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NUCLEAR MATERIAL SAFEGUARDS SURVEILLANCE AND ACCOUNTANCY
BY ISOTOPE-CORRELATION TECHNIQUES

by

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ABSTRACT

The purpose of this study is to investigate the applicability of isotope correlation techniques (ICT) to the Light Water Reactor (LWR) and the Liquid Metal Fast Breeder Reactor (LMFBR) fuel cycles for nuclear material accountancy and safeguards surveillance. The isotopic measurement of the inventory input to the reprocessing phase of the fuel cycle is the primary direct determination that an anomaly may exist in the fuel management of nuclear material. The nuclear materials accountancy gap which exists between the fabrication plant output and the input to the reprocessing plant can be minimized by using ICT at the dissolver stage of the reprocessing plant. The ICT allows a level of verification of the fabricator's fuel content specifications, the irradiation history, the fuel and blanket assemblies management and scheduling within the reactor, and the subsequent spent fuel assembly flows to the reprocessing plant. The investigation indicates that there exist relationships between isotopic concentration which have predictable, functional behavior over a range of burnup. Several cross-correlations serve to establish the initial core assembly-averaged composition. The selection of the more effective functionals will depend not only on the level of reliability of ICT for verification, but also on the capability, accuracy and difficulty of developing measurement methods. The propagation of measurement errors on the correlation functions and respective sensitivities to isotopic compositional changes have been examined and found to be consistent with current measurement methods.

I. INTRODUCTION

The general purpose of this study program is to investigate the applicability of isotope correlation techniques (ICT) to the Light Water Reactor (LWR) and the Liquid Metal Fast Breeder Reactor (LMFBR) fuel cycles for nuclear material safeguards surveillance and accountancy. The similarities in reactor designs and operating modes of the worldwide deployed LWRs (~ hundreds) and LMFBRs (~ ten) allows the ICT to be used in principle as a universal measure of safeguards. To reflect the mutually important complementary roles of isotopic measurements in maintaining continuity of knowledge in the history of the nuclear material flow prior to dissolution and in establishing reference measurement points for material balance accountancy, the distinction between ICT for safeguards surveillance and accountancy is emphasized.

The safeguards significance of the ICT has the following attributes for nuclear material surveillance and accountancy: establishes reactor irradiation history, indicates necessary measurement technology at the dissolver and predissolver stage, and identifies necessary error limits in support of instrumentation designs and measurement methods. The surveillance safeguards aspect relates to: identifying anomalies in reactor loading and unloading fuel management programs; confirming the operating history of the reactor, identifying the development needs of sensing devices, and maintaining an overview of the expected operational substages in the fuel cycle. The accounting aspect of ICT measurements for safeguards includes: measurements which may be used in close-out material balances, and measurements which provide for an independent near-real-time assessment approach to account for nuclear materials.

II. SAFEGUARDS SIGNIFICANCE

The feasibility of utilizing fission product and heavy element isotopic ratios has been explored by many investigators¹⁻⁶ for verifying burnup of fissile material input to a reprocessing plant for the LWR fuel assemblies. The heavy element isotopic correlations may be employed for the independent verification and evaluation of the Pu/U ratio measurement by the use of wet chemistry and isotope dilution mass spectroscopy, and to establish that the integrity of the fuel content has been maintained from the output of the fuel fabricator plant and the input to the reprocessing plant.

The changes in the isotopic composition of a fuel assembly with burnup are a measure of the irradiation history of the fuel in a power reactor. Initial experimental data on LWR reprocessed fuel indicates that there may exist relationships between isotopic concentrations which have a predictable, functional behavior over a range of reactor operating conditions and burnup. However, the range of burnups in the reprocessing data are found to be limited at very low burnups (<10,000 MWD/T) and very high burnups (>25,000 MWD/T). The validity of extrapolating the correlation functions much beyond the range of the burnup data bank is uncertain and should be established by computation and confirmed with precisely tailored experiments. In part, preliminary efforts in this direction have been initiated in Europe^{7,8,9,10}. This paper presents the initial phase of the U.S. study program and involves the computation of isotopic correlations for an LMFBR reference fuel cycle.¹¹ The LWR fuel cycle phase of the study is currently in progress.

