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ADVANCES IN NUCLEAR INSTRUMENTATION FOR SAFEGUARDS

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Abstract

Emerging issues impacting nuclear materials management include nonproliferation, disarmament, the Comprehensive Test Ban Treaty, a fissile material production cut-off, weapons material storage and disposal, waste storage and disposal, remote and environmental monitoring, and smuggling and export control. All of these issues require measurements of an increasingly varied range of material forms. This, in turn, requires the improvement of existing techniques and the development of new nondestructive assay methods, new sensors and detectors, and new data analysis techniques.

This paper describes detectors, instrumentation, and analytical methods under development to address the above issues. We will describe work underway on room-temperature semiconductors including attempts to model the response of these detectors to improve spectrum analysis procedures and detector design. Computerized tomography is used in many medical and industrial applications; we are developing both gamma-ray and neutron tomography for improved measurements of waste and direct-use materials. Modern electronics and scintillation detectors should permit the development of fast neutron coincidence detectors with dramatically improved signal-to-noise ratios.

For active measurements, we are studying several improved neutron sources, including a high-fluence, plasma-based, d-t generator. New analysis tools from information theory may permit one to better combine data from different measurement systems. This paper attempts to briefly describe a range of new sensors, electronics, and data analysis methods under study at Los Alamos and other laboratories to promote discussion of promising technology that we may bring to bear on these important global issues.

1. Introduction

The changing world order has brought new areas of concern to nuclear nonproliferation, nuclear material safeguards, and nuclear disarmament. These are very hopeful changes for world peace and cooperation, and they pose challenging technical problems to those who work in these fields. Issues that impact nuclear material safeguards include nonproliferation and the Nonproliferation Treaty (NPT), a fissile-material production cutoff, the Comprehensive Test Ban Treaty (CTBT), weapons dismantlement, nuclear material and waste storage and disposal, remote and environmental monitoring, and nuclear smuggling and export control. This is a long, and incomplete, list of issues that all relate to controlling the nuclear genie. All have requirements to detect and measure nuclear materials, and these requirements can not all be met by present measurement technology and instrumentation. This paper describes improvements in existing technology and the planned development of new technology, new sensors and detectors, and new data analysis techniques. It concentrates on research and development underway at Los Alamos National Laboratory, and it includes work that is ongoing at other government and educational institutions.

Over the past 50 years, there has been a global effort to develop a reliable measurement system for nuclear materials, whether for civil or defense applications. The initial effort was to develop very accurate chemical and mass-spectrometric techniques for relatively pure, concentrated uranium and plutonium materials. Approximately 30 years ago, a parallel effort began to develop nondestructive techniques based on measurement of nuclear radiation and heat produced by this radiation. These techniques proved especially useful where rapid, real-time measurements were required on pure materials and where measurements were required of heterogeneous nuclear scrap and waste. A variety of techniques using gamma-rays and neutrons, both passive and active, were developed to meet the requirements of the time.

The world situation that is emerging poses new challenges, changing safeguards requirements and institutions, and new international organizations created to verify compliance with the CTBT. The NPT is 25 years old and the International Atomic Energy Agency (IAEA), which verifies NPT compliance, is nearly 40 years old. The phrase, "IAEA safeguards," will take on an enhanced meaning if Member States accept Phase II of the new safeguards of Program 93+2. Both the Russian Federation and the United States have ambitious programs to dismantle nuclear warheads and store the resulting nuclear materials. The US has offered to place excess defense materials under IAEA safeguards, which results in a very dramatic increase of "direct-use" nuclear material under IAEA verification. In addition, the world is slowly increasing its reliance on nuclear-generated electricity, which further burdens the international safeguards regime. Finally, the cleanup of the weapon's complex has given rise to new requirements for the disposal of nuclear material and the termination of safeguards on nuclear material in waste.

