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CONCEPTUAL DESIGNS OF NDA INSTRUMENTS FOR THE NRTA SYSTEM AT THE ROKKASHO REPROCESSING PLANT

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ABSTRACT

We are studying conceptual designs of selected nondestructive assay (NDA) instruments for the near-real-time accounting system at the Rokkasho Reprocessing Plant (RRP) of Japan Nuclear Fuel Limited (JNFL). The JNFL RRP is a large-scale commercial reprocessing facility for spent fuel from boiling-water and pressurized-water reactors. The facility comprises two major components: the main process area to separate and produce purified plutonium nitrate and uranyl nitrate from irradiated reactor spent fuels, and the co-denitration process area to combine and convert the plutonium nitrate and uranyl nitrate into mixed oxide (MOX). The selected NDA instruments for conceptual design studies are the MOX-product canister counter, holdup measurement systems for calcination and reduction furnaces and for blenders in the co-denitration process, the isotope dilution gamma-ray spectrometer for the spent fuel dissolver solution, and unattended verification systems. For more effective and practical safeguards and material control and accounting at RRP, we are also studying the conceptual design for the UO₂ large-barrel counter. This paper discusses the state-of-the-art NDA conceptual design and research and development activities for the above instruments.

I. INTRODUCTION

The Rokkasho Reprocessing Plant (RRP) is a large-scale reprocessing facility for spent fuel from boiling-water and pressurized-water reactors. The facility comprises two major components: the main process area to separate and produce purified plutonium nitrate and uranyl nitrate from irradiated reactor spent fuels, and the co-denitration process area to combine and convert the plutonium nitrate and uranyl nitrate into mixed oxide (MOX). The facility is under construction by Japan Nuclear Fuel Limited at Rokkasho-mura, Aomori prefecture at the Shimokita peninsula of the northern end of Honshu, Japan, and is planned to start operation some time after the year 2003 with a reprocessing capacity of 800 tons of uranium per year. To ensure that an effective and efficient safeguards system is applied at RRP, national and international safeguards agencies require near-real-time accounting (NRTA). We are studying conceptual designs of selected nondestructive assay (NDA) instruments for NRTA at the RRP.

This paper discusses the state-of-the-art NDA conceptual design and research and development activities for some selected instruments in the order of the input solutions in the main chemical separation process, holdup measurement systems in the co-denitration process, MOX- and UO₂-product canister measurement systems in the co-denitration process, and unattended verification systems.

II. ISOTOPE DILUTION GAMMA-RAY SPECTROMETRY (IDGS) FOR THE INPUT SPENT FUEL DISSOLVER SOLUTION

A. Overview

Dissolver solutions are the input to the main chemical process area where separation and purification occur. Dissolver solutions contain high levels of fission products and uranium (to 250 g/L). Plutonium concentrations are generally about 1% of uranium concentrations. Accurate, rapid determinations of the plutonium and uranium in reprocessing input dissolver solutions are very important for input accountability analysis, nuclear material control, and on-site verification, which are essential elements of the NRTA system at reprocessing plants.

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Isotope dilution mass spectrometry (IDMS) has long been the most accepted technique for determining the plutonium content of the input dissolver solution in reprocessing plants. However, IDMS is time consuming, sample preparation is lengthy, the equipment and operation are costly, and the final sample is not always representative of the batch content. The Hybrid K-Edge Densitometry/X-Ray Fluorescence Densitometer (HKED) measures element concentrations, but does not measure isotopic compositions. It would therefore be desirable to have an alternative technique that can provide timely, less expensive, and simpler on-site verification of the plutonium in the input dissolver solution to avoid IDMS disadvantages and to complement the HKED measurement of concentrations. Such a novel technique, isotope dilution gamma-ray spectrometry (IDGS), has been recently developed by using low-energy gamma-ray spectrometry.