The selection of the more safeguards effective functionals will depend not only on the level of reliability of ICT for verification, but also on the capability and difficulty of developing measurement methods. Performance characteristics of existing and proposed isotopic measurement techniques cover the general areas including assay and isotopic wet chemistry and NDA:

(1) simultaneous multicomponent analysis techniques, (2) mass spectrometry, (3) x-ray fluorescence or densitometry with high flux monochromatic x-ray sources and high dispersion spectrometers, (4) passive and active neutron interrogation, (5) high level gamma-ray spectroscopy, (6) coulometry, and (7) potentiometry.

The accuracy and precision of the measurement methods may not be emphasized for this level of assessment since some of the methods are still being developed. However, a need exists to evaluate/improve measurement technology to serve the Isotope Correlation Surveillance and Accountability Methods (ICSAM) activity for safeguards.

Many proposed measurements involving integral-type facilities, such as critical facilities for reactivity measurements on spent fuel assemblies, or neutron source systems for nondestructive assay of total fissile content in irradiated fuel assemblies have not been included in this study. Although these systems could be utilized for "signature-type" accountability, the integral measures are limited in meeting the objectives of the ICT program.

A. INTERNATIONAL AND DOMESTIC SAFEGUARDS AND MANAGEMENT OF NUCLEAR MATERIALS

Isotope correlations coupled with measurement techniques have the potential as a safeguards surveillance and accountability measure in the dissolver stage of the nuclear fuel cycle. Throughout the major portion of the fuel cycle, the fissile surveillance and accountability measure is basically in the form of item accountability (fuel powder containers, pellets, elements, and assemblies). The measurements of the nuclear material input and product streams in the reprocessing phase of the cycle is the primary direct verification of fissile material flow subsequent to reactor operation. The nuclear materials assay measurement gap which exists between the fabrication plant output and the input to the reprocessing plant can be minimized by the utilization of Isotope Correlation Techniques (ICT) at the dissolver stage of the processing plant. The safeguards significance of the ICT is that the input accountability would allow a level of verification of the fabricator's uranium content specification, the reactor irradiation history, the fuel and blanket assemblies management and scheduling within the reactor, and the subsequent spent fuel assembly flow to the reprocessing plant.

III. THE BASIS FOR ISOTOPE CORRELATIONS

From the study of many functionals,¹¹ several of the isotopic relationships of interest found to exhibit a reasonably monotonic behavior over a broad range of reactor conditions and burnup were such functions as: Pu/U vs. depletion ^{235}U , Pu/U vs. $(100 - ^{239}Pu)$, Pu/U vs. $^{239}Pu \times ^{242}Pu / ^{240}Pu^2$, and ^{236}U vs. ^{235}U . Of the many preliminary functionals that have been suggested, the most effective functions are those having a linear or almost linear behavior. These linear relationships being independent of reactor operating conditions and burnup, effect a means of verifying the input to a reprocessing plant, and methods for establishing internal consistency of input analytical measurements, and a level of verification on initial isotopic concentrations prior to burnup.

A. REACTOR SYSTEM

The reference LMFBR was studied to gain an R&D insight in applying ICT to fast breeder reactor technology as part of the International and Domestic requirements in developing a safeguards systems for the currently and near-term operating fast breeder reactors.¹² For computational purposes the core is divided in azimuthally symmetric 1/6 segments and radially zoned into five concentric regions or columns surrounding a central column, as shown in Fig. 1.

Fig. 1. Core and Radial Blanket Refueling Scheme

The central column, is a control rod; columns two, three, and four comprise the inner core assemblies; column five, the outer core assemblies; and column six, the radial blanket. The initial compositions of the assemblies in the two respective core regions, inner (low enrichment) and outer (high enrichment), and the blanket regions are contained in Table I. The core compositions are based based on the LMFBR integral-assembly design of fuel elements and depleted-uranium axial blanket sections.

TABLE I. Initial Composition by Column

	Column 2, 3, 4	Column 5	Column 6
Pu/U (g/te)	1.32×10^5	2.04×10^5	0.0
^{235}U (w/o)	0.447	0.432	0.217
^{236}U (w/o)	0.0	0.0	0.0
^{238}U (w/o)	99.6	99.6	99.8
^{238}Pu (w/o)	1.00	1.00	0.0
^{239}Pu (w/o)	67.3	67.3	0.0
^{240}Pu (w/o)	19.2	19.2	0.0
^{241}Pu (w/o)	10.1	10.1	0.0
^{242}Pu (w/o)	2.40	2.4	0.0

The units for all of the graphs are weight percent of the isotope compared to the total weight of that element except for Pu/U which is in grams of plutonium per metric ton of uranium.

