnitude. ^{240}Pu spontaneous fission rate is taken from Ref. 11.

characterized before it leaves the fuel-fabrication facility. Normally the integrity of the fuel assemblies is preserved from the time they leave the fuel fabrication facility, through reactor irradiation, and on to spent fuel storage. Fuel assemblies bear serial numbers and can be identified and verified by safeguards inspectors as the assemblies move through this portion of the fuel cycle. Seals, surveillance cameras, and radiation monitors are used to complement the safeguards provided by verification inspections.

Standard low-enriched (2-4% ^{235}U/U) uranium fuels for LWRs have relatively low proliferation value; therefore, the focus of safeguards in an LWR facility is currently on the plutonium

TABLE IV. Neutron Cross-Sections for Selected Isotopes[12]

	Thermal			1 MeV			14 MeV		
Isotope	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	-

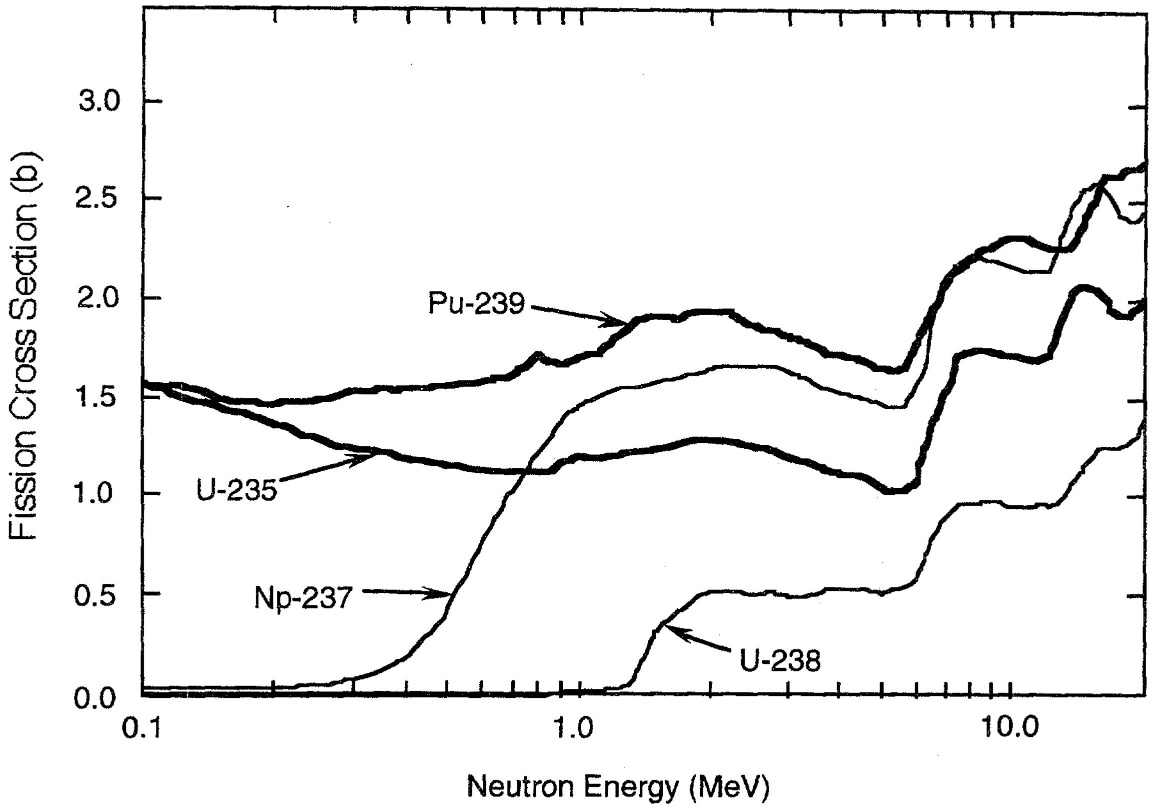

Fig. 3. Fast-neutron-induced fission cross-sections for ^{239}Pu, ^{235}U, ^{237}Np, *and* ^{238}U.

generated upon irradiating the fuel. Once the fuel is discharged from the reactor, it is "self-protected" by its intrinsic high-fission-product radiation. (This level of self-protection has been called the "spent-fuel standard.") The bulk of the safeguards effort is focused on item accounting, sometimes supplemented with NDA verification as described below.

Item accounting becomes more tedious if facility operations involve any disassembly and reconstituting of spent fuel assemblies, e.g., pin replacement for leakers or compaction of fuel rods for storage. Accounting for the plutonium in spent fuels is based on detailed calculations with burnup codes such as EPRI-CINDER[13,14] and ORIGEN,[15] which use input data of initial fuel enrichment, management and exposure, and cooling. Comparison with the amount of plutonium recovered from reprocessing has proved such calculations to be accurate, usually to within 5% for PWR fuel. The differences between the measured and calculated plutonium isotopic compositions ranged up to 13%;[2] however, recent agreement has been found as low as 2%.

