**NDA SAFEGUARDS TECHNIQUES
FOR LMFBR ASSEMBLIES**

by

P. J. Persiani and M. L. Gundy

ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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ARGONNE NATIONAL LABORATORY
9700 South Cass Avenue
Argonne, Illinois 60439

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P. J. Persiani and M. L. Gundy

Applied Physics Division
Argonne National Laboratory
Argonne, Illinois 60439

ABSTRACT

The significant safeguards concerns for liquid-metal fast breeder reactors (LMFBRs), and for the LMFBR fuel handling systems are the accountability, surveillance, and identification of fuel and blanket assemblies. The introduction of fuel assemblies with a high content of Pu into the receiving and shipping areas of the LMFBR fuel cycle does allow a more direct near-real-time assay profile of the disposition of Pu. Isotope correlations and neutron assay methods have been investigated and implemented for determining plutonium and burnup in fresh and spent LMFBR fuel assemblies. The methods are based on active and passive neutron coincidence counting (NCC) techniques. Preliminary studies on neutron yield rates from the spontaneous fission of plutonium and curium isotopes have indicated that the NCC system is a most effective measure in the verification of nuclear material flow in assembly form for the entire reactor fuel handling cycle i.e., from the fresh-to the spent-fuel stage. A consequence of the high plutonium concentration level throughout the fuel irradiation period in an LMFBR, is that the spontaneous fission neutron yield from the 242-curium and 244-curium does not dominate the spontaneous fission neutron yield from the plutonium isotopes in the spent fuel stage.

I. INTRODUCTION

This study describes selected nondestructive assay safeguards techniques which may be applicable for current and near-term fast breeder reactors (FBR).

Safeguards for nuclear fuel cycles should be considered in the context of a world deployment of various reactor types and a varying availability of fuel cycle services. This is particularly true of the current experimental, demonstration, and prototypal fast breeders, which must be fueled and safeguarded in the transition period. The quantities of plutonium and uranium fissile materials, the fuel handling systems, and the storage methods for the fuel assemblies of fast breeders will have a significant impact on safeguards techniques. The plutonium and fissile uranium throughput, the high fissile content, and the use of sodium coolant for the liquid-metal fast breeder reactor (LMFBR) fuel cycles may require development of additional safeguards techniques beyond those for the thermal light-water reactor (LWR) fuel cycle:

The feasibility of, and the limitations and constraints on, the application of certain safeguards measures and techniques for the LMFBR are not in common with LWR systems. Important distinctions of the LMFBR fuel cycle are:

the total quantity of nuclear material in inventory and in flow; the concentration levels of nuclear material in single items (assemblies); the assembly-handling operations and storage facilities; the reactor operating modes. The total quantity of nuclear material in inventory and in flow for each demonstration and commercial breeder reactor system would be 3,000-10,000 kg of plutonium, which is in the range more of the annual flow rates of 6,000-12,000 kg for current and proposed large LWR reprocessing plants. The content of the fissile material, 6-15 kg of plutonium, in each of the fresh and spent fuel assemblies is on the order of the IAEA significant-quantity guideline of 8 kg.

A timely perspective for developing LMFBR safeguards is obtained by considering that the existing experimental reactors have plutonium flow rates and inventory quantities (ranging from 1/2 to 2 tonnes of plutonium) which is comparable to some current size reprocessing and mixed oxide fabrication plants having flow rates of about 200 tonnes of heavy metal (1 to 2 tonnes of plutonium) per year. Consequently, in the context of near-term nuclear power development, the deployment of LMFBRs and reprocessing facilities appear to be on a comparable time-scale with impact on safeguards system development needs closely coupled.

II. REFERENCE LMFBRs AND FUEL HANDLING SYSTEMS

The facility design features of the generic LMFBR reference reactors and fuel handling systems are primarily based on two general classes of reactors: (1) demonstration and commercial, and (2) experimental -- currently in operation, under construction, or in the design stage.¹ The former class includes SNR-300 (Federal Republic of Germany, or FRG), Monju (Japan), Phenix and Super Phenix (France), BN-350 and BN-600 (USSR), and the CRBR (US). The scale of the experimental reactors falls into two categories: power ratings (1) higher than 100 MWt and (2) lower than 100 MWt. In the first category are the FFTF (US) and the PFR (UK). In the second are EBR-II (US), the KNK-2 (FRG), Joyo (Japan), FBTR (India), and the BOR-60 (USSR).