^{235}D represents the depletion of

^{235}U or the difference between the concentration at a given burnup and the initial unirradiated concentration. Similarly, $\text{D}(^{235}\text{U}^2)$ and $\text{D}(^{235}\text{U} \times ^{239}\text{Pu}^2)$ are the difference between a given burnup and the beginning of the equilibrium cycle (BOEC).

IV. USE AND IMPLEMENTATION OF ICT

A. DISSOLVER STAGE

The isotopic correlation functions of interest for this report are presented in Figs. 2a and 2b. The points on the graphs are the program output of the functions and are numbered with increasing burnup. In the lower right hand corner of the graph, an arrow is included to indicate the general direction of burnup. Also noted are the reactor region identification and the curve numbers that correspond to the specific correlation functions listed in Table II. In the set of functions for simulating substitutions, the graphs list the normal and the changed end-values of the ordinate and abscissa, and the percentage difference between the two for each of the axis variables.

TABLE II. Isotopic Correlation Functions

-
1. Pu/U vs. D-235
 2. Pu/U vs. 100-239Pu
 3. 236U vs. 235U
 4. 239Pu x 2 vs. 235U
 5. 240Pu vs. 239Pu(100-239Pu)
 6. 239Pu x 240Pu vs. 235U x 241Pu
 7. 240Pu x 241Pu vs. 235U x 242Pu
 8. 240Pu vs. D(235U x 2)
 9. 239Pu x 2(100-239Pu)/235U x 2 vs. 100-239Pu
 10. (239Pu x 2) (240Pu x 2) vs. 240Pu
 11. 239Pu(100-239Pu) vs. D(235U x (239Pu x 2))
 12. Pu/U vs. 239Pu x 242Pu/(240Pu x 2)
 13. Pu/U vs. 241Pu/240Pu
 14. Pu/U vs. (241Pu + 242Pu)/240Pu
-

The final point on the graph will be that measured at the reprocessing plant. The position of the measured correlation point when compared with the expected position of the correlation function for a specific assembly batch, may be utilized to detect inconsistencies in nuclear material management within some margin of error. The margin of error is influenced by the measurement accuracy and precision and the propagation of these measurement errors for the specific correlation function.

There are several cross-correlations that may serve, to establish the initial core assembly-averaged composition. Referring to Fig. 2a, the functional relationship

curve-3 (236U vs. 235U) indicates that for the core assemblies the correlation behaves in a linear manner. The slope can be used to extrapolate to the preirradiation condition from the measured point. This initial determination of 235U concentration may be used as a known quantity in the other functionals.

The correlation function 240Pu vs. D(235U²), curve 8, is found to be a well-behaved function and may be used to verify curve-3 and consequently provide the initial 240Pu concentration. This would use the value for 235U from curve-3 since the initial 235U enrichment is necessary to calculate the depletion. With the initial 240Pu determined in this manner, curve-10 239Pu² x 240Pu² vs. 240Pu may then be used to verify the initial 239Pu concentration. These three values may now be applied to curve-6 (239Pu x 240Pu vs. 235U x 241Pu), and curve-7 (240Pu x 241Pu vs. 235U x 242Pu), to obtain verification of 241Pu and 242Pu concentrations. The significance of this procedure is that this cross-correlation adds a constraint of consistency which in turn may be used to identify anomalies or unexpected variations in the isotopic compositions of the assemblies. The existence of the many monotonically well-behaved functions offers the possibility of identifying several patterns of cross-correlations.

Fig. 2a. Isotope Correlation Functions

Fig. 2b. Isotope Correlation Functions

Referring to Fig. 2b curve 1, Pu/U vs. ^{235}D , can provide the original plutonium enrichment which distinguishes the inner and outer core assemblies. This again would use the value for ^{235}U from curve-3. Several other correlations are also found to be well-behaved functionals and may be used in cross-correlating the plutonium isotopes.

