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Author(s,):	H. A. Smith, Jr., H. O. Menlove, T. D. Reilly, G. E. Bosler, E. A. Hakkila, and G. W. Eccleston
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## THE LOS ALAMOS NUCLEAR SAFEGUARDS AND NONPROLIFERATION TECHNOLOGY DEVELOPMENT PROGRAM\*

H. A. Smith, Jr., H. O. Menlove, T. D. Reilly, G. E. Bosler, E. A. Hakkila, and G. W. Eccleston Los Alamos National Laboratory Los Alamos, New Mexico 87545 USA

## Abstract

For nearly three decades, Los Alamos National Laboratory has developed and implemented nuclear measurement technology and training in support of national and international nuclear safeguards. This paper outlines the major elements of those technologies and highlights some of the latest developments.

#### Background

After the end of World War II, concern about the spread of nuclear weapons and nuclear technology spawned the organization of the International Atomic Energy Agency (IAEA) with a mandate under international treaty to verify the peaceful use of nuclear technology worldwide. This verification took the form of periodic inspections of nuclear facilities to verify operations and nuclear material inventories. State systems of accounting and control of nuclear material were then formed to coordinate nuclear material management for the states themselves and to interact with the international system.

At the facility level, where the nuclear material is controlled and accounted for at the source, measurement programs had to be established, and measurement technology was needed to provide the raw data for the national and international systems. A safeguards technology development program began in Los Alamos in the middle 1960s to provide technical support to meet facility, state, and international safeguards needs. Nuclear measurement technology was just beginning to mature, and there were many interesting research and development challenges arising from the needs of safeguards inspectors and nuclear materials managers. In the years that followed, a wealth of measurement techniques and instrumentation was produced to address these measurement needs.

In the present-day environment of fast-moving nuclear material transactions and heightened world concern over proliferation, it is even more important that safeguards systems provide timely information on the amounts and locations of nuclear material holdings.

## **Key Technologies/Applications**

Nuclear material movements must be controlled and documented, and nuclear material inventories must be carefully measured in a large variety of nuclear facilities worldwide. The broad range of possible material diversion scenarios requires that versatile measurement systems be established and comprehensive controls on nuclear material movements be in place at facilities. The wide spectrum of nuclear material chemical and physical forms and packaging dictates a broad repertoire of nuclear measurement techniques and associated instrumentation.

## **Safeguards Systems Technologies**

In the United States, a systems approach is used to establish facility and national safeguards. Modeling and simulation technologies are used to determine optimum system configurations and to specify the most effective measurement programs. New

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technologies aimed at improving the effectiveness and efficiency of safeguards at nuclear facilities include the development of advanced nuclear materials accounting techniques. Los Alamos has been instrumental in the development of near-real-time accounting (NRTA) as an effective method of providing acceptable timeliness for materials balance closures at large, sensitive, bulk-handling facilities such as nuclear fuel reprocessing plants and plutonium fuel fabrication facilities. To handle the large volume of data generated by NRTA systems, statistical data analysis packages, such as Materials Accounting With Sequential Testing have been developed. To design the NRTA systems, facility simulation codes have been developed to simulate facility operations and analytical measurements. Any large NRTA system is designed to detect the loss or diversion of nuclear material during operation of the process. However, because of the way some statistical tests are designed, some alarms can be generated that are not significant in detecting material loss. Methods are required to differentiate these false alarms from real indications of material loss. Los Alamos is developing various techniques to accomplish this. These techniques rely on intelligent systems, such as neural networks.

#### Safeguards Measurement Instrumentation

Nondestructive assays (NDA) of special nuclear materials (SNM) exploit the radiative properties of the materials, which emit gamma, neutron, heat, and beta radiation with specific radiative signatures for each isotope. The intensity of emitted radiation is related directly to the amount of nuclear material present. Techniques have been developed to assay nuclear material in many chemical and physical forms throughent the nuclear fuel cycle using this simple approach. Assays are obtained, using computer-based instrumentation, with increased levels of sophistication and automation and a growing emphasis on miniaturization for field use.

