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NONDESTRUCTIVE ASSAY INSTRUMENTATION FOR A
SAVANNAH RIVER PLANT UPGRADE PROJECT

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NONDESTRUCTIVE ASSAY INSTRUMENTATION FOR A SAVANNAH RIVER
PLANT UPGRADE PROJECT*

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Abstract

We have designed and are developing three different computer-based spectrometer systems. Two will measure the concentration of Pu solutions by gamma-ray and by stimulated x-ray fluorescence emissions. The third will measure relative isotopic abundances from the gamma-ray emissions of solid samples in closed containers. All systems are coupled to remote terminals and bar code readers, and also to mini-computer based MCAs, which in-turn are linked to another computer to provide a state-of-the-art NDA measurement capability. Installation at SRP is planned in late 1985.

Introduction

There is a continuing need to improve the accountability of nuclear material. Over the past five years, the DOE's Office of Safeguards and Security has supported the test and evaluation of many different instruments, techniques, and methods at numerous defense facilities. Among the more successful non-destructive assay (NDA) instruments tested and evaluated have been neutron counters, calorimeters, gamma-ray spectrometers, and x-ray fluorescence analysis systems. A major cooperative effort is under way between E. I. DuPont de Nemours & Co., Inc., the contractor for the Savannah River Plant (SRP); the Los Alamos and Livermore National Laboratories; and the Mound Laboratory of Monsanto Research Corporation to design and install a

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state-of-the-art, near-real-time, nuclear accounting measurement capability.

There are six types of NDA instruments to be implemented - a neutron counter and a waste solution assay system (Los Alamos), a 4-station calorimeter (Mound), two gamma-ray and two x-ray fluorescence analysis systems to measure plutonium solution concentrations, and a two-detector gamma-ray system to measure the relative plutonium isotopics. The latter three NDA systems will be supplied by the Lawrence Livermore National Laboratory (LLNL). Los Alamos also has the responsibility of interfacing all of the above NDA mini-computer based analyzers into a larger sized computer, that will itself be interfaced to the present nuclear material accounting system used at SRP. For discussion of the other laboratory instruments, see references [1-4].

There are three rooms to be instrumented in the scrap recovery facility at SRP. The first two consist of a 'wet' laboratory, in which Pu solutions will be handled, and a 'dry' laboratory, which will handle sealed containers of solids. Many, but not all, of the instruments will require the use of germanium detectors, which in turn are coupled to computer-based, multi-channel analyzers. The latter electronic racks will be installed in the third room, creating an instrument equipment room that is separate from the 'wet' and 'dry' sample laboratories. Because it is plutonium solutions that are to be analyzed in the 'wet' lab, there will necessarily be glove boxes in that lab, with the requirement that the NDA measurement systems must be mechanically connected to the glove boxes.

Solution Assay by Gamma-Ray Spectrometry

The first type of NDA system consists of a precision machined stainless steel cell mounted inside a glove box in the 'wet' sample analysis

laboratory. An operator working on the opposite side of the glove box will draw into the cell about 20 milliliters of plutonium solution. The cell will be very closely coupled to a germanium detector low energy photon system (LEPS) that is outside the glove box; but the detector and its shielding are integrally coupled to the cell to prevent any geometric variations.

The plutonium solution concentration is determined from the measured intensity of the emitted gamma rays. A large collimator will be inserted if solution concentrations are below a pre-determined value, while a small collimator will be used for concentrations above that value. A vacuum deposited and sealed plutonium metal source can be moved into position to periodically check the system performance. Finally, a Cd-109 monitor source is also integrally mounted into the system's hardware. The 88-keV peak from this source is measured for all analyses and its decay corrected intensity is compared to the calibration data intensity. Any difference indicates a change in detector efficiency, and thus, the final plutonium concentration is adjusted accordingly.

The LEPS detector will measure the low-energy region of the spectrum; i.e., from 50- to 220-keV, with some important information coming from the complex 94- to 104-keV region of the plutonium spectrum. The measurement methodology is very similar to that developed for the Tokai-mura reprocessing plant located in Japan and described in reference [5]. It is premature to provide any further details of this measurement system at this time, since it has not yet been delivered to SRP.

Solution Assay by X-Ray Fluorescence Analysis

The second type of NDA system to be implemented in the 'wet' analysis laboratory is an x-ray fluorescence analysis system. It consists of a

cylindrically machined stainless steel cell into which about 15 ml of plutonium solution is drawn. Two collimated 20 mCi Co-57 sources are used to ionize the K-shell electrons of Pu atoms, which immediately decay by K x-ray emissions. The 122-keV gamma rays of Co-57 and the actinide K-alpha x rays (about 100 keV) easily penetrate the 0.9 mm thick stainless steel cell walls. The emitted x rays are measured by a LEPS germanium detector and solution concentration is essentially proportional to the measured x-ray intensity.