New technologies, like remote monitoring, unattended monitoring, and environmental monitoring, are already being tested to address some of the new problems and they require new sensors and new data analysis methods. To detect nuclear material smuggling, personnel, package, and vehicle monitors at airports and border crossings must handle large traffic volumes with minimal attention except that required to respond to alarms. These monitors must also be able to respond to low signal levels from material that is distant from the sensor and possibly shielded from it.

Residues and waste provide a number of new and interesting challenges to the developers of nondestructive assay instruments and systems. These materials are typically impure, dense, and heterogeneous. Impurities such as Am-241 or fission products when present in sufficient quantities can interfere with passive and active radiation signatures. Because the matrix material in these samples is non-uniform, the assumptions of matrix uniformity underlying established NDA methods are invalid. Non-uniformity is particularly important when the matrix interference is large (e.g. dense materials for gamma-ray assay or highly moderating or absorbing materials for neutron assay). Finally, the increased variety of residue and waste forms is driving the need for generalized, robust assay technology that is capable of compensating for these effects.

The safeguards research program at Los Alamos National Laboratory has developed a number of the nondestructive assay (NDA) techniques that are currently being used for domestic and international safeguards, including neutron coincidence counting, segmented gamma-ray scanning, Cf-252 shuffler, and neutron multiplicity counting. For the last 10-15 years, the program has emphasized the implementation of these techniques. During the past year, we began a search for technical solutions to the new problems facing domestic and international safeguards. While keeping in mind the new user needs, we have hired new scientific staff and begun a broad research program to develop new detectors, new interrogation sources, new data acquisition systems, and new data analysis procedures. This program covers both active and passive measurements and gamma-ray, neutron, and calorimetry techniques. In some cases, we hope to improve existing technology, and, in others, borrow technology from other fields and apply it to nuclear material measurement. We began by asking rather basic questions regarding radiation detection, radiation-matter interactions, and the analysis of radiation data. We hope to make contributions to applied radiation science and, in this way, develop technology to help answer the questions above.

For example, in the past we have looked at applications of room-temperature, semiconductor detectors such as CdTe, CdZnTe, and HgI_2. Their small size, poor resolution, and poor peak shape have limited application to nuclear materials measurements. However, the recent invention of the coplanar grid technique and improved pulse processing methods have enabled the development of medium-resolution CdZnTe with improved efficiency. Consequently, we have begun a program to study the engineering aspects of CdZnTe detectors and to model their response in order to enable the development of detectors that meet our needs.

New scintillation detectors and fast electronics are being studied to develop fast neutron detectors that show promise as coincidence detectors with much improved signal-to-noise ratios as compared to present thermal neutron coincidence counters. We have already successfully developed gamma-ray tomography, primarily to measure heterogeneous residues and waste.[1,2,3] In addition to improving the existing technology, we have begun a study of neutron tomography, which could permit more accurate assays of heterogeneous nuclear material samples. For active assay, we are developing improved interrogation methods and high-intensity neutron sources. In addition, we are developing methods to combine complementary NDA instruments to improve the accuracy and dynamic range of assay systems. While our approach to data fusion is based fundamentally on the physics of the measurements, we are also examining a variety of procedures from information theory and artificial intelligence that could help combine data from different measurement instruments to produce a better measurement than is possible with either one. Selected examples of the new technologies we are developing are discussed in more detail in the body of this paper.

2. Room Temperature Semiconductors

Improvements in materials quality, charge sensing techniques, and pulse processing electronics, are leading to the development of large-volume CdZnTe detectors (between 1 and 5 cm^3) that are suitable for medium-resolution, room-temperature gamma-ray spectroscopy. A primary limitation of CdZnTe, poor hole-collection, has been overcome in large measure by the development of novel electrode configurations, such as coplanar grids, that register only the charge induced by electron motion.[4] The development of coplanar grids has renewed interest in CdZnTe as a substitute for high-resolution gamma-ray spectroscopy for many applications, particularly those where detector size, ruggedness, and maintenance needs are limiting factors (Fig. 1). We are examining the application of these detectors to nuclear materials search, holdup measurements, and gamma-ray tomography.