Nuclear measurement techniques range from small, portable gamma-ray-based systems to large, in-plant neutron-based instruments. The measurements are applied to input (feed) materials, product materials (for example, oxides, fluorides, powders, metals, fuel pellets, and fuel assemblies), scrap and waste from processes, in-plant nuclear material holdup, and facility effluents. Measurements are made at key transfer points in facility processes, at the perimeter access points to facilities, and at shipment and receipt points for material transfers among facilities. These nuclear material measurement techniques have also been useful for process monitoring and quality control applications in the facilities.

## **Gamma-Ray NDA Measurement Systems**

Gamma radiation from the decay of SNM, SNM daughters, and fission products can be used to identify and quantify the amount of isotopes present in a sample. The key problems in using this type of radiation for assays are to identify the gamma-ray signatures uniquely and to correct the measured data for absorption effects that are usually present. Samples that are highly heterogeneous, massive, or large in size exhibit excessive and sometimes drastically varying absorption effects that severely undermine the quality of the assay. Such samples are usually inappropriate for gamma-ray assays and are better analyzed with techniques that use more penetrating neutron radiation.

The basic gamma-ray assays involve a photon detector, data acquisition and analysis electronics, and a method to manipulate the sample to be assayed. In addition, the absorption losses are often measured by using an external gamma-ray source to determine the absorption properties of the sample. Passive measurements of gamma-ray emissions from SNM are isotope-specific, giving the isotopic composition of the sample and yielding individual isotopic masses. When applied to spent fuel, gamma-ray measurements give isotopic ratios of fission products that can be used to determine the burnup and cooling time of the fuel assemblies.

An example of the automated gamma-ray measurement systems is the segmented gamma scanner (SGS, Fig. 1). The sample is assayed in successive segments by

stepping the sample past the narrowly collimated field of view of the detector. An absorption correction is measured for each segment with an external gamma-ray source, making the overall quality of the assay less vulnerable to inhomogeneities in sample material density. Sample movements, assay data acquisition, absorption correction, and data analysis are carried out by the instrument computer. SGS measurements are best suited for low-density uranium or plutonium scrap and waste and yield assay results to better than a few percent for SNM masses from a few grams to as much as a kilogram.

The basic SGS approach has recently been broadened to provide a three-dimensional view of SNM samples through tomographic gamma scanning (TGS). The notion that computed tomography could be applied to the problems of assaying highly inhomogeneous samples was conceived long ago; but only recently have improved computer technology and more stringent waste assay requirements provided the impetus to pursue the idea. A prototype system is now under development at Los Alamos (Fig. 2). Through this technique, density variations in the sample can be located, and more accurate absorption corrections can then be applied to the assay data.

For applications that require more portability but continued sophistication, the portable multichannel analyzer (PMCA) was developed. This instrument provides all the necessary power, data acquisition, data recording, and data analysis needed to perform reliable assays in field operations (See Fig. 3). The instrument is used extensively by the IAEA, many nuclear facilities, and other international safeguards inspection operations (See Fig. 4). This type of instrument is currently being miniaturized further at Los Alamos. The latest prototype is approximately 1/5 the volume of the PMCA, has increased functionality, and is interfaced with the latest hand-held compater technology (see Fig. 5). NDA measurements of uranium enrichment, plutonium isotopic composition, and in-plant nuclear material holdup as well as other verification measurements can be accomplished with accuracies of a few percent or better using portable gammaray spectroscopy instrumentation. The portable nature of this instrumentation also makes it very effective in safeguards and nonproliferation inspection activities.

Active gamma-ray measurement techniques that have been developed for nondestructive assay of SNM include X-ray fluorescence (XRF) and absorption-edge densitometry. These techniques are most effective when applied to highly uniform samples and hence tend to be used to analyze SNM solutions. Since these techniques involve the use of external photon sources to generate the assay data, they are successful in assays of samples with high passive gamma-ray output, such as process solutions with high fission product content. These techniques generate highly precise and accurate assays, with precisions as good as 0.2% and sufficient accuracy in as little as 15 minutes of data acquisition. The latest development in this area is a combination of the two techniques into one instrument (Fig. 6) to determine simultaneously the concentration of uranium, plutonium, and mixtures of the two elements in solutions. This application combines the strengths of the two techniques to determine the concentrations and uranium/plutonium ratios. It has been successfully applied in Europe to assay input dissolver solutions for reprocessing plants as well as product solutions.