Detailed studies indicate that the change in the Pu/U ratio as a function of ^{235}D is plutonium enrichment dependent. The Pu/U ratio increases by about 11% in the inner (low enrichment) core assemblies and decreases by about 8% in the outer (high enrichment) core assemblies (see Reference 11) for the burnup range considered in this study. Consequently the intermixing of the two differently enriched assemblies should be associated. This implies that safeguards requirements may influence, in part, fuel reprocessing management programs. However, it may develop that in a preplanned intermixing of different rows of assemblies in a reprocessing program would also yield well-behaved functionals which could be programmed for cross-correlations.

The isotopic correlations studied for the radial blanket assemblies were found to be linear functions or extremely well-behaved functions over the 6-year burnup period. There should be a strong incentive to process the radial blanket assemblies separately from the core assemblies. With the adoption of this reprocessing program, the potential exists to enhance independent verification of nuclear material flow between the fabrication plant output and the reactor operational phase of the fuel cycle.

B. PREDISSOLVER STAGE

Isotope correlations and neutron assay methods have been investigated and implemented for determining plutonium and burnup in fresh and spent LMFBR and in spent LWR fuel assemblies.^{13,14} The methods are based on active and passive neutron coincidence counting (NCC) techniques. Preliminary studies^{15,16} on neutron yield rates indicates that as a consequence of the high plutonium concentration level throughout the fuel irradiation period in an LMFBR, 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. This is a significant result when compared to the LWR spent fuel interrogation problems and experience where the 244-curium neutron yield rate is found to be several orders of magnitude greater than the plutonium yield rate at the same level of burnup and after the decay of the 242-curium contribution.

Preliminary results of isotopic content experiments from the reprocessed fast breeder fuels of the UKAEA Dounraey Prototype Fast Breeder reactor (PFR)¹⁷, indicated 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 corroborate the computed estimates for the reference LMFBR^{15,16}, outlined in Table III, 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-curium isotope.

TABLE III. Isotopic Neutron Yields Experiments
vs Computations (ISPO Task C.43)¹⁶

Reprocessed PFR Assembly: 70,000 MWd/T (5th cycle),
180 days decay cooling period

Total Neutron Yields	
Experiment	Computed (ISPO Task C.43)
7% Pu	12% Pu
93% Cm	88% Cm

Reprocessed Single PFR Fuel Pin: 65,000 MWd/T,
3 year decay cooling period

Total Neutron Yields		
Experiment	Computed (ISPO Task C.43)	
	(19 w/o Pu Fuel)	(27 w/o Pu Fuel)
30% Pu	27% Pu	40% Pu
70% Cm	73% Cm	60% Cm

Although the relative concentrations of the plutonium isotopes, may vary over extended periods of burnup and decay cooling times, the spontaneous fission neutron yield from the plutonium is found to be almost constant. The determination of the curium isotope concentrations is made by a difference measurement of spontaneous fission source strengths between the fresh and spent fuel for a given assembly. Consequently the NDA measurements are considered a primary safeguards technique to be developed for the LMFBR and LWR fuel cycles.

V. SENSITIVITY OF ICT TO ANOMALIES IN NUCLEAR MATERIAL FLOW

To scope the sensitivity of the isotope correlation functions, a substitution or dilution equivalent of one assembly from the radial blanket for core assemblies (TC-2) in a seven assembly dissolver batch was simulated (15% dilution). The dilution may represent the partial interchange of the fuel pins in the assemblies. All assemblies were assumed to have completed their final burnup stages. Representative changes in the correlation functions end-points (outliers) are presented in Figs. 3a and 3b. Most of the correlation functions were found to be very sensitive to this type of substitution.

The above scheme of substitution or dilution was extended to include the case of one radial blanket assembly substitution in a twelve core assembly reprocessing batch (8% dilution). The results, included in Figs. 3 a,b and Fig. 4, indicate that the TC-2 end-value differences (outliers) are approximately half those of the previous case. There appear to be many correlations with large enough differences to detect substitution, with six of the correlation functions having changes between 7 and 15% and two of the correlation functions having changes greater than 15%.

The interpretation of these changes in the correlation function end-values (outliers) will depend on the propagation of measurement errors. The selection of the more safeguards effective functionals will depend on the level of reliability of the correlation function for verification, and on the capability and difficulty of developing measurement methods.