Once again, the solution cell is mounted inside the glove box, while the detector is mounted outside, but both are integral members of the same hardware housing that uses a polycarbonate window to separate components inside the glove box from those that are on the outside. Behind the cell and co-linear with the detector axis is a paralene coated, circular Th metal foil. It is also fluoresced by 122-keV photons that pass through the cell. The emitted thorium K α x-rays are measured by the LEPS detector and provide a means of quantitatively measuring the solution density - that is, their intensity is sensitive to acid molarity, to temperature, to any anions or cations in the solution, or any other unknown but minor solution components. Thus, the Pu solution concentration is proportional to the Pu K α x-ray intensity, while the Pu/Th K α ratio is independent of solution density and the Co-57 half life.

A computer controlled tungsten shutter is used to eclipse the exciter sources so that the passive solution radioactivity can be measured. This is necessary because gamma rays emitted by several of the Pu isotopes fall directly beneath the Pu K α x-ray line. The net fluoresced x ray intensity is obtained by using sophisticated peak shape mathematical algorithms to fit the x-rays in the difference spectrum; i.e., the active spectrum minus the passive

radioactivity spectrum. This system is similar in operational principle to that described in reference [6].

Two-Detector Solids Plutonium Isotopics System

A two-germanium detector, gamma-ray spectrometry system is being developed to measure isotopic ratios of the plutonium component in feed scrap materials and oxides in the sealed containers. A LEPS germanium detector is the ideal detector to measure the low energy plutonium gamma-ray emission spectrum. As this spectrum is being acquired, a large coaxial germanium detector is measuring the higher energy gamma rays emitted from the sample. Once both spectra are obtained, a sophisticated analysis code is used to simultaneously solve the 120 linear equations that represent all of the major peak regions, and thus determine the isotopic abundances with their attendant errors. This approach to the analysis of Pu gamma-ray spectra has been called MGA; i.e., multiple group aalysis. In principle, no better results for plutonium isotopic abundance ratios can be obtained for the counting period used, because all information content in the spectra are utilized to obtain the composite isotopics. The details of this methodology can be found in reference [7].

Computer-Based Electronic Support Instruments

Each of the three types of NDA measurements discussed above will be supported by a computer-based, analyzer unit. Each station will consist of a series 90 Canberra multichannel analyzer interfaced to a Digital Equipment Corporation (DEC) Micro-11, 16 bit computer, which includes a 10 megabyte hard disc with a dual-sided, double-density, mini-floppy disc. Remote terminals are located immediately adjacent to each of the NDA instruments and are

interfaced to one of three, computer-based, analyzer units. A bar code reader is also located at each NDA instrument and is interfaced to its terminal and one computer-based multichannel analyzer station. Once an operator initiates spectrum accumulation through any one of the terminals, the subsequent control of a shutter, or differencing or combining of spectra is automatically controlled by the computer-based analyzer systems. Results from any one of these analyzer stations is relayed to an instrument control computer, a DEC VAX 750. In the event that the VAX-750 is not operational, the individual analyzer units have the capability to function as stand alone instrument controllers and to store results for at least one week. More detailed information about the instrument control computer can be found in another paper in these proceedings [4].

One gamma-ray spectrometer measurement cell and one x-ray fluorescence analysis cell are connected to one of the series 90 MCAs, which is multiplexed into two separate 8192 channel memory groups. Since the gamma-ray and x-ray fluorescence analyses will, in most cases, be measuring the same solution, the concentration results should be the same.

There will be two gamma-ray and x-ray analysis systems, each paired as above. In this manner, each system will offer replicate and redundant analysis capabilities to the other. This will be the first time analytical measurements for plutonium concentrations in the USA will rely on two totally different and independent nondestructive assay instruments, rather than on traditional 'wet' chemical analytical methods. Therefore, it would be very difficult to successfully tamper with both instruments in ways that would permit the successful diversion of material by insiders.

Because two detectors are required by the solids isotopics system, they will be connected to the third series 90 Canberra, Micro-11 based MCA

system. As mentioned earlier, each of the three analyzer systems will be remotely located from the NDA measurement systems in an instrument equipment room, which will be temperature and humidity controlled.

Prior to installation at SRP, all of the NDA systems being developed by all of the laboratories will be sent to Los Alamos, where they will be "staged" or operated in a manner similar to that to be used at the final installation. This staging offers several advantages. Each of the instruments must be coupled to bar code readers, remote terminals and to the instrument control computer; then the interfacing between this computer and each peripheral and each of the NDA Micro-11 based analyzer systems must be optimized. Another advantage is that the staging area can serve as a preliminary training ground for the senior operators and supervisors that will eventually be in charge of the NDA instruments after installation. Finally, it will permit the training of service personnel and at the same time serve as a test of the documented operational procedures necessary to use each instrument.

Conclusion

It is not possible to discuss the success of these instruments at this time. However, LLNL and the other laboratories involved have been working for almost two years individually with DuPont and SRP, and for over one year in a group planning mode. All of the meetings to date have been very productive and these efforts represent one of the most comprehensive approaches to the design of an integrated NDA facility attempted to date. When it becomes operational, it will represent the state-of-the-art in NDA instrumentation and in near-real-time nuclear material accounting systems within the Department of Energy's Savannah River Plant scrap recovery facility.

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