#### **Neutron NDA measurement systems**

Neutron radiation comes from prompt, multiple emissions during the fission process, delayed emissions from fission fragments, and random emissions from matrix materials that are bomharded by alpha particles from SNM decays. Neutron radiation is very penetrating, so sufficient radiation intensity is available to allow assays of even highly dense or heterogeneous materials or both.

Simple counting techniques for single neutrons were developed primarily to verify neutron levels. Detailed assay information is not usually practical with this method, in view of the many possible sources of neutrons from chemical and nuclear effects and the lack of spectral detail inherent to the neutron detection process.

Neutron detectors that can count coincidence neutrons are sensitive to fission processes in samples and as such allow for the quantitative assay of SNM. The basic passive neutron coincidence counter (Fig. 7), with a well designed to surround the sample with neutron detectors, was developed at Los Alamos in the early 1970s. The counting electronics registers the coincidence count rate which is proportional to the fission rate in the sample. Some of the plutonium isotopes commonly encountered in SNM have a significant spontaneous fission rate, so passive neutron coincidence counting is well suited for plutonium assays. Uranium, on the other hand, has a very low spontaneous fission rate, so the radiation signal is not adequate for passive neutron coincidence assays of uranium. The fissions must be induced by an external source, and then the coincidence fission neutrons can provide adequate assay data. The so-called active-well coincidence counting after inducing fissions with an external source of single neutrons.

The conventional two-fold coincidence counting of prompt fission neutrons has recently been extended to measurement of higher multiplicities of neutrons from SNM samples. Measurement of three-fold and higher coincidences requires higher detection efficiency, and new counter designs have been developed at Los Alamos to achieve this (Fig. 9). High-multiplicity counting of SNM samples yields additional information about the sample and permits accurate measurements of highly impure SNM-bearing materials that were difficult or impossible to assay in the past.

Active neutron coincidence counting has also been adapted for assaying fresh reactor fuel assemblies. The neutron coincidence collar (Fig. 10) irradiates a segment of the fuel with single neutrons, and the detector electronics counts the coincidence neutron events from the induced fissions. This instrument determines the fissile content per unit length of the fuel assemblies and is designed to detect the removal of fuel rods from the assembly.

Other active neutron assay applications include the differential die-away technique, photo-neutron measurements, and delayed neutron interrogation. This latter active neutron technique detects the delayed neutrons following induced fissions. An intense neutron source is brought from behind shielding to irradiate the sample and induce fissions. Then, after the exciting source is returned to its shielding, the delayed neutrons from the fission products are counted, giving an assay signal that is proportional to the amount of fissile material in the sample. This type of assay technique is quite sensitive and can detect milligram quantities of fissile material in  $200-\ell$  drums of waste. In addition, the technique is also useful for high-gamma-yield samples (such as spent fuel), because the detectors can be heavily shielded from gamma radiation and also can be designed with low intrinsic gamma sensitivity. The shuffling motion of the exciting source to and from the sample, in repeated generation of assay data, gives the instrument the name of "shuffler." These shufflers tend to be quite large in-plant installations (Fig. 11).

A variation on delayed neutron interrogation is exemplified in the ruel rod scanner (Fig. 12). This device excites the loaded fuel rod with neutrons as the fuel rod moves past a stationary neutron source. Delayed gamma radiation from the resulting fission fragments is counted downstream from the excitation source. This technique provides the fissile profile of the rod for product quality control as well as the total number of grams of fissile material in the rod for materials accounting.