Because the MAs are also generated in the reactor and with abundances of at least a factor of ten less than plutonium, the same safeguards procedures will suffice for these materials until the spent fuel is reprocessed. The MA isotopes are already included in the burnup/generation codes. A very small cost for safeguarding MAs would be incurred for carrying them on the material book inventory. An issue to be addressed is the accuracy of the burnup/generation codes for predicting MA concentrations.

The characteristics of power reactor fuel involved with typical 1000-MWe LWRs are presented in Table V.[1,16,17,18] For the transuranic production values, we assumed a reactor exposure of 31 GWd/tonne and off-loading/refueling once each year. The numbers given are approximate. The americium in the spent fuels results from the decay of ^{241}Pu and, after a few decades, grows in by the amount of ^{241}Pu (13% isotopic abundance relative to total plutonium) generated during reactor exposure. Thus, the total potential MA (neptunium + projected americium) produced per year by one LWR is about 55 kg. Higher exposure would increase these MAs approximately proportionally. For MOX recycle reactor feed (include recycle U), the MA amounts would be considerably larger, e.g., about a factor of two greater for neptunium.

The inventories of fuel assemblies at different locations within typical power stations are given in Table VI. In the US, spent-fuel assemblies that have been operated for several to many years are usually stored on-site because the back end of the fuel cycle, either reprocessing or permanent storage, is not done. In Japan and France, on-site residence times for spent fuel are only a few years.

The IAEA's significant quantities of ^{235}U in LEU, and plutonium in LWR MOX and spent fuel, together with the approximate number of LWR fuel assemblies to contain them, are shown in Table VII.

TABLE V. Typical Power Reactor Fuel Characteristics

Category	Typical PWR	Typical BWR
Fuel enrichment	2-4%	2-3%
Core inventory	200 fuel assemblies (100,000 kg)	750 fuel assemblies (150,000 kg)
Reload	65 fuel assemblies/yr (approx. 1/3 core)	190 fuel assemblies/yr (approx. 1/4 core)
Spent fuel	65 fuel assemblies/yr	190 fuel assemblies/yr
Plutonium production	320 kg/yr	320 kg/yr
Plutonium content of spent fuel	4.9 kg/assembly	1.7 kg/assembly
Neptunium production	14 kg/yr	14 kg/yr
Neptunium content of spent fuel	215 g/assembly	74 g/assembly

TABLE VI. Typical Fuel Inventories at Nuclear Power Stations (Number of Fuel Assemblies)

Reactor Type	Fresh-Fuel Storage	Spent Fuel in Core	Spent-Fuel Storage
PWR	75	200	Few hundred
BWR	200	750	Several hundred

TABLE VII. **Significant Quantities and Detection Times**

Material	Mass (kg)	Detection Time	No. of PWR Assemblies	No. of BWR Assemblies
^{235}U in LEU Feed	75	1 year	Approx. 6	Approx. 13
Plutonium in LWR MOX Feed	8	1-3 weeks	1	2
Plutonium in Spent Fuel	8	1-3 months	2	5

We conclude from this table and other data that the presence of MAs in spent fuels in a reactor facility does not add significantly to the existing problem of safeguarding the plutonium contained in the same fuel.

The only added safeguards requirement would be to carry on inventory, for each fuel assembly, the MA abundances obtained from reactor operating history and burnup codes. Applicable decay or growth of these materials would have to be propagated, as is currently done for the isotopes of plutonium. Verification measurements of spent fuels that correlate with declarations by facility operators based on burnup codes would indirectly corroborate the MA assignments.[19,20]

Although we have not included details of typical heavy water reactors (HWRs), the essence of their safeguards is the same as for LWRs. HWR fresh fuel is natural uranium; the unloading/refueling is continuous. The HWR core inventory is about 98 tonnes, comprising 4560 fuel bundles. The continuous discharge rate is 16 bundles (21.5 kg each) per day, resulting in a much greater annual accumulation of spent fuel assemblies than from LWRs. Also, the plutonium production is approximately 50% greater for HWRs than PWRs because of greater neutron capture in ^{238}U. Despite much lower burnup, the plutonium isotopic composition at discharge for HWRs (~10 GWd/tU) is similar to that for PWRs at discharge (~30 GWd/tU). Because of the lower exposure compared with LWRs, spent HWR fuel assemblies are not highly self-protecting (because of fission products) after a cooling time of more than 3 years.

B. Safeguards for Minor Actinides at Reprocessing Plants

Application of safeguards to MAs at reprocessing plants presents the greatest challenge of any component of the fuel cycle. In the reprocessing plant, the first appearance of separated, direct-use MAs in the fuel cycle could likely occur.