To predict the performance of the new detectors, we have developed methods to calculate the complete response of semiconductor detectors from first-principles. Both the production and collection of charge carriers are treated. Monte Carlo is used to model the production of charge carriers in the detector by gamma-ray interactions.

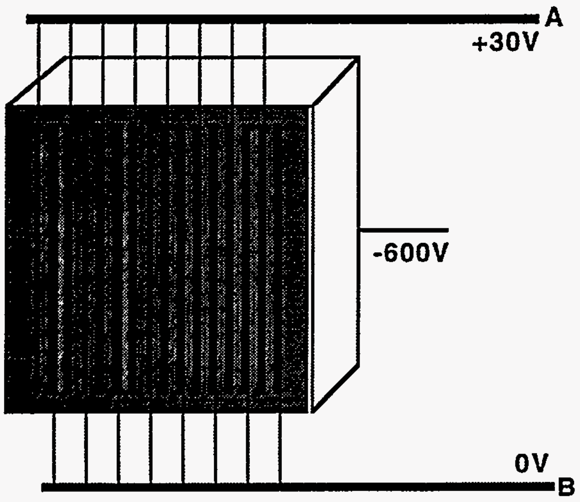

Fig. 1. Example arrangement of electrodes for a coplanar grid CdZnTe detector (from Prettyman, et al.[5]).

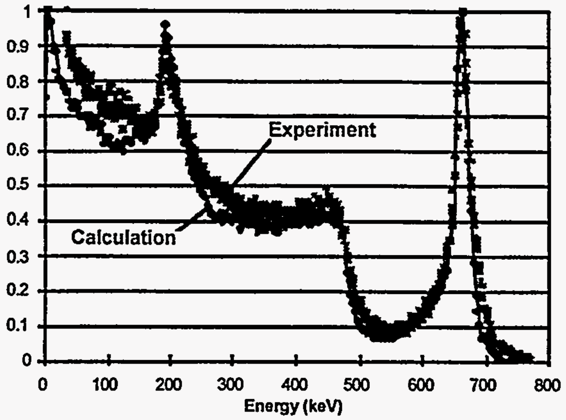

Fig. 2. Comparison between simulated and experimental CdZnTe detector spectra for Cs-137 (from Prettyman, et al.[5]).

Second-order transport effects, such as electron escape, are included in the calculation. Deterministic methods are used to estimate pulse height and rise time for arbitrary detector and electrode configurations. A variant of the Trammell-Walter equation is used to estimate the pulse shape for discrete charge production sites created by individual gamma-rays.[6] A finite elements code is used to model the electric field and electrode weighting potentials[7] needed to predict charge carrier drift and induction efficiency for arbitrary detector and electrode geometry. Charge carrier attenuation by trapping is treated empirically. Pulse-height spectra for a large number of detector configurations can be calculated in a single Monte Carlo simulation, enabling the efficient calculation of the sensitivity of the response to changes in detector parameters.

The combined radiation-transport and charge-collection model can be used to predict the effect of nonuniform trapping and electric fields on detector response and the performance of CdZnTe detectors for nonstandard geometry and electrode configurations. While intended as a tool for detector design optimization and evaluation, the model may also be useful for detector characterization. Comparisons of the model to experimental spectra obtained for planar detectors show that the model can accurately simulate detector response. Figure 2, for example, shows a comparison between simulated and experimental spectra for a Cs-137 source. The detector is one of the first coplanar grid detectors made by eV Products and is currently being evaluated by our group for search and holdup applications.