B. Recommended Measurement System

IDGS is similar to IDMS except that the isotopic distributions of both unspiked (unknown dissolver solution) and spiked (adding to the dissolver solution a spike of well-characterized plutonium) samples are measured by high-resolution gamma-ray spectrometry rather than mass spectrometry, and that sample preparation is simpler for IDGS.

Details of the measurement principle and algorithms of the IDGS technique are described in Refs. 1 and 2. Briefly, the isotopic composition is determined by measuring the respective low-energy gamma rays of isotopes from the dissolver solution. With regard to the total plutonium concentration, the unknown solutions are spiked with a plutonium isotope of accurately known concentration and isotopic composition. The total plutonium concentration is determined by calculating the difference between the isotopic ratios of the spike, spiked (unknown sample plus the spike), and unspiked samples (unknown sample only). By measuring the isotopic compositions of both unspiked and spiked dissolver-solution samples, the concentration of plutonium in the unknown dissolver solution, \( C_u \), can be determined as

\[
C_u = \frac{M_s}{V_u} \cdot \frac{W_{240}^9}{W_{239}^9} \cdot \frac{R_m - R_x}{R_u - R_m},
\]

where

\[
M_s = \text{mass of plutonium in the spike},
\]

\[
R_m = \frac{W_{240}^9}{W_{239}^9}, \quad \text{the } ^{240}\text{Pu} / ^{239}\text{Pu} \text{ ratio in the spiked sample},
\]

\[
R_u = \frac{W_{240}^9}{W_{239}^9}, \quad \text{the } ^{240}\text{Pu} / ^{239}\text{Pu} \text{ ratio in the dissolver-solution sample}.
\]

In this equation, the values of \( M_s, W_{239}^9, R_s, \) and \( V_u \) are known. Therefore, only the values of \( R_u \) and \( W_{239}^9 \) in the unspiked dissolver-solution sample and \( R_m \) in the spiked sample need to be measured by gamma-ray spectrometry.

Gamma-ray measurements of highly radioactive dissolver solutions from reprocessing plants require rapid and efficient separation of plutonium from fission products and other actinides and efficient recovery of purified plutonium. Ion-exchange separation and extraction chromatograph techniques were used to obtain satisfactory purification and recovery of plutonium for the IDGS measurement.

C. Preliminary Design Specifications

The IDGS system is a standard high-resolution gamma-ray spectrometer, which consists of a hyperpure germanium planar detector and associated electronics, an ORTEC Multichannel Buffer (MCB), and an IBM-compatible personal computer (PC). The detector is a high-resolution hyperpure germanium planar detector with dimensions of 1000 mm$^2$ by 13 mm and a resolution of about 580 eV at 122 keV. The computer is designed to interact with the user, to control the MCB hardware to acquire spectra data for measurement information, to automate the analysis of these measurements, and to store gamma-ray spectra on disk for future analysis. A PC at least as advanced as a 486 33 MHz with 4 Mb memory and 120 Mb hard disk is needed.

D. Performance Specifications

IDGS has been successfully demonstrated in measuring plutonium isotopic composition and elemental concentrations of spent fuel dissolver solutions with burnups up to 28 000 MWD/T. Using IDGS to analyze dissolver solutions for plutonium isotopic compositions, the precision is approximately 0.5% for the \(^{240}\text{Pu} / ^{239}\text{Pu} \) ratio and 0.2% for the \(^{239}\text{Pu} \) (weight percent) within a 1-h count time. The agreement between IDGS and IDMS for dissolver solutions is very good, especially for the \(^{240}\text{Pu} / ^{239}\text{Pu} \) ratio (average IDGS/IDMS ratio is 0.997) and the weight percent of \(^{239}\text{Pu} \) (average IDGS/IDMS ratio is 0.999), which are important for determining the total plutonium concentration.
For plutonium concentrations in dissolver solutions, bias between IDGS and IDMS is less than 0.15% with a precision of better than 1%, within a 1-h count time.