	NO SUBST	1/7 SUBST	% DIFF	1/12 SUBST	% DIFF	TC - 2
X	5.876×10^{-2}	4.657×10^{-2}	-2.076×10^1	5.159×10^{-2}	-1.220×10^1	3
Y	2.061×10^{-1}	1.903×10^{-1}	-7.662×10^0	1.969×10^{-1}	-4.503×10^0	0.5 %

	NO SUBST	1/7 SUBST	% DIFF	1/12 SUBST	% DIFF	TC - 2
X	1.574×10^{-1}	1.637×10^{-1}	3.978×10^0	1.612×10^{-1}	2.576×10^0	8
Y	2.449×10^1	2.273×10^1	-7.203×10^0	2.357×10^1	-3.787×10^0	0.5 %

	NO SUBST	1/7 SUBST	% DIFF	1/12 SUBST	% DIFF	TC - 2
X	2.449×10^1	2.273×10^1	-7.203×10^0	2.357×10^1	-3.787×10^0	10
Y	2.658×10^6	2.471×10^6	-7.054×10^0	2.562×10^6	-3.604×10^0	0.5 %

Fig. 3a. Measurement Errors (0.5%) Propagation on Isotope Correlation Functions

	NO SUBST	1/7 SUBST	% DIFF	1/12 SUBST	% DIFF	TC - 2
X	5.876×10^{-2}	4.657×10^{-2}	-2.076×10^1	5.159×10^{-2}	-1.220×10^1	3
Y	2.041×10^{-1}	1.903×10^{-1}	-7.662×10^0	1.969×10^{-1}	-4.503×10^0	2.0 %

	NO SUBST	1/7 SUBST	% DIFF	1/12 SUBST	% DIFF	TC - 2
X	1.574×10^{-1}	1.637×10^{-1}	3.978×10^0	1.612×10^{-1}	2.376×10^0	8
Y	2.449×10^1	2.273×10^1	-7.203×10^0	2.357×10^1	-5.787×10^0	2.0 %

	NO SUBST	1/7 SUBST	% DIFF	1/12 SUBST	% DIFF	TC - 2
X	2.440×10^1	2.273×10^1	-7.203×10^0	2.357×10^1	-5.787×10^0	10
Y	2.658×10^0	2.471×10^0	-7.054×10^0	2.562×10^0	-3.604×10^0	2.0 %

Fig. 3b. Measurement Errors (2.0%) Propagation on Isotope Correlation Functions

Fig. 4.
Isotopic Correlation
Function, Pu/U vs. ^{235}D

	NO SUBST.	1/7 SUBST.	% DIFF.	1/12 SUBST.	% DIFF.	TC - 2
X	2.410×10^{-1}	2.568×10^{-1}	6.554×10^0	2.503×10^{-1}	3.852×10^0	1
Y	1.464×10^{-5}	1.137×10^{-5}	-2.234×10^1	1.271×10^{-5}	-1313×10^1	0.5%

VI. MEASUREMENT METHODS FOR ICSAM

A. WET CHEMISTRY SYSTEM

The most widely used analytical method to measure Pu, U and/or its isotopes in nuclear materials includes the following:

- a) Isotope Dilution Mass Spectrometry (IDMS)
 - filament loading
 - resin bead loading
- b) Controlled potential coulometry
- c) Potentiometric titrimetry
- d) Optical Spectroscopy using inductively-coupled plasma (ICP)

1. Mass Spectrometry Filament Loading

The advantage of the mass spectrometry isotope dilution method technique is that micro quantities of a sample containing spiked isotopes e.g., Pu-244, U-233, are used on a filament of a mass spectrometer to obtain

total Pu, U and their isotopes. Some problems exist with this technology if the Pu loaded on the filament has some U or if the U loaded on the filament has some Pu. Mass spectrometry resolution is not capable of distinguishing between Pu-238 and U-238 masses. Therefore, care has to be exercised to separate U from Pu to load respective elements on filaments of the mass spectrometer.

2. Mass Spectrometry Resin Bead Loading

To minimize or eliminate contamination of Pu-238 with U-238 using mass spectrometry, a resin bead sampling technique has been developed.¹⁸ A Dowex anion exchange bead selectively adsorbs and concentrates, in defined quantities, Pu and U from a nitric acid solution. The dried bead containing both the sample and spike is mounted on a filament of a spectrometer and the temperature of the bead is elevated. At about 1400°C, Pu is selectively volatilized off the bead and measured. Subsequently, the temperature of the bead is raised to about 1800°C, and the U is selectively volatilized and measured. The added advantage of the resin bead technology is that subnanogram to nanogram levels of Pu and U can readily be transported on the bead between labs and countries with essentially no health hazards to shippers or operators of instruments.