Take the example of a large reprocessing plant such as the Japanese Rokkasho-mura facility with a design basis of 800 tHM/yr and assume single-cycle LWR fuel reprocessing. The fuel is assumed to have experienced 50 GWd/tU exposure and a 2-year cooling time. The initial LWR fuel enrichment is taken to be 2.5%. The data of Table VIII are calculated from Ref. 2 and Table I. For the MAs, 1 SQ is assumed to be at least 25 kg for this comparison, as mentioned in Section II.A.

TABLE VIII. SNM and MA Throughputs for an 800-tHM/yr Commercial Reprocessing Plant

Material	Throughputs (kg/SQ):
^{235}U (LEU)	1408/19
Pu	7200/900
^{237}Np	422/17
$^{241}Am + ^{243}Am$	493/20

The data of Table VIII show that, compared with plutonium and ^{235}U present as LEU, MAs would contribute ~4% to the total number of SQs moving through the plant each year. These quantities, while small compared to plutonium, are large enough to warrant measurements and accounting, even if the established SQ values are much larger than the assumed 25 kg.

For all streams with concentrations of SNM and/or MAs above discard limits, volumes and concentrations are usually required for accountability. Adaptation and modification of existing measurement methods would be required for MA measurements in dissolver and product streams. Waste streams with significant concentrations and flows of MAs could require accountability measures not required before. MA waste stream concentrations depend strongly on specific processes. Examples of waste streams with potentially accountable MA concentrations include those containing fission products. NDA methods will need to be developed or adapted for international verification measurements of MAs in high-level waste (HLW) streams. As is common practice, calibrated accountability tanks will provide volume measurements at KMPs. Referring to Fig. 1, KMPs could be required at all points indicated for the reprocessing plant. These include the dissolver solution, leached hulls, plutonium product solution, uranium product solution, any MA product solution streams, and waste streams. Measurements of separated plutonium product and uranium product will need to include MA. Most importantly, NDA measurement methods will need to be adapted or developed for separated MAs. Destructive analysis methods exist for MA measurements in reprocessing plants. However, these are probably too costly to use as the sole measurements for material accounting.

Following is a summary review of measurement methods currently available or applicable to SNM and MA measurements for MC&A in reprocessing plants. Development needs for MA are identified along with approximate development costs. Plant-specific systems studies are required to define the measurement performance requirements necessary to meet inspection criteria, which also have to be developed for MAs. Therefore, not all methods would be required in any one plant, or in addition to existing measurement capabilities, e.g., at Rokkasho, Tokai, or THORP.

1. Dissolver, Intermediate, and Product Solutions. 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 matrices, in the various solution streams.

a. Traditional Destructive Analysis (DA) Methods. Traditional methods for determining plutonium and uranium concentrations in reprocessing streams involve sampling, sample preparation, and analyses. At least three different analytical techniques have been used for plutonium concentrations; isotope dilution mass spectrometry (IDMS) is the current standard method, but coulometry is also common. Wavelength-dispersive x-ray fluorescence (XRF) of L x-rays has been used at Karlsruhe for a number of years on a laboratory scale.[21,22] Spectrophotometry of Pu (VI) has also been used.[23] These methods provide accurate results, but require complex procedures and skilled operators for sample preparation and calibration. Further development of DA methods for MAs is required.

b. Hybrid XRF K-edge Densitometer. An alternative method that requires no sample preparation combines two NDA techniques in one instrument: energy-dispersive K-edge, x-ray absorptiometry (K-edge XRA) and energy-dispersive XRF analysis of K x-rays. The hybrid K-edge XRA/K-XRF system was originally developed at Karlsruhe[24] and later implemented at the French La Hague reprocessing plant. A similar system has been implemented by Los Alamos at the Japanese Nuclear Fuel Cycle Safety Engineering Research Facility (NUCEF)[25]under JAERI contract.

A simulation code[26] has been developed that predicts Hybrid XRF K-edge Densitometer assay precisions for uranium, plutonium, neptunium, and americium. Table IX gives a summary of the MA measurement precisions for dissolver and product streams for the reprocessing of LWR and FBR fuels. Typical ratios of MAs to plutonium and a 20-minute counting time were assumed.

Using this system for the input dissolver stream with typical uranium and plutonium concentrations of 200 and 2 g/l, respectively, a Pu/Np ratio of 30, and a Pu/Am ratio of 30, neither the neptunium or americium can be detected because of uranium interference. This implies an isotope dilution step would be required to reduce the uranium concentration, as indicated in Table IX. This would be similar to the sample-preparation step used with isotope dilution gamma spectrometry (IDGS) on resin beads.[27] With a dilution of the uranium concentration, this system should produce acceptably accurate and precise assays for neptunium and americium. Depending on the uranium dilution, 2% precision for neptunium and americium could be obtained.

Precision of MA measurements could be improved by adding another sample preparation step, which involves putting the resin bead into solution and evaporating it dry on filter paper. When this sample is measured in the hybrid system, measurement precision would be improved because incoherent scattering produced by the matrix liquid would be eliminated. The simulation results for partial uranium removal and uranium removal combined with evaporation are shown in Table X.