Spent nuclear fuel emits not only intense gamma radiation fields but also intense neutron radiation. Gross neutron counting of spent fuel, combined with gross gamma counting, can be used to infer the cooling time and burnup of the fuel in its storage position. A simple detector and electronics package was developed at Los Alamos to obtain this type of data in underwater storage ponds at reactor facilities (Fig. 13).

The high penetrability of neutron radiation requires large amounts of shielding and tends to make neutron assay instruments quite bulky. Developments are under way, however, to reduce the size of detectors and instruments to achieve greater portability for these techniques.

## Perimeter Safeguards Technology

Los Alamos has pic seered in the application of radiation detection to portal monitoring of personnel and vehicles crossing the boundaries of nuclear facilities (Fig. 14). These monitors measure the radiation levels from items passing into or out of a facility to catch the unauthorized movement of SNM across the facility boundary.

## Training and Technology Transfer

An important part of the development of assay instrumentation is the transfer of that technology to capable suppliers and the transfer of the technical knowledge to the potential users. Thus an integral element of the Los Alamos Safeguards Technology Development Program is an extensive training program involving in-depth lecture materials on the measurement physics and extended laboratory exercises in which real nuclear material is assayed with the instrumentation developed (Fig. 15). Training courses currently offered at Los Alamos are as follows:

- Materials Accounting for Nuclear Safeguards: A course in lecture/workshop format that presents the principles of safeguards systems, including system design, system operation, MC&A data acquisition and analysis, and decision analysis.
- Nondestructive Assay Techniques for Safeguards Practitioners: A basic course that introduces the basic concepts of neutron and gamma-ray nondestructive assay techniques, using hands-on classroom exercises with actual SNM samples.
- Nondestructive Assay of Special Nuclear Materials Holdup: Provides hands-on field practice in the measurement of SNM deposits in simulated plant equipment, piping and ductwork, along with the special data acquisition and analysis problems peculiar to these types of measurements.
- Gamma-Ray Spectroscopy for Nuclear Materials Accountability: A hands-on laboratory course that illustrates the application of high-resolution gamma-ray spectroscopy to a variety of NDA measurement techniques, along with the detailed data analysis that accompanies the use of high-resolution spectroscopy equipment.
- Nondestructive Assay Inspector Training Course: A two-week training module for IAEA inspectors that covers basic NDA principles and measurement exercises, with measurement of SNM in the various forms encountered in international inspections. This course is an integral part of the formal training of all new IAEA inspectors.
- International Training Course on State Systems of Accounting for and Control of Nuclear Materials (SSAC): A course mandated by the US Congress for safeguards practitioners in developing countries. This threeweek-long lecture/workshop course describes the elements of a State System and its required interactions with the International Safeguards (IAEA) system. The course includes lectures on existing SSACs and provides workshops on safeguards technologies, SSAC design, and international safeguards. The course includes participation by staff from a model US nuclear facility at an illustration of a facility in which materials accounting is set up to accommodate SSAC requirements.

The safeguards technology developed at Los Alamos has widespread application, and so convenient commercial sources of the technologies must be made available to potential users worldwide. As a result, Los Alamos maintains an on-going program of transferring proven technologies to commercial US manufacturing firms, and many NDA instruments can now be purchased from US companies. Examples include segmented gamma scanners, portable multichannel analyzers, portal monitors, neutron coincidence counters, and neutron coincidence collars.

## **US Projects**

The Los Alamos Safeguards Technology Development and Training programs provide full-scope technical support to US nuclear facilities and regulatory agencies in nuclear stafeguards. Materials accounting and process control needs are supported by safeguards systems expertise and by numerous installations of automated NDA instruments. Figure 16 shows the locations in the US where Los Alamos safeguards projects are in progress or have taken place.

## Worldwide Projects

The international safeguards community is also heavily supported by expertise and technology from Los Alamos. NDA instrumentation is used by IAEA and Euratom inspectors as well as by many facility operators worldwide (Fig. 17). The US maintains bilateral and multilateral cooperation activities with many countries to help enhance materials accounting and safeguards in international facilities. For many years, international inspectors have been trained at Los Alamos in the areas of NDA measurements. Training is now being developed in areas of nonproliferation inspection procedures and skills to broaden the effectiveness of existing inspection activities.