3. Coulometry

The use of classical wet chemistry techniques to analytically measure elemental Pu and U in characterized material has now become a relatively routine methodology. Manual as well as automated equipment is being employed throughout the nuclear community for wet chemical analyses of Pu and U. The two more extensively used wet analytical techniques to assay Pu and U are: a) Controlled-potential coulometry,^{19,20} and b) Potentiometric titrimetry.²¹ The former method is based on the quantitative oxidation or reduction of Pu and U using controlled conditions. The number of coulombs required is a measure of the number of electro-chemical equivalents present of the element in question.

4. Potentiometry

The potentiometric method²¹ utilizes the change in the ratio of the concentration of oxidized to reduced species on addition of an appropriate reagent. The change of voltage is monitored until a large potential break, which occurs at the equivalence point is achieved. The concentration of Pu or U is obtained by the measure of the amount of titrant needed to reach the equivalence point.

5. Optical Spectroscopy

An analytical technique being developed is based on the use of emission optical spectroscopy to simultaneously measure U, Pu and their isotopes at the dissolver stage of the fuel cycle. This in-line approach employs an Inductively Coupled Plasma (ICP)²² to excite the liquid sample and a high resolution instrument to resolve the spectral lines of Pu, U and their isotopes. The intent is to assay Pu, U and their isotopes simultaneously to obtain real time accountability and surveillance of nuclear materials. Preliminary results have been obtained for U and its isotopes using a simulated

unirradiated dissolver solution without Pu. A high resolution optical spectrometer is capable of resolving the isotopic structure of spectral lines emitted by a uranium solution that had been atomized and excited using an inductively coupled plasma. Isotopic ratios measured optically over two orders of magnitude exhibit a linear calibration curve with no detectable bias. The application of this analytical technique to nuclear safeguards

will be tested subsequently using plutonium solutions to simultaneously assay total U, Pu and their isotopes. This will achieve near-real-time surveillance and accountability of nuclide material. A summary of wet analytical chemistry measurement techniques are listed in Table IV.

TABLE IV. Wet Analytical Chemistry Measurement Techniques

Method	Status	Accuracy for Pu, U
Isotope Dilution Mass Spectroscopy (IDMS) using filament loading	Developed	<0.5% for assay and isotopes
IDMS using Resin bead collection technique	Developed	<0.5% for assay and isotopes
Controlled-Potential Coulometry	Developed	About 0.1% or better for Pu
Potentiometric Titrimetry	Developed	About 0.1% or better for U
Optical Spectroscopy (Inductively Coupled Plasma)	Under Development	About 1% for assay and isotopes

B. NDA TECHNOLOGY

The safeguards approach in the long-term handling and interim storage of fresh and spent fuel assemblies, dissolver solutions, and the transfer of ownership of the fuel and the resolution of shipper/receiver balances, indicates a need for direct verification of the integrity of fissile content. The safeguards approach in the fuel handling system will involve a combination of sensors and inspection plans supplemented by NDA techniques.

NDA technology being developed includes the following components:

- a) Gamma-Ray Spectrometry techniques,
- b) Neutron coincidence counting techniques (NCC) to assay nuclear material fresh fuel assemblies,
- c) Active neutron coincidence counting techniques to assay nuclear materials in spent fuel assemblies,
- d) Wavelength dispersive x-ray fluorescence,
- e) X-Ray Absorption Edge Densitometry.

The currently available NDA techniques for assembly accountability can only distinguish between different classes of assemblies, i.e. fuel, blanket and/or dummy replacement. There is no distinction made between the individual assemblies. It is not apparent how effective NDA techniques may be if this information is not supplemented by subsequent confirmatory information that a replacement or partial replacement has occurred. An added level of consistency in flow verification and nuclear material accountancy may be effected if the difference in isotopic-radiation measurements reflect the irradiation history of a specifically identified assembly.

The NDA safeguards instrumentation considerations for fuel assembly accountability and verification of the integrity of nuclear material content

have included assessing the implementation of the High Resolution Gamma Spectroscopy (HRGS) for the passive gamma-ray spectrometry of uranium and plutonium; the High Level Neutron Coincidence Counter (NCC) for the passive measurement of spontaneous fission neutrons from plutonium, and the active well neutron interrogation systems (AWCC) for the measure of induced fission neutrons in the fissile isotopes.