TABLE IX. Simulated Precisions of the Hybrid XRF K-edge Densitometer for Measurements of MAs in Reprocessing Plant Streams

Stream	U conc. (g/l)	Pu conc. (g/l)	Pu/Np ratio	Pu/Am ratio	Np precision (%)	Am precision (%)
LWR Dissolver	200	2	30	30	<2*	<2*
Pu Product (LWR)		200	30	300	0.5	3.1
FBR Dissolver	200	50	10	10	0.5	0.5
Pu Product (FBR)		200	10	300	0.2	3.3

*Requires partial removal of uranium and concentration of solution. Improved precision can be obtained with an additional evaporation step.

TABLE X. Simulated Precisions of the Hybrid XRF K-edge Densitometer for Measurements of Pu, Np, and Am in LWR Reprocessing Plant Pu Dissolver Solutions: Pu/Np = 30, Pu/Am = 30. Partial U Removal, Evaporation

Stream	U conc. (g/l)	Pu conc. (g/l)	Pu/Np ratio	Pu/Am ratio	Pu precision (%)	Np precision (%)	Am precision (%)
LWR Dissolver	200	2	30	30	1		
U removal, evaporation	120	40	30	30	0.3	2.8	2.1
U removal, evaporation	40	40	30	30	0.3	2.6	2.0
U removal, evaporation	10	100	30	30	0.2	0.8	0.6

Calculated MA measurement precisions for reprocessed plutonium product solutions from LWRs and FBRs are shown in Table XI. Fixed Pu/MA ratios were assumed and the plutonium concentration was varied.

The cost of an existing Hybrid XRF K-edge Densitometer system would be approximately $500K. A plutonium densitometry system exists at Tokai, but it does not have XRF capability. Physics development, testing, standards preparation, and experimental analyses are needed to determine the capability of the Hybrid system to assay MAs. Also, specialized physics analysis software will need to be developed.

TABLE XI. Simulated Precisions of the Hybrid XRF K-edge Densitometer for Measurements of Pu, Np, and Am in Reprocessed LWRs and FBRs

LWR Pu Product Solution **Pu/Np = 30, Pu/Am = 300**			
Pu concentration	Pu precision (%)	Np precision (%)	Am precision (%)
50	0.5	1.2	10
100	0.3	0.7	5.8
150	0.2	0.6	3.7
200	0.2	0.5	3.1
FBR Pu Product Solution **Pu/Np = 10, Pu/Am =300**			
Pu concentration	Pu precision (%)	Np precision (%)	Am precision (%)
50	0.5	0.6	8.3
100	0.3	0.5	5.1
150	0.2	0.3	3.8
200	0.2	0.3	3.3

c. Isotope Dilution Gamma Spectrometry (IDGS). This technique[27] simultaneously determines the plutonium concentration and isotopic composition of dissolver and intermediate solutions from the reprocessing plant. The method combines high-resolution, low-energy gamma-ray spectroscopy; isotope dilution; and plutonium spiking. The IDGS technique involves adding a well-characterized (in concentration and isotopics) plutonium isotope to the unknown solution, extracting the plutonium from the spiked (mixed) solution on resin beads, and subsequently measuring the beads using high-resolution gamma-ray spectroscopy (HRGS). In the dilution step, fission products, uranium, and americium are removed. The HRGS analysis is based on intense low-energy plutonium gamma rays.

It is assumed that in the isotope-dilution step, neptunium is not diluted but carried along with the plutonium. It must be determined whether or not ^{233}Pa, the short-lived daughter of ^{237}Np, would be separated in this step. If so, a non-equilibrium state would exist for several months, but could easily be corrected by knowing the date of separation. Whether or not equilibrium between ^{237}Np and ^{233}Pa is the dominant case, a separate physics analysis algorithm for the HRGS analysis of ^{237}Np would have to be developed for IDGS.

Developments are required to determine the feasibility and accuracy of the IDGS method for measurement of MAs.

d. Neutron Inventory Sample Counter (INVS). References 2 and 19 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 INVS neutron coincidence counter,[28] 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, as described above (IV.B.1.b.). A similar approach has been inferred by Miura and Menlove.[29]

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 an americium balance. The feasibility of this method can be determined only if details of facility-specific processes are known. The "elemental balance method" appears to possess significant promise for verifying SNM and MA in waste streams, but has yet to be demonstrated.

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

2. Product Oxides. For separated, pure plutonium oxide, the standard NDA methods are neutron coincidence counting and HRGS. The High-Level Neutron Coincidence Counter (HLNCC)[30] 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[31] or MGA[32] 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.[33] 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[34] 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.

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.