We are currently using the modeling tool to assist in the design of improved electrode configurations, to design detectors for gamma-ray imaging, to evaluate new methods for pulse-processing, to aid in the determination of CZT materials properties, to evaluate the performance of new detectors for safeguards and nonproliferation, and to develop analytical methods for spectral analysis. For example, we are working closely with Lawrence Berkeley National Laboratory to help evaluate improved electrode configurations for the coplanar grid technique. Figure 3 illustrates the usefulness of the code for evaluating detectors for specific applications. The improvement observed for the large detector (1 mm thick) is a consequence of both increased detector efficiency and improved grid design. Simulations of this kind can be used to determine how well a detector will perform for a specific task and can help identify ways to modify detector design to produce optimal performance.

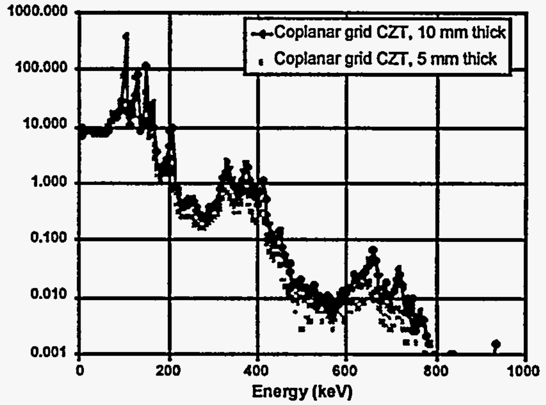

Fig. 3. Simulated low-burnup plutonium spectra (14% Pu-240) for two coplanar grid, CdZnTe detectors (from Prettyman, et al.[5]).

3. Fast-Neutron Multiplicity

A fundamental limitation in the performance of existing thermal neutron coincidence counters is their long neutron die-away time, on the order of 50 µs, which results in severe precision degradation for materials that have high (α,n).[8] Neutron detectors that have a short die-away time, while maintaining high counting efficiency, would enable accurate, high-precision assays of samples that are currently impractical to measure with conventional methods.

A promising candidate for the next generation of neutron detection systems is based on the boron-loaded plastic scintillator, BC454 (available from BICRON Corp.). This material is similar in composition to other organic scintillators with the addition of 5% by weight natural boron (~1% ^{10}B), homogeneously mixed throughout the detector volume.[9] Fast neutrons (E>0.5 MeV) enter the detector and elastically scatter, primarily off hydrogen, slowing down in the process and creating a proton recoil light pulse. Once thermalized, these neutrons can be captured by the ^{10}B(n,α) reaction generating another light pulse, with a characteristic energy deposition of ~93 keV_{ee}.

Capture of the neutron by other nuclides in the detector occurs less than 1% of the time due to the combination of boron uniformity within the detector volume and the large cross section for the ^{10}B(n,α) reaction. The scatter and capture process within the scintillator takes about 2 μs on average. About 94% of the time, the ^{10}B(n,α) reaction populates the first excited state of ^{7}Li, which decays by the emission of a 478-keV gamma ray. The addition of an inorganic scintillator, in our case bismuth germanate (BGO), enables the detection of the 478-keV gamma ray.

Isolation of neutron events is accomplished by utilizing the spectral information inherent in the detector response. A fast coincidence between the 93-keV_{ee} pulse in the BC454 with the 478-keV gamma ray in the BGO identifies a neutron event unambiguously. Further isolation of only fast neutron events can be done by requiring a proton recoil event to precede the capture within a few microseconds. The resulting detection system is one that has a neutron die-away time an order of magnitude shorter than existing counters and the potential for high efficiency.

Figure 4 shows the history of a fast neutron in schematic form. Simulation of the neutron response of an array of these detectors in a well-counter configuration has been performed and indicates that reduction in counting time for high (α,n) samples by a factor of 4 to 10 is achievable in a first generation instrument.[10] Successful implementation of this hardware will benefit assay of both plutonium- and uranium-bearing materials by allowing coincidence gate lengths to be much shorter, thereby greatly reducing the accidental coincidences observed, and by utilizing spectroscopic information not available in thermal neutron counters. We are currently building a prototype well counter employing an array of 10 detectors that will be used in proof-of-principle measurements of items that are currently hard to measure.