III. HOLDUP MEASUREMENT SYSTEM FOR CALCINATION AND REDUCTION FURNACES AND FOR BLENDER GLOVE BOXES

A. Overview

Holdup is material that remains within the processing equipment after the process has been shut down and process materials have been run out. This material typically remains within the processing equipment from one operation period to the next. In-process inventory consists of materials that reside within the processing equipment during operation and at the time of shutdown of the process. These materials typically leave the process as product during the subsequent processing period. Both in-process inventory and holdup can be directly measured and quantified in situ by portable NDA methods.

The calcination, reduction, and blending glove boxes require NDA measurements to determine the amount of plutonium residing in the glove boxes for safeguards accounting. The plutonium may be in the category of in-process inventory or holdup material depending on when the measurements are carried out. Both gamma-ray and neutron coincidence measurements have been considered for the on-line quantitative determinations of plutonium inventory in these glove boxes. Based on previous experience with both of these methods at the Plutonium Fuel Processing Facility (PFPF) in Japan, neutron coincidence measurements are recommended as the primary method for measuring the plutonium inventory combined with secondary portable gamma-ray measurements to quantify possible biases from the neutron assay. For brevity, only conceptual design of neutron coincidence measurements will be addressed in this paper.

B. Recommended Measurement System

Previous measurement experience at PFPF, which has similar process equipment, has shown the measurement uncertainty from neutron coincidence assay (~5%) was less than that for gamma-ray assay (25-30%). The primary source of uncertainty in the gamma-ray measurements is the large equipment attenuation correction factors; however, secondary gamma-ray measurements are useful for quantifying the amount of material on the surfaces and floor of the glove box. Determining the amount of material in these locations individually is not possible with the current neutron coincidence assay system. These secondary measurements can be used to correct the neutron coincidence assay for biases that are possible when the quantity of material in these locations becomes large.

The time-correlated neutrons from plutonium that are used for the assay signal in the neutron coincidence measurements arise mainly from the spontaneous fission of the $^{240}$Pu isotope. For nonmultiplying quantities of in-process plutonium inventory, the coincidence count rate is proportional to the (effective) $^{240}$Pu mass. Because the in-process inventory can accumulate over many campaigns, it is difficult to improve the prediction of the $^{240}$Pu isotopic fraction based on a known origin of the spent fuel input material during a given processing campaign. It is recommended that on-line measurements of the plutonium isotopic composition be implemented, in addition to the neutron coincidence counting, to minimize this source of uncertainty.

C. Preliminary Design Specifications

The neutron coincidence assay system consists of four slab detectors composed of polyethylene, cadmium, and $^3$He tubes. Figure 1 is a top view showing the four slab detectors positioned around a glove box for a measurement and Fig. 2 is a more detailed diagram of one of the four slabs. Each slab is approximately 13 cm x 50 cm x 163 cm and contains 12 $^3$He tubes. The tubes are 2.54 cm in diameter with an active length of 150 cm and 6-atm of $^3$He. The junction box seals the area where the $^3$He tubes are connected to the AMPTEK preamplifiers and where the cable connections are made between the tubes/preamplifiers and the coincidence counting electronics.

Each slab detector is composed of two sections: the detector-half and the shield-half. The detector-half is the portion of polyethylene where the $^3$He tubes are placed and faces the glove box being assayed. The shield-half consists of 5 cm of polyethylene to thermalize neutrons coming in from neighboring boxes so they are more readily absorbed in the cadmium liner located along the center of the slab. The shield reduces the number of neutrons that are detected in the $^3$He tubes that came from glove boxes other than the one being assayed. Typically in fuel processing facilities, many rows of glove boxes are present in close proximity to one another, so “cross-talk,” neutrons from neighbor glove boxes contributing to the measured signal of the assay glove box, is a concern. Although cross-talk can be accounted for in the system calibration, it is time
Fig. 1. Overview of the Glove Box Assay System proposed for the Rokkasho Reprocessing Plant illustrating the system setup around a typical glove box with associated electronics.

Fig. 2. Detailed diagram of one of the four slab detectors used in the Glove Box Assay System.