3. Residues. This 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.

4. Waste Streams.

a. High-Level Waste. As mentioned in Sec. IV.B.1.d, for the head-end of the process, including the spent fuel, dissolver solution, and leached hulls, a direct INVS measurement of ^{244}Cm, using neutron multiplicity counting and combined with measurements of elemental ratios using DA or NDA, can yield mass balances for plutonium, neptunium, and americium. The elemental ratios are expected to remain nearly constant at the head-end of the process. This elemental balance method can be used at any other measurement point in the process where desired elemental ratios remain constant. The Hybrid XRF K-edge Densitometer could be used to measure elemental concentrations of samples with known volumes. Because the sample volumes are known, the total element mass can be determined and results of the two measurements can be combined to yield elemental ratios. The ratios are then used to determine the mass balances for elements other than curium. In addition to mass balances, the ratios or "tags" could be used as signatures of declared separation ratios at selected points in the process. To lower the overall costs of DA, it is necessary to develop the ability of the Hybrid XRF K-edge Densitometer to measure neptunium and americium concentrations. As mentioned earlier, this could involve an extra isotope-dilution step and possibly a matrix evaporation step, e.g., for the dissolver solution.

At the back-end of reprocessing, where the high-level waste stream is evaporated and the fission products and residual uranium and transuranics are entrained in glass, the neutrons from the ^{244}Cm content in the vitrified glass logs can be measured. Neutron coincidence counting would be required to assay the ^{244}Cm content because the boron in the glass produces many more, interfering, (α,n) neutrons. The method is currently being pursued for independent IAEA verification of plutonium in the glass.[35] In contrast to the plutonium concentration in the head-end dissolver solution of a few grams per liter, the concentration in the glass may be a few tenths of a gram per liter. The elemental curium ratios needed to infer the MA as well as the plutonium contents of the glass logs could be obtained by sampling the upstream, calibrated, high-level-waste accountability tank (if there is one) and performing DA or NDA. The capability of the Hybrid K-edge/XRF system, in combination with INVS, to measure Cm/Pu and Cm/Am ratios should be investigated.

IDMS, a DA technique in which an isotopic spike of a known amount is added, can be used to analyze low concentrations of transuranics, as may be present in high-level liquid waste. Similarly, IDAS (isotope dilution alpha spectroscopy) can be used with good accuracy in the concentration range 10-100 mg/l. At Los Alamos, Laser-Induced Breakdown Spectroscopy (LIBS) has been investigated for several applications, including transuranic concentrations. Recently, a new technique with only one chemical preparation step has been investigated by the staff of PNC, Tokai-mura, Japan.[36] This technique, called LIPAS (laser induced photoacoustic spectroscopy), employs an 830.6 nm beam of light from a YAG laser with an optical parametric oscillator to excite plutonium VI. The induced photoacoustic signal is then detected with piezoelectric transducers. The limit of sensitivity for plutonium is 1 mg/l, and it is free of interference from fission products. The method has not yet been applied to MA; clearly, more research is needed to establish its feasibility.

Once the facility-specific processes and physical layout of the high-level waste stream are known, a specific measurement strategy can be pursued. For example, if neptunium is separated upstream, the residual amounts in the high-level waste may be so small as to be completely negligible in the material balance compared to the level of one significant quantity. Conversely, if all of the americium goes out with the fission products, its concentration would probably be larger than that of plutonium. In this case, the NDA method of the Hybrid K-edge/XRF system may be well suited to the measurement of americium.

In all cases, requirements for measurements of SNM and MA in vitrified glass are conditioned upon concentrations, discard limits, and termination of safeguards.

Development of standards and experimental tests is required to determine the feasibility and accuracy of these techniques.

b. Low-Level TRU Waste. Low-level, low-density, heterogeneous radioactive waste containing SNM and MA isotopes, each with sufficiently intense and characteristic gamma-ray emission, can be assayed using the recently developed method of tomographic gamma scanning (TGS).[37] Earlier methods for NDA of SNM and transuranic (TRU) waste in 208-L drums can give assay errors of 100% or more when the drum matrix or radionuclide distribution or both are nonuniform. This problem is addressed by TGS, which extends the well-established segmented gamma-scanner (SGS)[38] method by forming low-resolution tomographic emission and transmission scans on the drum, yielding coarse three-dimensional images of the matrix density and radionuclide distributions. The images are used to make accurate point-to-point attenuation corrections. A TGS system can be operated in both SGS and TGS modes.

C. Fuel Fabrication Plants

For bulk-handling facilities, where neptunium is present with plutonium in trace amounts (0.5 to 1.0%) and americium is present because of ^{241}Pu decay, HRGS can be adapted to determine the neptunium, americium, and plutonium. Neutron coincidence and multiplicity counting can be used to verify the effective ^{240}Pu mass of bulk material or items. Traditional DA methods are also applicable. This topic is covered in Sec. IV.B.2 (Product Oxides).

In the future, bulk-handling facilities processing MAs in greater quantities, e.g., a Pu-Np-Am-U fuel fabrication plant, could require improved DA procedures because the MAs can no longer be treated as trace impurities. Existing NDA methods would be applied to future Pu-Np-Am-U fuels (HRGS and neutron multiplicity counting), but precision and accuracy criteria for MAs will be more stringent because of larger flow rates.