An example of these multilateral cooperative activities is the investigation of the direct use of spent pressurized water reactor (PWR) fuel in Canadian depleted tranium (CANDU) reactors (DUPIC), which is being carried out by the Korean Atomic Energy Research Institute and Atomic Energy of Canada Limited, with the assistance of Los Alamos safeguards staff. The DUPIC concept involves the reuse of spent PWR fuel in CANDU reactors without reprocessing the fuel. The spent fuel would be reconstituted into CANDU-type fuel pins with most of the fission products intact for proliferation resistance. The DUPIC fuel cycle extends the lifetime of the fuel and reduces spent-fuel storage requirements.

## **The Future**

Both the US and the international safeguards regimes are changing, and there are many new challenges as we move to the future. Weapons dismantlement activities in both the US and the Former Soviet Union will produce highly strategic nuclear material to store and safeguard. The down-sizing and reconfiguration of the US nuclear material complex will create many safeguards measurement needs to characterize the facilities and establish SNM inventories before decommissioning or reconfiguring these facilities. As the world emphasis shifts from arms reduction and arms control to nonproliferation, greater emphasis will be placed on the use of more sophisticated measurements and inspection techniques to identify undeclared facilities and proliferant activities. This will drive the needs for further technology developments and enhanced safeguards training for international and national inspectors.

## Further Reading on Los Alamos Safeguards Technology Development

- 1. "Program Feature Nuclear Safeguards," Los Alamos Science 1, (1), 67-137 (1980). This section of the premiere issue of this journal contains articles on the history of safeguards, safeguards technology, and safeguards Systems.
- 2. Proceedings of the Symposium Commemorating 20 Years of Safeguards Technology Development at Los Alamos, *Journal of the INMM*, 15, (4), 18-99 (1987). This symposium contained presentations from major figures in US and international safeguards and recounts the developments of safeguards technology and safeguards and nonproliferation policy worldwide.
- 3. Keepin, G. Robert, "Los Alamos Safeguards Program Overview and NDA in Safeguards," Los Alamos National Laboratory document LA-UR-88-3190 (1988). This paper, by the founder of the Safeguards Program at Los Alamos, recounts the

history of safeguards and the then current status of safeguards technology developments at Los Alan.os.

- 4. Keepin, G. Robert, "Nondestructive Assay," Los Alamos National Laboratory document LA-UR-89-1105 (1989). This paper gives a brief overview of NDA technology and applications.
- 5. T. D. Reilly, N. Ensslin, H. A. Smith, Jr., and S. Kreiner, (Eds.), *Passive Nondestructive Assay of Nuclear Materials*, Nuclear Regulatory Commission Textbook NUREG/CR5550 (1991). This is a con prehensive treatise on all aspects of gamma-ray and neutron NDA using passive techniques, with treatment of some active techniques. Available from the Superintendent of Documents, US Government Printing Office, P. O. Box 37082, Washington, DC 20013-7082.



Figure 1. Segmented gamma-ray scanner (SGS) developed at Los Alamos and now commercially available through Canberra Industries. The highresolution gamma-ray detector (left) looks through a narrow collimator at a segment of the sample (thin can in center of figure). Gainma-ray absorption corrections are measured with the help of an external gamma-ray source on the opposite side of the sample from the detector (right-hand portion of instrument). Instrument electronics (far right of picture) manages instrument operation and data acquisition, displays spectra, and conducts data analysis. Inset shows the measurement geometry for assay segments.

## **SGS Measurement Geometry**





Figure 2. The tomographic gamma scanner (TGS). This prototype unit uses tomographic scanning procedures and analysis techniques to locate density variations in the sample container. The passive gamma-ray assay data can then be corrected more explicitly for these density variations, yielding more accurate assays.



Figure 3. Portable multichannel analy. er (PMCA) manufactured by D.S. Davidson and Co. This battery-powered urit provides detector power, pulse-processing electronics, data recording devices, spectral display, computer communication, and operator/instrument communication for portable gamma-ray assays.