Fig. 3. Glove Box Assay System scanning pattern for the measurement of holdup in the blending glove box. The numbers in the figure indicate the measurement positions.
B. Recommended Measurement System

Verification of the MOX powder contained in storage canisters requires both gamma-ray and neutron instrumentation. The high-resolution gamma-ray spectroscopy which is needed for the plutonium isotopic measurement will not be discussed in this paper. The passive neutron coincidence counter (NCC) is used to measure the spontaneous-fission rate from the plutonium for the $^{240}$Pu-eff mass determination. When this rate is combined with the plutonium isotopic compositions, the plutonium mass is determined.

The recommended measurement system would use the neutron coincidence counting of the spontaneous-fission neutrons to determine the $^{240}$Pu-eff. In the large mass samples such as the canister, there is a significant multiplication of the neutrons in the massive sample. Thus, we must use the multiplication-corrected $^6$ coincidence rate, $R_{mc}$.

The measured quantities are the totals (T) and the reals (R). However, the unknowns are the $^{240}$Pu-eff mass, the multiplication (M), and the alpha (alpha,n neutrons/spontaneous-fission neutrons). For the known alpha measurement method, the alpha value is assumed known from the chemical composition and M is calculated. The analysis of T and R give the quantity $R_{mc}$ that is directly proportional to the $^{240}$Pu-eff mass. The calibration function has the linear form

$$R_{mc} = a \times ^{240}\text{Pu-eff} + b,$$

where $a$ and $b$ are the calibration coefficients and $b$ is very nearly zero.

C. Preliminary Design Specifications

The neutron detector must be designed large enough to hold the tall canister. The efficiency for neutron detection should be the same for all of the plutonium positions in the canister. This results in a requirement for a 1130-mm-long flat zone (uniform efficiency) in the counter. The long flat zone is difficult to achieve because of the large diameter (435 mm) of the top of the canister. The large opening required to lower the canister into the neutron counter allows many neutrons to leak out of the ends of the counter with a resulting decrease in the efficiency at both ends of the counter.

Figure 6 shows a schematic diagram of the neutron counter including three port-holes in the side for simultaneous neutron and gamma-ray measurements. The neutron detector contains 18 $^3$He tubes. The maximum
counting rate in the detector electronics is \(-2.4\) MHz when the neutron emission rate from the sample is very large and varies with burnup and age since separation. We have designed the detector with an 8\% counting efficiency to give \(-1,000,000\) counts/s for the design basis fuel. Thus, the electronics can accommodate counting rates about twice that high for very high burnup and high \(^{241}\text{Am}\) content.

The efficiency of the detector in the central region is about 16\% prior to the custom tailoring, discussed below, to achieve a flat response over the 1130-mm sample height. Variable thicknesses of \(\text{CH}_2\) and thin cadmium strips are applied to each \(^3\text{He}\) tube to reduce the efficiency to \(-8\%\) and improve the uniform response over the sample region. Because the sample can contain one to three cans of MOX with variable fill heights, the multiplication is unknown and the multiplication-corrected result is needed for the assay. A uniform or flat counting efficiency is required to obtain accurate results using the multiplication-corrected coincidence technique.

Figure 7 shows an example of the custom tailoring using \(\text{CH}_2\) and cadmium from the plutonium canister counter\(^7\) that was designed for the COGEMA canisters at PFPP. The initial efficiency was reduced by a factor of \(-1.5\) to extend the flat zone from \(-700\) mm out to \(-1300\) mm. This was an easier case than for the present design because the COGEMA canister head has a smaller diameter than the REP canister head.

The initial design of the canister counter only needs to provide the excess efficiency \((-16\%)\) and active length for the detector as shown in Fig. 6. The final tailoring of the efficiency profile, as shown in the bottom of Fig. 7, is best done experimentally during the fabrication phase of detector development.