V. SUMMARY AND RECOMMENDATIONS

While most of the MAs produced by nuclear power reactors reside in spent fuels in temporary storage, some countries, most notably France, Japan, the United Kingdom, and Russia, have chosen to reprocess their spent fuels and to utilize the recovered plutonium, and possibly, the MAs, as reactor fuels. The MAs constitute a significant fraction, 13-16%, of the amount of plutonium in high-burnup LWR spent fuel. Worldwide annual production of neptunium is about 3.5 tonnes, and the rate of production of americium, including the potential ^{241}Am from the decay of ^{241}Pu, is comparable. In addition to the incentive for recovering the MAs for their fuel value, there is a very strong motivation to remove them from the fission product waste to reduce the costs, hazards, and complexities of permanent nuclear waste storage. On the other hand, the separation of MAs has been suggested to fall in the context of nuclear nonproliferation.

For those who reprocess, the options for the separation and utilization of the MAs include passing them through with the plutonium, separating MA product solutions and blending them with plutonium nitrate to be used for specific reactor fuels, and producing separate MA oxides for future blending with plutonium oxide.

Because of the presence of the MAs in many forms, either separately or with plutonium, there is a challenging need to extend existing and develop new measurement methods that are fast and economical. Even small concentrations of MAs in plutonium mixes could compromise the accuracy of the plutonium measurement if not properly taken into account: consequently, safeguards of plutonium could be affected. To date, little effort has been directed to the specific problem of measuring the MAs in the reprocessing environment, especially nondestructive methods. Measurement has not been a problem as long as MAs passed [illegible] with the fission product waste or remained in stored spent fuels.

Fortunately, there are a number of NDA methods and instruments, reviewed here, that appear to be readily adaptable to the measurement of the MAs. [illegible] should be particularly well suited for measurement of MAs because of the economic factors of handling and accounting for the smaller (than plutonium) abundances in processing facilities. The added costs for inclusion of MAs in materials accounting should be partially mitigated by adapting the same instruments and procedures that are used for measuring plutonium. Much of the adaptation will involve changes in physics algorithms and software as well as testing on typical, real MA standards. Standards are needed to establish feasibility and determine measurement accuracies. In some cases, the combination of NDA and some sample preparation involving chemistry may be required. A summary of the NDA applications for MAs within reprocessing and fuel fabrication facilities is given in Table XII. Estimates of capital (instrument) costs and personnel development time are included in the table.

Destructive (chemical) analysis methods will also have to be adapted and extended to measure MAs. Such methods as isotope dilution mass spectroscopy, isotope dilution alpha spectroscopy, and laser-induced breakdown spectroscopy as well as purely chemical techniques, will need to be tested and calibrated with standards.

To keep pace with current experimental advanced fuel programs, we recommend that a vigorous development and testing program for the measurement of MAs in the environments of reprocessing and fuel fabrication facilities be initiated. The first phase of such a program should comprise the following:

1. a systems study of a real reprocessing facility, e.g., the 800-tonne/yr Rokkasho-mura plant, to characterize the process streams and establish measurement criteria and

2. laboratory measurements of characteristic MA samples using existing NDA instruments adapted for this purpose.

Under subsequent phases of this program, techniques for measuring MAs would be fully developed, tested, and the results incorporated into appropriate technology transfer and training activities.

ACKNOWLEDGMENTS

The authors would like to thank John Rooney and Ira Goldman of the US Department of Energy; Bill Murphey of the Nuclear Regulatory Commission; and Jim Sprinkle, Norbert Ensslin, Jack Parker, Tom Sampson, and T. K. Li of the Los Alamos Safeguards Science and Technology Group for many helpful discussions and comments provided during the preparation of this report.

TABLE XII. Providing Nondestructive Measurement Capabilities for the Minor Actinides: Applications and Estimated Costs

Measurement Method	Measured Quantities	Measured Quantities	Equipment Cost ($K)	Development Effort (FTEs)[c]
Hybrid XRF K-edge Densitometer	Dissolver, intermediate and product solutions; high-level waste	U, Pu, Np, Am concentrations	500[a]	2-4
Isotope Dilution Gamma Spectrometry (IDGS)	Dissolver and intermediate solutions	Pu concentration and isotopic composition, ^{237}Np concentration	150[b]	2-4
Neutron Inventory Sample Counter (INVS)	Dissolver solution sample, high-level waste	^{244}Cm concentration	100[a]	2-4
Neutron Multiplicity Counting (NMC)	Product oxides, residues	^{237}Np, ^{240}Pu$_{eff}$ masses	450	4-6
High-Resolution Gamma Spectrometry (HRGS)	Product oxides, residues	^{237}Np/Pu, Am/Pu, Pu isotopic composition	50	4-6
Neutron Coincidence Counter (NCC)	High-level waste in vitrified glass logs	^{244}Cm content	400	4-6
Destructive Analysis Methods (IDMS, IDAS, LIBS, LIPAS, etc.)	High-level waste	SNM, MA concentrations	200	4-6
Segmented Gamma Scanner (SGS)/ Tomographic Gamma Scanner (TGS)	Low-Level Transuranic (TRU) Waste	SNM, MA masses	250/400	6-9