Figure 4. United Nations inspectors in Iraq, verifying 200-liter drums of uranium yellowcake for proper <sup>235</sup>U enrichment values, using the Davidson portable multichannel analyzer (PMCA) and Nal scintillator gamma-ray detectors.



Figure 5. Miniaturized prototype of the PMCA, currently under development at Los Alamos. Full functionality with gamma detectors (right of figure above) and data storage capacity are achieved by interfacing the main unit (center in figure above) with hand-held computers (left in figure above), for maximum portability of in-field measurements. In figure below, operator is measuring a gamma-ray spectrum with a NaI detector (in her right hand) and recording the result in a hand-held computer (left hand. The multichannel analyzer is mounted in a holster on her belt.





Figure 6. The hybrid XRF and K-Absorption-Edge Densitometry Assay Instrument. The instrument assays mixed-oxide solutions for U/Pu ratio as well as uranium and plutonium concentrations.



Figure 7. Passive (high-level) neutron coincidence counter (HLNCC). The counter (at the right) is cylindrical to provide a well in which to place the sample (in the can on the counter) and surround it with neutron detectors. The assay electronics (on the benchtop) powers the detectors, acquires the data, and performs an analysis to obtain mass of fissioning material.



Figure 8. Active well coincidence counter (AWCC). A sample (in the operator's right hand) is placed in the sample chamber, which is surrounded by neutron detectors. The top and bottom end caps (top cap shown in the figure) contain random (AmLi) neutron sources to induce fissions in the sample material. The neutron detectors and counting electronics then determine the coincidence neutron count rate, which is proportional to the fissile mass in the sample.



Figure 9. Passive neutron multiplicity coincidence counter. The neutron detector tubes are partially removed from one half of the counter to show their arrangement and number. The larger counter volume and greater number of detector tubes (compared with the HLNCC, Fig. 7.) provides the high counting efficiency necessary to measure higher coincidence multiplicities.



Figure 10. Active neutron coincidence collar, shown measuring a simulated boiling water reactor fuel assembly. A random single-neutron source excites the assembly, inducing fissions in the fissile isotopes of the fuel material. The neutron detectors and coindicence electronics then determine the induced coincidence count rate, thereby determining the grams of fissile material in the segment of the assembly being measured.



Figure 11. Delayed neutron interrogator for 200-liter (55-gal) drums. The large size of the instrument is primarily to accommodate the shielding needed to protect operators and the detectors from the excitation source when it is stored.



Figure 12. Fuel rod scanner, employing the principle of neutron irradiation of the fuel material, followed by measurement of the delayed gamma activity in the excited fuel material. The instrument gives a measure of the fissile content of the rod and can measure rod loading as well as fissile mass for material accounting reasons. Upper figure: schematic of in-plant rod scanner showing the neutron source (bulky structure at top) and the rod conveyor (the long rack passing through the neutron source). The lower figure shows the prototypes of rcd scanners, as they were under development at Los Alamos.



Figure 13. Los Alamos "FORK" detector and GRAND electronics system for measurement of spent nuclear fuel. The detector is positioned around an underwater spent fuel assembly, and gross gamma and neutron rates from the assembly are measured. From these data, it is possible to determine the burnup and cooling time for the assembly.



Figure 14. Portal radiation monitors for detection of unauthorized movements of SNM across a facility boundary. These monitors have been developed for checking both personnel (left portion of the figure) and vehicles (right portion of the figure) at the points where they enter or leave the facility.



Figure 15. Los Alamos Safeguards Technology Training Courses provide in-depth instruction in safeguards, materials accounting, and NDA measurement principles, with hands-on experience in NDA measurements with SNM samples. Los Alamos instructor (center) and IAEA inspectors in a classroom exercise for measurement of uranium enrichment use high-resolution gamma-ray spectroscopy.



Los Alamos National Laboratory, Nuclear Safequards Research and Development

Figure 17

# Nuclear Safeguards Research and Development Experience at US Facilities