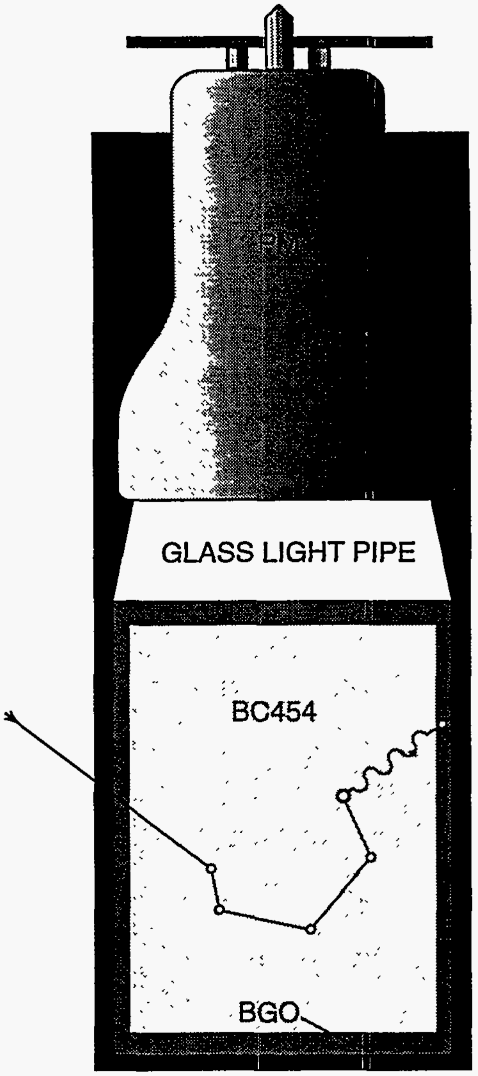

Fig. 4. Schematic history of a fast neutron in the BC454/BGO detector showing the proton recoil and capture process. Capture of the neutron results in a characteristic energy deposition in both the BC454 and BGO elements.

4. New Concepts for Active Neutron Interrogation

The mass determination of HEU (highly enriched uranium) and ANM (alternate nuclear materials) in uncontrolled and unknown configurations will likely become an issue for national and international safeguards. At present no well-established method exists that can be used with confidence for this mass determination when the material is shielded (e.g. HEU surrounded by depleted uranium). We have used active interrogation using high energy photons from a linear electron accelerator (LINAC), or 14 MeV neutrons from a neutron generator, in conjunction with a high efficiency neutron detector to verify the presence of HEU within shielded configurations.

The method relies upon the observation of "correlated" neutrons with a high efficiency neutron detector. These correlated neutrons are from fission events induced by delayed neutrons from fission products. The majority of the fission products result from fission events in the HEU produced by the high energy radiation probe. The correlated neutrons are detected between the high energy probe beam pulses by recording the time of detected neutron events in a data list. A statistical analysis of the neutron arrival time history based upon the Feynman variance technique is performed to extract the correlated neutron signal.[11]

Interrogation of the fissile material using delayed neutrons has a number of interesting applications, including the discrimination between HEU and LEU. Future research will be directed to developing methods to extract quantitative values for the mass, extracting information relevant to the configuration of the HEU such as values of k-effective, or self multiplication, and determining the limits of applicability for the method with respect to lower mass limits, and matrix effects.

5. High Fluence Neutron Source

High-fluence neutron sources can potentially be used to achieve screening criteria for low level and transuranic waste, and to assay material with radiation signatures that interfere with passive neutron measurements, such as spent fuel. In addition, high-fluence neutron sources can be used to increase the sensitivity of prompt gamma-ray neutron activation analysis (PGNAA), which we are currently using for a number of applications, including matrix characterization to verify item descriptions and matrix-corrections in neutron assays. Other applications include fast-neutron radiography and tomography. We are currently developing an intense neutron source using a neutron generator because isotopic sources are difficult use at these levels, particularly for field applications. Present neutron generator systems have short operating lifetimes and produce only 10^8 n/s. Our objective is to develop a system that produces at least 10^{11} n/s in roughly the same size, weight, and power consumption as existing 10^8 n/s systems.