1. Passive Gamma-Ray Assay (HRGS)

The uranium and plutonium assay by the HRGS system²³ may be applicable for individual fresh fuel elements of an assembly and in a more qualified manner possibly for total fuel assemblies of both LWRs and LMFBFRs. The self-shielding effects in the fuel elements is expected to be minimal and consequently the gamma-ray spectra may be used as a quantitative measure of the uranium and plutonium content of the assembly. The self-attenuation of the gamma-rays in a total assembly would compromise the quantitative measure of the nuclear material content in the fresh fuel assembly. However, for purposes other than plutonium content determinations, gamma-ray spectroscopy may be found useful in certain assembly identification modes.

The direct assay of plutonium in an irradiated assembly by gamma-ray spectroscopy does not appear to be an effective measurement technique except to establish the existence of fission-products.

2. Passive Neutron Assay (NCC)

Preliminary studies on neutron yield rates from the spontaneous fission of the 238-plutonium, 240-plutonium and the 242-plutonium isotopes have indicated that the NCC system^{13,14,15} 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. Of major safeguards importance in attaining these NCC measurements is in establishing the verification of the plutonium flow accountancy in an NDA mode well within the time period prior to the reprocessing operation. The modification in the design of the NCC systems would have to minimize the interaction of the high gamma-ray radiation background of the spent fuel.

3. Active Neutron Interrogation (AWCC)

A safeguards measure of complementary importance is the direct determination of plutonium/235 uranium concentrations in the fresh and spent fuel assembly. The measure of interest for verification of material-flow and physical inventory is the induced fission neutrons obtained by using an external neutron source of sufficient strength to effect a neutron multiplication above the total neutron yield rate. The measure of fissile content in the assemblies may be determined by the AWCC system.²⁴ The ²⁴¹Am-Li source system may be redesigned for use on spent fuel assemblies and made consistent for use as a dual purpose NCC.

The determination of the fissile plutonium/235-uranium content of the fuel assembly before and after irradiation is also a surveillance measure for maintaining the integrity of item flow and physical inventory accounting of assemblies.

4. Wavelength Dispersive X-Ray Fluorescence

An in-line NDA wavelength dispersive x-ray fluorescence technique²⁵ is being developed to assay Pu and U in dissolver solution. As a first step approach a continuous "white light" technique is being evaluated to excite and analytically measure fluorescent Pu and U and intensities. Additional work is contemplated in the x-ray area to improve signal-to-noise ratio for Pu and U assay and includes the use of wavelength monochromatic radiation.

5. X-Ray Absorption Edge Densitometry

X-ray absorption edge densitometry can be used to measure uranium in LWR and the uranium and plutonium LMFBR solutions.²⁶ No sample treatment is required even with fission product activity of several hundred curies per liter.

A summary of NDA measurement techniques are included in Table V.

TABLE V. NDA Measurement Techniques

Method	Status	Accuracy for Pu, U
High Resolution Gamma Ray Spectroscopy (HRGS)	Developed for Fuel Assemblies (SAM/BSAM)	Qualitative measure of fission product gamma-ray radiation
High Level Neutron Coincident Counter (NCC) Passive Mode	Developed for Unirradiated fuel Assemblies	1 to 2% in Pu assay for a 10 minute counting time
Active Well High Level Neutron Coincidence Counter (AWCC)	Under Development for Fuel Assemblies	To be determined
Wavelength Dispersion Spectrometry (WDS) XRF-WDS	Under Development	~ 1% expected for a 2-5 minute counting time
X-Ray Absorption Edge Densitometry	Under Development	~ 0.2% for uranium 0.7% for plutonium

VII. IMPACT OF ICT REQUIREMENTS ON INSTRUMENTATION ERROR DESIGN CRITERIA AND MEASUREMENT METHODOLOGY

The two aspects of the ICT and related measurement methods as referred to in this study maintains the distinction between measurements for nuclear material accounting in the sense of Inventory Differences, and NDA and wet chemistry measurements for the verification of the integrity of the nuclear material content. This latter mode of utilizing ICT and related measurements relegates the accuracy of measurements to a lower critical level for safeguards surveillance or verification of nuclear material content and/or accountancy.