The basic units of one fuel assembly and one blanket assembly have been adopted for item accountability with reactors in the normal operating mode. However, for the experimental reactors, experimental requirements and experimental-assembly designs necessitate the more restrictive item accountability by fuel element (pin). Generally, the fuel assemblies for the operating power reactors differ from those for the experimental reactors in dimensional design, dismantling capabilities, frequency and mode of handling, plutonium and ²³⁵U content, and burnup rate and average burnup. These differences necessitate investigating variations in requirements for diversion path analysis, safeguards measures, item accountability, and containment and surveillance (C/S) techniques.

Since the safeguards system will have some components that are plant-specific even within each class and category of reactor, the safeguards analysis usually includes three general considerations: (1) verification of the inventory of fresh fuel assemblies in handling and storage; (2) verification of the inventory of irradiated fuel assemblies in storage and in preparation for transport; and (3) handling systems for irradiated fuel assemblies. In some plants, experimental assemblies or fuel assemblies must be dismantled and given special handling and storage. The requirements for

verifying inventory and flow of these dismantled assemblies can be satisfied with item accountability adopted to the level of the individual fuel elements within the assembly when necessary.

The differences between the core and blanket design details for the demonstration and commercial reactors and the experimental reactors have been categorized and are summarized in Table I.

TABLE I. Data for Reference LMFBR Fuel and Blanket Assemblies

	Experimental Systems		Demonstration Systems	Commercial Systems
	<100 MWt	>100 MWt		
Number of Assemblies				
Inner Core	60	30-80	60-110	170-200
Outer Core		50	50-90	160-170
Blanket	200	50	90-170	200-400
Fuel Elements/Ass.	90	200-325	160-220	130-270
Uranium Elements/Blanket Ass.	30	80-90	80-90	40-90
Nuclear Fuel Content (Fresh/Irradiated)				
Material Form	PuO ₂ /UO ₂	PuO ₂ /UO ₂	PuO ₂ /UO ₂	PuO ₂ /UO ₂
Heavy Metal, kg/Ass.	2-14	30-50	30-55	20-95
Pu/ ²³⁵ U, kg/Ass.	1-3 Pu, 2-3 ²³⁵ U	8-13 Pu, 10-12 ²³⁵ U	6-14	3-18
Pu/ ²³⁵ U, kg/Element	0.03-0.07	0.03-0.04	0.03-0.06	0.025-0.05
Irradiated Blanket				
Pu, kg/Ass.	1	1-3	1-3	1-3
Pu, kg/Element	0.03	0.01-0.04	0.01-0.04	0.01-0.04

A. Item Flow in Fuel Handling System

The item-accountability measure of fuel and blanket assemblies in the fuel-handling storage and operations, or in the reactor-operations facility, is formed by adding all measured receipts to the initial inventory and subtracting all measured removals from the final measured inventory. During the normal flow of uniquely identified assemblies, nuclear-materials control emphasizes administrative controls supplemented by secure storage for the identified assemblies, tamper-indicating assembly seals or systems of seals, sealed containers or transport casks for receiving and shipping, etc. Limited measurement capabilities and the uncertainties of the nuclear-material content in the assemblies, may obscure the diversion of significant quantities of nuclear materials from a plant.

Item accountability and the accountability of the nuclear-material contained in the assembly become equivalent only when there is assurance that the integrity of the assembly and its contents has been maintained. The use of neutron interrogation techniques for nondestructive assay (NDA) would provide direct assurance that the integrity of the nuclear-material content has not been compromised since the fabrication of the assembly. The introduction of fuel assemblies with a high content of Pu/²³⁵U into the receiving and shipping areas of the fuel cycle does allow a more direct near-real-time

assay profile of the disposition of Pu/ ^{235}U . Information on the timely disposition of assembly flow is obtained from the direct applicability of material accountancy by NDA methods (HLNCC)^{2,3,4,5} and from the selective application of effective C/S techniques.