The large metal head on the storage canisters will significantly change the neutron response profile. Thus, final application of the cadmium shims must be done with a dummy canister in the detector. This is best done as a final step after the neutron detector has been fabricated. The excess efficiency (16\% vs 8\%) is provided in the design to allow the cadmium tailoring to give a flat profile over the 1130-mm sample length.

On the bottom of the detector is an aluminum plug to help flatten the profile by scattering neutrons back into the detector. This also provides a robust stop for the canister.

The electrical design of the MOX canister counter is very similar to the other passive neutron NDA systems.\(^8\)

The canister counter uses the new fast-counting circuitry based on the miniature AMPTEK hybrid chip. These chips are located near the end of the \(^3\text{He}\) tubes and contain the preamplifier, amplifier, and discriminator circuits. Nine of these amplifier units are located in the top part of the detector. The outputs of these eight amplifiers are added and sent to the input of the shift-register (SR) board. The connection is made through the “external SR input” on the back panel of the shift-register.
V. UO3-PRODUCT CANISTER COUNTER

Uranium oxide, nominally 1–1.6% enrichment UO3 powder, is another product from the co-denitration process and will be stored in 304SS canisters having a volume of 340 liters. The assay method selected for the UO3 canisters is a combination of passive NCC and high-resolution gamma-ray spectroscopy. The gamma-ray measurement is used to determine the uranium enrichment. The passive NCC is used to measure the quantity of 238U, 235U is inferred from the combination of the quantitative assay of 238U using neutron coincidence counting and the gamma-ray enrichment measurement. Extensive MCNP simulations were conducted to evaluate the neutron counter performance. Details of the UO3 canister counter are discussed in a separate paper in this proceeding.

VI. UNATTENDED VERIFICATION SYSTEMS FOR MOX- AND UO3-PRODUCT MATERIALS

A. Overview

The ability of NDA instrumentation to run in unattended operation has been demonstrated at several facilities. The need for these types of systems was prompted by increased numbers of facilities to safeguard with the same number of personnel as before the new facilities came on line. This unattended operation has increased the safeguards effectiveness of instrumentation and decreased associated costs for both operators and inspectorates.

The UO3 and MOX counter systems will assay plutonium and uranium. The unattended verification systems for these counters will consist of (1) local NDA cabinets located near the detectors, (2) computers located in a central inspection room, and (3) a local operating network (LON) interconnecting the NDA cabinets and computers. To make the required measurements, the MOX and UO3 counters will measure coincidence neutrons and acquire gamma spectra. It is assumed that International Atomic Energy Agency (IAEA) personnel would come to the plant at infrequent intervals, such as once every 30 days. The system consists of two parts, those that run in unattended operation when the inspector is not present and those that run only when the inspector is present. During the time that inspectors are not present, the system would collect and store data continuously in unattended operation. When inspectors arrive at the plant, the data collected during their absence would be reviewed and safeguards decisions made to determine when and how much material moved through the counters. The equipment would be under IAEA seal when inspectors are not present.

B. Preliminary Design Specifications

1. Preliminary Configuration. The overall configuration of the unattended monitoring systems for the plant is shown in Fig. 8. It consists of NDA cabinets located near the detectors, computers located in a central inspection room remote from the NDA cabinets, and a LON connecting the NDA cabinets with the computers. In addition to the computers and NDA cabinets on the LON, a master timer is also present to provide time synchronization of all devices on the LON.

The equipment contained in the NDA cabinets runs continuously in unattended operation. The cabinets contain different configurations of these types of equipment: intelligent shift register (ISR), intelligent multichannel analyzer (IMCA), identification (ID) reader and IBM-compatible PC. The ISR is a specialized data acquisition electronics device for the NCC that contains a specialized program for unattended operation. In addition to acquiring the neutron data, an ISR has the capability to temporarily store data in battery backup memory, to perform adaptive processing to determine if some of the statistically redundant data can be eliminated, and to record information about the instrument’s “state of health.” The ISR is capable of battery operation. An IMCA is used to acquire high-resolution spectra from high-resolution gamma-ray spectroscopy (HRGS) detectors in unattended operation. Like the ISR, the IMCA is capable of battery operation and performs several other functions in addition to basic data collection: storing data, determining which data can be eliminated, and reading the instrument’s “state of health.” Also in the cabinets is an ID reader that provides a means to identify the sample. Depending on the sample container, it could be a bar code reader or a camera that captures an image of the part of the container with the serial number; this image could be optically scanned and analyzed to obtain a serial number.