[a] includes some sample shielding and handling provisions

[b] existing facility systems, e.g., resin-bead system could be used

[c] full-time equivalent years of personnel effort

REFERENCES

1. L. Koch, "Formation and Recycling of Minor Actinides in Nuclear Power Stations," *Handbook of Physics and Chemistry of the Actinides*, A. J. Freeman and C. Keller, Eds. (Elsevier Science Publishers B. V., 1986), Chapter 9, pp. 457-490.

2. G. E. Bosler, J. R. Phillips, W. B. Wilson, R. J. LaBauve, and T. R. England, "Production of Actinide Isotopes in Simulated PWR Fuel and Their Influence on Inherent Neutron Emission," Los Alamos National Laboratory report LA-9343 (July 1982).

3. N. Sasao, "PNCs View and Programs on the Actinide Recycle," A Presentation for the CEA/PNC Meeting on Actinide Recycle, Japan, 1994.

4. R. J. S. Harry, "IAEA Safeguards and Non-Proliferation," presented at "The Clingendael Nuclear Non-Proliferation Workshop," September 8-10, 1994, at the Netherlands Institute of International Relations Clingendael, Netherlands Energy Research Center report ECN-RX–95-005 (February 1995).

5. K. Chitumbo, "IAEA Safeguards at Power Reactors" Session 18 - International Training Course on Implementation of State Systems of Accounting for and Control of Nuclear Materials, April 19-May 5, 1995, Los Alamos National Laboratory.

6. C. M. Lederer and V. S. Shirley, Eds., *Table of Isotopes*, 7th ed. (John Wiley & Sons, Inc., New York, 1978).

7. R. Gunnink, J. E. Evans, and A. L. Prindle, "A Reevaluation of the Gamma-Ray Energies and Absolute Branching Intensities of ^{237}U, ^{238,239,240,241}Pu, and ^{241}Am," Lawrence Livermore National Laboratory report UCRL-52139 (1976).

8. "Calibration Techniques for the Calorimetric Assay of Plutonium-Bearing Solids Applied to Nuclear Materials Control," ANSI N5.22-1975 (American National Standards Institute, Inc., New York, 1975) and 1986 revision.

9. R. T. Perry and W. B. Wilson, "Neutron Production from (α,n) Reactions and Spontaneous Fission in ThO_2, UO_2, and (U,Pu) O_2 Fuels," Los Alamos National Laboratory report LA-8869-MS (June 1981).

10. D. L. Johnson, "Evaluation of Neutron Yields from Spontaneous Fission of Transuranic Isotopes," *Transactions of the American Nuclear Society* **22**, 673 (1975).

11. P. Fieldhouse, D. S. Mather, and E. R. Culliford, "The Spontaneous Fission Half-Life of ^{240}Pu," *Journal of Nuclear Energy* **21**, 749 (1967).

12. H. M. Fisher, "A Nuclear Cross Section Data Handbook," Los Alamos National Laboratory report LA-11711-M (December 1989).

13. T. R. England, W. B. Wilson, and M. G. Stamatelatos, "Fission Product Data for Thermal Reactors, Part 1: A Data Set for EPRI-CINDER Using ENDF/B-IV," Electric Power Research Institute report EPRI NP/356, Part 1 (December 1976). Also published as Los Alamos Scientific Laboratory report LA-6745-MS (December 1976).

14. T. R. England, W. B. Wilson, and M. G. Stamatelatos, "Fission Product Data for Thermal Reactors, Part 2: Users Manual for EPRI-CINDER Code and Data," Electric Power Research Institute report EPRI NP/356, Part 2 (December 1976). Also published as Los Alamos Scientific Laboratory report LA-6746-MS (December 1976).

15. A. G. Croff, M. A. Bjerke, G. W. Morrison, and L. M. Petrie, "Revised Uranium-Plutonium Cycle PWR and BWR Models for the ORIGEN Computer Code," Oak Ridge National Laboratory Report ORNL-6051 (September 1978).

16. S. Glasstone and A. Sesonske, *Nuclear Reactor Engineering*, 3rd Edition (Van Nostrand Reinhold, 1981).

17. "World Nuclear Industry Handbook 1994," *Nuclear Engineering International* magazine annual supplement, ISSN 0029-5507, pp. 12-100 (1994).

18. A. V. Nero, Jr., *A Guidebook to Nuclear Reactors* (University of California Press, Berkeley 1979), ISBN 0-520-03482-2.