The distinction must be maintained between NDA neutron-coincidence counting, NCC systems of measurements for nuclear-material accounting in the sense of nuclear material balance accountancy and NDA for the verification of nuclear-material content for accountability in an assembly. It is only in the latter sense that NDA is recommended in this report.

B. Direct Accountability of Fissile Material

Both the IAEA safeguards approach for the long-term handling and interim storage of fresh and spent fuel assemblies, and the international transfer of ownership of the fuel with resolutions of shipper/ receiver balances, require direct identification of fissile content and accountability of nuclear-material balance. The currently available NDA techniques for item accountability can distinguish only between different classes of assemblies, e.g., fuel, blanket, and dummy replacement. No distinction can be made between the individual assemblies of one class. It is not clear how effective NDA techniques may be if the NDA information is not supplemented by subsequent confirmatory information that a replacement or partial replacement has been made. An added level of assurance in flow verification and nuclear-material accountancy is provided by establishing that the difference between preirradiation and postirradiation isotopic measurements reflects the irradiation history of a specific assembly.

A number of instruments which have been assessed for application as NDA safeguards for item accountability include: high resolution gamma spectroscopy (HRGS) for passive gamma-ray spectrometry of plutonium; the high level neutron-coincidence counter HLNCC for passive measurement of spontaneous fission neutrons from ^{240}Pu , and the active-well neutron-coincidence counter AWCC interrogation system for measuring induced-fission neutrons in fissile isotopes.

C. Instruments and Methods for Determining Nuclear-Material Content

1. High-resolution Gamma Spectrometry (HRGS)

In view of the high plutonium content in LMFBR mixed-oxide fuel, the plutonium assay by HRGS appears to be more effective for fresh fuel elements than for complete fresh fuel assemblies. The self-shielding effects in fuel elements are expected to be minimal, and consequently the gamma-ray spectra may be used as a quantitative measure of the plutonium content. The self-attenuation of the gamma rays in an LMFBR assembly, with or without sodium, would introduce uncertainty in the quantitative measure of the plutonium-content in a fresh fuel assembly. The direct assay of plutonium in an irradiated assembly by HRGS does not appear to be an effective measurement technique except to establish the existence of fission products.

2. High-level Neutron Coincidence Counter (HLNCC)

Preliminary studies on neutron yield rates from the spontaneous fission of the ^{238}Pu , ^{240}Pu , and ^{242}Pu have indicated that the HLNCC is

effective in the verification of nuclear-material flow in assembly form throughout the entire reactor fuel handling cycle, i.e., from the fresh-fuel through the spent-fuel stages. Because of the high plutonium concentration throughout the fuel-irradiation period in an LMFBR, the spontaneous-fission (SF) neutron yields from the ^{242}Cm and ^{244}Cm do not dominate the SF neutron yield from the plutonium isotopes in the spent-fuel stage. This is significant when compared to the problem with LWR spent-fuel interrogation where the ^{244}Cm neutron yield rate is several orders of magnitude greater than the plutonium yield rate at the same level of burnup and after the decay of the short-life (162.8 d) ^{242}Cm contribution. The relative curium isotopic concentration buildup as a function of average burnup of LWR and LMFBR fuel assemblies are compared in Fig. 1.

In the LWR system, the long-life ^{244}Cm isotope begins to dominate at burnup levels of about 20,000 to 25,000 MWd/T. In the LMFBR systems the cross-over occurs at average burnup levels of greater than 100,000 MWd/T.

Fig. 1. Curium Isotope Concentrations vs. Burnup

Initial computations of total neutron yield rates [SF and $(\alpha, n)^{18}\text{O}$] for the prominent isotopes are plotted as a function of fuel burnup in Figs. 2-3. The computations are results from a study of isotopic-safeguards techniques in LMFBR fuel cycles.⁶ Figures 2a, 2b, and 2c represent different core regions where possible differences in enrichment and spectral effects were studied. In all regions, the results indicate that, for mixed-oxide fuel with a burnup of 35,000 MWd/T, about half the total neutron yield rate is contributed by plutonium after a cooling period of 1-2 yr and subsequent decay of the ^{242}Cm isotope. From the specific isotopic neutron

Fig. 2. Neutron Yield vs. Burnup in Three Core Regions

Fig. 3. Neutron Yield vs. Burnup in Blanket Region

yields, listed in Table II, it is estimated that the plutonium SF neutrons emitted primarily by the ^{240}Pu (and some ^{238}Pu), contribute about 25-35% of the total SF neutrons emitted from the fuel.