Nondestructive assay instruments that require high neutron fluence typically use isotopic californium sources. Isotopic sources, however, cannot be turned off and are sometimes inconvenient to use when high fluence is required because the shielding can be cumbersome, particularly for mobile or field applications. A more effective alternative would be a neutron generator that would operate only during measurements. Unfortunately, existing neutron generator systems have generally low output (for a reasonable site, weight, and cost) and have short operating lifetimes. Normally these systems last only 500 hours. An effective system would require an operating lifetime of 10,000 hours to be reliable in a field application. The neutron yield would have to be raised to 10^{11} n/s to improve speed and proximity. The system should have a size, weight, cost, and power requirement comparable to present 10^8 n/s neutron generator systems. The target operating parameters for an effective neutron generator system are:

Parameter	Value
Neutron Yield	10^{11} neutrons/second
Operating Lifetime	10,000 hours
Size	1 60 cm sphere and 1 19 in. rack of power and control electronics
Power Required	< 10 kW

We are developing a neutron generator with these characteristics based an the Internal Electrostatically Confined (IEC) plasma technology. The IEC concept was originally developed in the 1950s as a possible fusion candidate for the production of electrical power. In the 1960s a peak output of 2×10^{10} n/s was achieved on a table-top experiment.[12] The idea has been only modestly pursued; IEC devices were not expected to achieve fusion power energies. However, it works nicely as a neutron generator. Experiments at the University of Illinois have already demonstrated very long operating lifetimes >1,000 hours and modest size and power requirements.

The IEC plasma neutron generator is a deceptively simple device. It consists of a spherically symmetric vacuum chamber containing a spherical grid. The grid is held at high negative potential (~80 - 100 kV). A plasma forms between the grid and vacuum chamber. The positive ions are accelerated toward the center, where they collide. In a deuterium-tritium (D-T) plasma at sufficient energies, the colliding ions fuse to produce ^{4}He and neutrons.

A fundamental result of the early research by Hirsch and the subsequent research at the University of Illinois, INEL and Los Alamos, is that the neutron yield improves as the plasma density is lowered. The reason is that at high density the plasma is collisional. Accelerated ions are more likely to collide with background neutrals rather than with other accelerated ions at the plasma core. Two effects reduce the fusion yield: the ions collide multiple times and never achieve the full accelerating potential energy, and ions collide with essentially stationary neutrals rather than with other accelerated ions. The result is that the center of mass energy can be far below the accelerating potential.

In a lower-density plasma, ions would be more likely to avoid collisions with background neutrals and would collide at the plasma core, where the local density peaks. The center of mass collision energy would increase as would the neutron yield. However, an arbitrarily low density is prevented by Paschen's Law and for nominal, single-grid IEC geometry and accelerating potential, the IEC operates in the collisional regime.

A novel Los Alamos design circumvents the Paschen limit in the IEC. The Los Alamos IEC uses a triple grid design.[13] Each grid is spherical as in the single grid system. The inner most of the 3 grids serves the same function as the single grid: it is at high potential. The central grid provides shielding. The outer grid is held at modest positive potential (~200 volts) and with electron injectors creates a plasma at a lower density as on the Paschen curve. The ions drift toward the center and are accelerated as before. The triple grid design enables a plasma breakdown at low density. The plasma is collisionless, the collision energy increases, and the neutron yield scales up. Figure 5 is a schematic of a triple grid IEC device.