19. S. T. Hsue, J. E. Stewart, K. Kaieda, J. Halbig, J. Phillips, D. Lee, and C. Hatcher, "Passive Assay of Irradiated Nuclear Fuels," Los Alamos Scientific Laboratory report LA-7645-MS (February 1979).

20. J. R. Phillips, "Irradiated Fuel Measurements," Chapter 18 in *Passive Nondestructive Assay of Nuclear Materials*, Doug Reilly, Norbert Ensslin Hastings Smith, Jr., and Sarah Kreiner, Eds. (Washington, D.C., U.S. Gov. Printing Office, 1991) NUREG/CR-5550, p. 529.

21. A. V. Baeckmann, D. Ertel, and J. Neuber, *Advances in X-ray Analysis* **18**, 62 (1975).

22. Mainka, et al., "Fully Automated, Wavelength Dispersive X-Ray Fluorescence Analysis of Uranium and Plutonium in Reprocessing Plant Solutions," Int. Symp. on Recent Advances in Nuclear Materials Safeguards, IAEA, Vienna, 8-12 November 1982, Contribution IAEA-SM-260/32, Vol. 2, p. 249.

23. P. Patigny, "Analyses de Bilan Entree Sortie A. L'Usine de la Hague Verification de L'Exactitude Des Resultates Obtenus," *ESARDA* **15** 145 (1982).

24. H. Ottmar and H. Eberle, "The Hybrid K-Edge/K-XRF Densitometer: Principles-Design-Performance," Kernforschungszentrum Karlsruhe report KfK 4590 (February 1991).

25. S. T. Hsue, J. K. Sprinkle, Jr., R. Cole, W. Hanson, and G. Walton, "Development of Hybrid [K-edge/X-Ray Fluorescence (XRF)] Densitometer," in Safeguards and Security Research and Development Progress Report, October 1992-September 1993," Los Alamos National Laboratory report LA-12816-PR (October 1994).

26. S. T. Hsue, private communication, Los Alamos National Laboratory.

27. T. K. Li, et al., "Determination of Plutonium Concentration and Isotopic Compositions by Isotope Dilution Gamma-Ray Spectroscopy on Resin Beads," Los Alamos National Laboratory report LA-11827 (1990).

28. H. O. Menlove, D. Davidson, J. Vermuelen, H. G. Wagner, R. Wellum, B. Brandelise, and K. Mayer, "Design and Performance of a New High Accuracy Combined Small Sample Neutron Gamma Detector," *Nucl. Mat. Manage.* **XXII** (Proc. Issue), 872-880 (1993).

29. N. Miura and H. O. Menlove, "The Use of Curium Neutrons to Verify Plutonium in Spent Fuel and Reprocessing Wastes," Los Alamos National Laboratory report LA-12774-MS (May 1994).

30. H. O. Menlove and J. E. Swansen, "A High-Performance Neutron Time-Correlation Counter," *Nucl. Technol.* **71,** 497-505 (1985).

31. T. E. Sampson, G. W. Nelson, and T. A. Kelley, "FRAM: A Versatile Code for Analyzing the Isotopic Composition of Plutonium from Gamma-Ray Pulse-Height Spectra," Los Alamos National Laboratory report LA-11720-MS (December 1989).

32. R. Gunnink and W. D. Ruhter, "MGA: A Gamma-Ray Spectrum Analysis Code for Determining Plutonium Isotopic Abundance," Lawrence Livermore National Laboratory report UCRL-LR-103220, (Vol. 1 "Methods and Algorithms," April 1990; Vol. 2, "A Guide to Using MGA," Sept. 1990).

33. S. Baumann, et al., "Determination of Plutonium-Uranium Isotopes and Fission Products with a Newly Developed Telescope-Germanium Detector and a Modified Two Detector MGA Code," presented at the 15th ESARDA Symposium, Rome (May 1993).

34. D. G. Langner, M. S. Krick, and N. Ensslin, "Pyrochemical Multiplicity Counter Design," *Nucl. Mater. Manage.* **XIX**, 411 (1990).

35. H. O. Menlove, private communication, Los Alamos National Laboratory.

36. N. Surugaya, S. Sato, A. Kurosawa, M. Kamata, Y. Kuno, and J. Masui, "Study on Determination of Small Amount of Plutonium in Highly Radioactive Solutions," in *Proceedings of the IAEA Symposium on International Safeguards* (IAEA, Vienna, Austria, 1994).

37. R. Estep, T. H. Prettyman, and G. A. Sheppard, "Tomographic Gamma Scanning to Assay Heterogeneous Radioactive Waste," *Nucl. Sci. Eng.* **118**, 145-152 (1994).

38. E. R. Martin, D. F. Jones, and J. L. Parker, "Gamma-Ray Measurements with the Segmented Gamma Scan," Los Alamos Scientific Laboratory report LA-7059-M (December 1977).

Los Alamos
NATIONAL LABORATORY
Los Alamos, New Mexico 87545