The plutonium isotopes will make a significant contribution to the SF neutron yields at all the levels of burnup and decay-cooling times that are anticipated in the next two decades of LMFBR operation. This maintains even for a burnup of 100,000 MWd/T, where the plutonium isotopes are estimated to contribute 5-7% to the SF neutron yield, depending on decay cooling times. At burnups in the range of 75,000 MWd/T, the estimates indicate that the plutonium isotopes contribute 5-12% of the total SF neutrons after a 2-yr cooling period. For the core region with the higher concentration level of plutonium (region 56, Fig. 2c), the fraction of the plutonium SF neutrons is higher than in the core axial-center region 26 and the core axial-edge region 23 of lower enrichment. At burnups below 33,000 MWd/T, the plutonium SF neutrons dominate the neutron yield rate. These significant contributions to the neutron yield by the plutonium isotopes

make passive-neutron-assay techniques a viable method for item accountability and identification of the spent fuel and blanket assemblies in the LMFBR cycle.

TABLE II. Principal Sources of Neutrons in Irradiated UO_2 Materials

Isotope	Half-life, yr	(α, n) Reaction	Neutrons Produced per gram-second	
			Spontaneous Fission ^a	Total
^{235}U	$7.038 \pm 0.005 \times 10^8$	$7.21 \pm 0.72 \times 10^{-4}$	$3.86 \pm 0.99 \times 10^{-4}$	$1.11 \pm 0.12 \times 10^{-3}$
^{238}U	$4.4683 \pm 0.0024 \times 10^9$	$8.43 \pm 0.84 \times 10^{-5}$	$1.36 \pm 0.02 \times 10^{-2}$	$1.36 \pm 0.02 \times 10^{-2}$
^{238}Pu	87.71 ± 0.03	$1.56 \pm 0.16 \times 10^4$	$2.60 \pm 0.11 \times 10^3$	$1.82 \pm 0.16 \times 10^4$
^{239}Pu	$2.4131 \pm 0.0016 \times 10^4$	$4.25 \pm 0.43 \times 10^1$		$4.25 \pm 0.43 \times 10^1$
^{240}Pu	$6.570 \pm 0.006 \times 10^3$	$1.56 \pm 0.16 \times 10^2$	$8.85 \pm 0.10 \times 10^2$	$1.04 \pm 0.19 \times 10^3$
^{242}Pu	$3.763 \pm 0.009 \times 10^5$	2.27 ± 0.23	$1.743 \pm 0.015 \times 10^3$	$1.743 \pm 0.015 \times 10^3$
^{241}Am	432.0 ± 0.2	$3.17 \pm 0.32 \times 10^3$		$3.17 \pm 0.32 \times 10^3$
^{242}Cm	0.4456 ± 0.0001	$4.48 \pm 0.45 \times 10^6$	$2.25 \pm 0.08 \times 10^7$	$2.70 \pm 0.09 \times 10^7$
^{244}Cm	18.099 ± 0.015	$8.82 \pm 0.88 \times 10^4$	$1.081 \pm 0.007 \times 10^7$	$1.090 \pm 0.007 \times 10^7$

^aReference value for spontaneous values.

$\bar{\nu}_{\text{sp}} = 3.756$ for ^{252}Cf .

The unique characteristic of the LMFBR system that relates to the verification of nuclear-material flow in core assemblies is the almost constant plutonium concentration in reactors with core conversion ratios of approximately 0.7 and greater. Although the relative concentrations of the plutonium isotopes may vary over extended periods of burnup, the SF neutron yields from the plutonium are almost constant over an irradiation period extending to about 100,000 MWd/T. Table III gives the SF neutron yields

for a reference core assembly containing approximately 35 kg of heavy metal. The variation is within 10% between the nonirradiated and irradiated fuel over this burnup range, and the apparent slight increase is found to be linear within an error of 0.3%.