The MOX counter detectors and NDA cabinet are shown in Fig. 9. The NDA cabinet consists of two ISRs labeled ISR-A and ISR-B only for description purposes, one local PC, and three IMCAs labeled IMCA-A, IMCA-B, and IMCA-C. Cables connect the neutron and gamma-ray detectors to their associated electronics units. In addition, ISR-A is connected to a LON node that connects to a Central Collect computer located elsewhere in the plant. ISR-B is connected to the local
Another required interface is an ID reader; this device would interface to the ISRs to record the sample ID along with the neutron data. Both ISRs receive signals from the detector and the ID reader. Each of the IMCAs is connected to one HRGS detector. Each IMCA is also connected to the LON over which the accumulated spectra are transmitted to the Central Collect computer. The UO, cabinet would be similar to the MOX system except that it would have two IMCAs.

In the central inspection room, the Central Collect computer would be connected to local NDA cabinets via the LON. This computer would run unattended and would continuously collect data from the remote systems. When inspectors are present, this computer could be connected to the Review computer in the inspection room over a high-performance, local area network.

Archived data would be moved from the Collect computer to the Review computer, where the data acquired during the inspection period would be reviewed. This review would involve using analysis programs to determine the isotopics of the samples and to determine the amount of material present in each sample. Possibly there would be some interaction with the video system at this level so that images associated with measurements might be displayed simultaneously.

2. Preliminary Software Design and Method of Operation. The software design of the unattended monitoring systems is based on the assumption that the systems would run unattended for periods of 30–90 days and that the inspectors would visit the facility once at the end of the inspection period. The system can be thought of as consisting of two parts: those that run in unattended operation while the inspector is not present and those that are run with the inspector present. The majority of the unattended operations occur in the NDA cabinets located near the detectors. During the time the inspector is not present, the equipment running unattended is under IAEA seal. When the inspector arrives at the plant, he collects data from all the systems, reviews it and determines when and how much material has moved through the detectors.

The main software to be run when the inspector is present is the Review software, which would allow him to review and analyze the data that was collected in unattended operation. This program would be run after the data has been transferred from the Central Collect computer to the Integrated Review computer. All the NDA data (neutron, spectral, and ID) from both systems would be imported into a database. In addition, the Integrated Review program would import the operator declarations for the inspection period.

The primary purpose of the Review program is to determine when movements of material have taken place. Other analysis programs, such as isotopics programs like PC-FRAM or MGA and neutron coincidence counting programs such as NCC, would also be run to calculate the amount of material that had moved through the counters. The results from these programs would be returned to the Integrated Review program where they would be stored in the database and their results compared to the operator declarations. Figure 10 is an example of showing the comparison between the declared and measured data. The top part of the figure shows the time display of the detector with the peaks indicating when a sample was present in the detector. The user could specify a certain peak by clicking on it.
and then the bottom display would appear. Here the region around the peak is expanded and the total number of counts in the peak would be displayed as well as the calculated and declared amounts of material.

State-of-the-art safeguards systems at the time this system is being proposed for installation would also have the ability to integrate information as well as operator data from the video system into the Integrated Review program and would have a capability for some remote access to the data. Inspectors would use the video picture to verify that the sample was correctly positioned in the detector when the data were being acquired. A complete system design must cover the possibility that the final system, when installed would have the capability to handle these additional needs. The system designed here would be able to accommodate such requests in that a provision is made for data to be centrally located in the Central Collect computer during the inspection period. The capability to remotely access the data could be added to this Central Collect computer if the final system requirements specify such a need.

REFERENCES