The sensitivity of the correlation functions to the propagation of isotopic measurement errors were investigated. The study included errors at the 0.5, 1.0, 2.0 and 4.0% levels and were assumed to apply uniformly to each

of the isotopes. This was adopted only for the scoping study and the next level of study will reflect the measurement accuracies for each isotope as experienced in current experiments.

The preliminary results of the effect of measurement errors are also included in Figs. 3 a,b for only the 0.5 and 2% measurement errors. The top set of relations are based on the 0.5% error level and the lower set of functions are based on the 2.0% error level. The resolutions appear to be most adequate to discern one-seventh and one-twelfth substitution in some of the functions. The results serve two purposes: 1) It identifies the more insensitive functions to measurement accuracies and as such would aid in optimizing the set of functions for cross-correlations, and 2) It establishes measurement methods design criteria which should be used as a guide in the development and selection of measuring instruments.

The sensitivities of some correlation functions to the mixing of inner and outer core assemblies or blanket and core assemblies are maintained for fabricator's isotopic specification accuracies within this range of measurement errors. However, much tighter fabricator's specifications are to be expected.

The Pu/U range of measurement error limits expected in the assay of dissolver solutions, would readily allow discerning a substitution or dilution equivalent of a blanket assembly in a twelve core assembly batch. As shown in Fig. 4, the substitution results in a 13% decrease in the Pu/U ratio and 3.5% change in the ^{235}U depletion by the ICT method. The substitution of an inner-core assembly in a batch of twelve outer-core assemblies can also be detected within the error limits. Consequently the ICT method can be used as an independent verification of those measurement requirements needing more demanding accuracy limits such as in estimating the plutonium input to the reprocessing plant.

VIII. R&D PROGRAM PLANS AND IMPLEMENTATION

The isotopic correlation technique shows considerable promise for use in verifying the initial isotopic composition and isotopic depletion of discharged assemblies based on the measured ratios of several key isotopes, obtained at the dissolver stage in reprocessing. The verification of the quantitative estimate of the plutonium input into the reprocessing plant obtained by the Pu/U or Pu/U+Pu ratio method (Gravimetric Method) is not sufficient to detect certain anomalies in the nuclear material flow. The Gravimetric Method must be supplemented by the ICT. Determining the initial and depleted isotopic concentrations of the heavy metals using the ICT method is a necessary condition to identify anomalies in the flow of nuclear material between the output of the fabrication plant and the input to the reprocessing plant.

The ICSAM program for testing and evaluating these techniques is being planned for LWRs in cooperation with relevant U.S./DOE Laboratories and Commonwealth Edison Company. The fuel elements (pins) for the experiments would be obtained from the Zion-2 power system. The measurement techniques to be tested include the methods being developed for mass spectrometry techniques (filament and resin bead loadings), NDA, and classical wet analytical chemistry, coulometry and potentiometry. The measurements tests would be performed by the respective principal investigators. The sampling of the fuel elements are to be representative of the fuel assembly and selected to critically test the computational methods and the feasibility of the measurement systems.

IX. SUMMARY OF CONCLUSIONS

- Many of the correlation functions displayed linear, near-linear, or reasonably monotonically well-behaved functions over the expected burnup period for LMFBR fuels, and consequently may be utilized in verifying reactor input fuel specifications.
- The functionals were found to be sensitive to certain anomalies from pre-planned fuel management programs.
- The correlation functions were found to exhibit significant and detectable differentiation between inner core, outer core, and radial blanket assemblies.
- The propagation of measurement errors on the correlation functions and the respective sensitivities to isotopic compositional changes have been examined and found to be consistent with current measurement methods.

- Dependence Upon Initial Enrichment

Differences observed between corresponding correlations suggests that intermixing two differently enriched assemblies should be avoided.

This implies that safeguards requirements may influence, in part, fuel reprocessing management programs.

However, a pre-planned intermixing of different assemblies in a reprocessing program may yield well-behaved functionals which could be programmed for cross-correlations.

- Predissolver Stage (Assembly Item Accountability)

Isotope correlations and neutron assay methods have been investigated and may be applicable in determining the plutonium content and burnup of fresh and spent LMFBR and LWR assemblies. The NDA methods based on active and passive neutron coincidence counting (NCC) techniques are considered a primary safeguards technique to be developed for the LMFBR and the LWR fuel cycles.

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