TABLE III. Spontaneous Fission Neutron Yield
Reference (35 kg HM) Core Assembly
(10^6 Neutrons/s)

Average Burnup (MWd/T)	Plutonium				Curium	
	238	240	242	Total	^{242}Cm ($t_c = 0$)	^{244}Cm
0	0.18	1.2	0.30	1.68	-	-
12,000	0.17	1.23	0.30	1.72	1.51	0.30
35,000	0.16	1.29	0.33	1.78	12.38	2.69
77,000	0.13	1.39	0.36	1.87	58.63	14.12
96,000	0.12	1.42	0.36	1.91	70.74	22.90

^aHalf-life = 162.8 days

The significance of the neutron source strength is that the curium-isotope concentrations can be determined from a difference measurement of SF neutron source strengths

between the fresh fuel and the spent fuel in a given assembly. The resulting curium concentration gives a direct measure of the irradiation history of the assembly. This difference-method of determining the curium concentration has the advantage of allowing use of the time decay of ^{242}Cm as a consistency check in the physical-inventory verification of the stored assemblies. The high concentration of ^{242}Cm at high burnups is unique to LMFBRs, and presents the opportunity for the inspector to make a timely in-the-field assessment as to a possible anomaly in the flow of nuclear material in the assembly handling operations of the reactor.

Figure 3 indicates that the plutonium should dominate the SF neutron yield rate over the burnup periods anticipated for the blanket assemblies in all classes of reactor systems.

Preliminary results of isotopic content experiments from the reprocessed fast breeder fuels of the UKAEA Dounraey Prototype Fast Breeder reactor (PFR)⁷, indicate that for spent fuels at the burnup level of about 70,000 MWd/T, and a decay cooling period of 180 days, the plutonium isotopes contributed 7% of the S.F. neutrons measured, with the remaining 93% being those of the curium isotopes. Additional total neutron emission measurements made at Dounraey on a single irradiated PFR pin, having a burnup history of 65,000 MWd/T and a cooling period of approximately 3 years, showed that 70% of the neutrons were emitted by the curium isotopes, and the remaining 30% (including some contribution from α, n reactions) were emitted by the plutonium isotopes. These experimental results outlined in Table IV, corroborate the computed estimates for the reference LMFBR and appear to give a consistent correlation between the isotopics and neutron emissions, including the time decay of neutron yields due to the time decay of the ^{242}Cm isotope.

The HLNCC measurements are of major safeguards importance in verifying plutonium flow accountancy data well within the time period before the reprocessing operation. The HLNCC systems would have to be modified in order to minimize the interference of the high gamma-ray radiation background of the spent fuel. This may be done through selection of neutron detectors and/or interspersing a gamma-ray attenuation material such as lead between the assembly and the detection system. The placement and operation of the HLNCC measurement system at key measurement points are plant-design-specific, and modifications in designs would be required to accommodate the specific plant features.

TABLE IV. Isotopic Neutron Yields
Experiments (PFR) and Computations (Ref. LMFBR)

Reprocessed PFR Assembly: 70,000 MWd/T (5th cycle), 180 days decay cooling period		
Total Neutron Yields		
Experiment (PFR)	Computed (Ref. LMFBR)	
7% Pu 93% Cm	12% Pu 88% Cm	
Reprocessed Single PFR Fuel Pin: 65,000 MWd/T, 3 year decay cooling period		
Total Neutron Yields		
Experiment (PFR)	Computed (Ref. LMFBR)	
	(19 w/o Pu Fuel)	(27 w/o Pu Fuel)
30% Pu 70% Cm	27% Pu 73% Cm	40% Pu 60% Cm

3. Active-well Neutron-interrogation System (AWCC)

An IAEA safeguards measurement of complementary importance to the HLNCC measurements is the direct determination of total fissile Pu/²³⁵U concentrations in the fresh and spent fuel assembly. For verification of material flow and physical inventory, the induced fission neutron rate measurements can be obtained by using an external neutron source of sufficient strength to effect a neutron multiplication above the total neutron yield rate. Fissile content in the assemblies may be determined by the AWCC system. The ²⁴¹Am-Li source system may be modified for use on spent fuel assemblies and designed for use as a dual purpose HLNCC.

D. Diversion Analysis

A diversion analysis study involving the substitution of 10% (22 of 217 fuel rods) and 20% (59 of 217 fuel rods) of the central rods in a fuel assembly with depleted uranium rods was performed to explore the sensitivity to detect the substitution by both the passive and active modes of the HLNCC methods. The computational model for the detector assumed cylindrical geometry of the HLNCC with a polyethylene detector region within which are placed the detectors (³He or fission chambers). The detector radius was maintained throughout the series of computations to avoid geometric effects on the flux at the detector. The model for the case of using the HLNCC in the active mode assumed the external neutron source to be cylindrical and uniformly distributed with zero radial extent, and positioned between the fuel assembly and detector region. The results of the scoping neutron transport computations for the passive and the active mode (with an external neutron source strength of 1.23×10^7 n/sec) on an LMFBR fuel assembly are presented in Table V. The fuel assembly composition was that of a spent fuel assembly having undergone an average 40,000 MWd/t.

TABLE V. Diversion Analysis Neutron Interrogation Sensitivity

PASSIVE NEUTRONS					
% Fuel Diversion	Total Source (n/sec)	Induced Fission (n/sec)	M	Total Flux at the Detector (n/sec-cm ²)	% Reduction in Detector Flux
0	8.237E+6	1.277E+6	1.155	5.066E+3	0
10.0	7.413E+6	1.025E+6	1.138	4.520E+3	10.8
26.2	6.084E+6	6.9595E+5	1.114	3.653E+3	27.7

EXTERNAL NEUTRON SOURCE STRENGTH (1.232×10^7 n/sec)					
% Fuel Diversion	Total Source (n/sec)	Induced Fission (n/sec)	M	^a Total Flux at the Detector (n/sec-cm ²)	% Reduction in Detector Flux
0	2.055E+7	3.590E+6	1.175	1.250E+4	0
10.0	1.973E+7	3.206E+6	1.163	1.193E+4	4.2
26.2	1.840E+7	2.650E+6	1.129	1.114E+4	11.5

^aExternal source background has been subtracted.

In the case of the passive HLNCC mode, the percent change in the induced fission and the total flux entering the detector regions indicate that the multiplication decreases as more fuel is removed. This multiplication effect results in a higher percentage decrease in the induced fission reactions than the change in the fuel and internal source. The flux at the detector changes by a slightly higher percentage than the change in the internal source. The major fraction of this change in the induced fission source is due to the reduction in the fuel region. Since the internal source overwhelms the induced fission, the change in the flux at the detector is more representative of the change in the internal source. The flux levels predicted at the detector indicate that a 10% fuel substitution or less can be detected if the initial flux level is known.

The introduction of a 0.3 MeV energy spectra external neutron source is to determine the fissile content. The low energy neutron source is selected to avoid the fast fission effect in ²³⁸U. The external total source strength of 1.23×10^7 n/sec increases the induced fission component by a factor of 3 and increases the flux at the detector by a factor of 3 and increases the flux at the detector by a factor greater than 2. The percent reduction of the induced fission follows the percent change of the fuel substitution. However, the respective change in the neutron flux of only 4.2 and 11.5% are to be expected at the detector region. This results from several considerations: a) the induced fission in the substituted fuel region is from the depleted uranium fast fission, and the specific induced fission radial profile is somewhat flatter in the center than at the outer edges of the fuel region. It appears that fuel substitutions in the order of magnitude used in this study may be detected using the HLNCC in the active mode, although not as readily as in the passive mode.

III. SUMMARY REMARKS

The need for IAEA safeguards systems to be applied to LMFBRs is near-term (a decade or so). Present programs to develop devices and techniques for improved safeguards do not address these near-term needs. The assessment for inspection requirements emphasizes those safeguards measures that are currently available or may be available for field implementation by the Agency within the next 3 to 5 years. At the present time, the neutron coincidence counting (NCC) system can provide some verification of the nuclear-material content in the fuel and blanket assembly, but further development will be necessary before it can be applied as a material balance accountancy measurement.

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