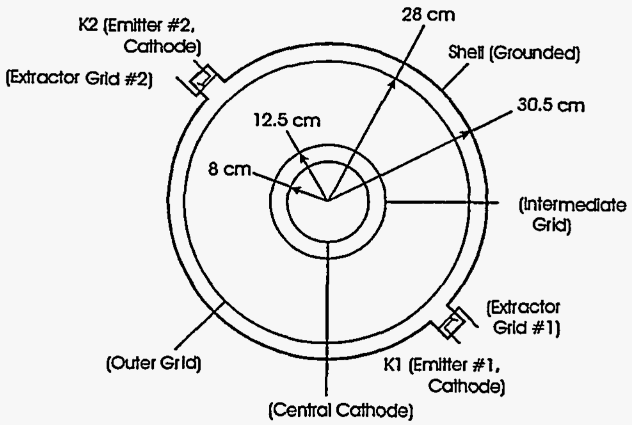

Fig. 5. *Schematic of a triple grid IEC device.*

This concept has been tested using a fully kinetic, 1.5 dimensional, plasma simulation code at Los Alamos. The code also implements full atomic physics. These simulations indicate that the neutron yield does indeed scale up using the triple grid design and low density. The target neutron rate of 1 x 10^{11} n/s seems readily achievable. Figure 6 compares the density profile for both a single grid and a triple grid design.

Los Alamos has designed a triple-grid IEC device to test this concept and a 3-year experimental program to test and evaluate the technology is underway. The experimental IEC generator is currently under construction and will be completed shortly. This system will be used to benchmark the plasma simulations and to experimentally verify the scaling relations. Based on these results, a second chamber will be designed, built and tested. The system operation will be optimized for maximum yield while meeting a minimum power consumption requirement. The final system technology would then be transferred to an industrial partner.

6. Data Fusion and Information Analysis

Data fusion is a promising approach for rapidly extending existing nondestructive assay capabilities to measure materials for which individual instruments perform poorly. The intent is to combine disparate measurements from different instruments into a single result. The assumption is that complimentary techniques more reliable assays and diagnostic information needed to assess the validity of the results. Data fusion also has obvious extensions to unattended monitoring, where different types of sensors are used to ensure the reliable detection of anomalous events.

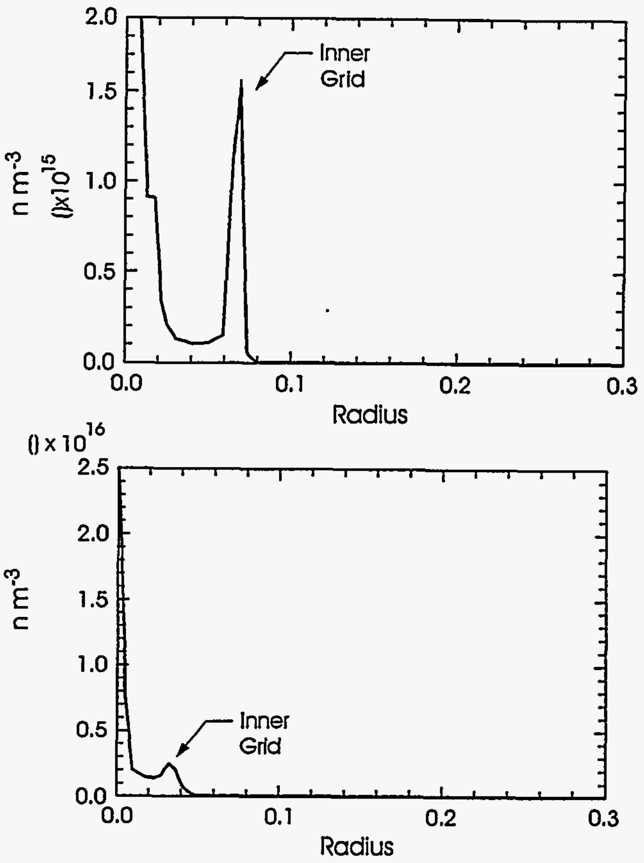

Fig. 6. Top plot is the radial density profile from simulations for a single grid IEC device. Bottom plot is for a triple grid IEC device. Note that the triple grid design more effectively peaks the density at the device center and that the density is lower at the accelerating grid. These simulations show that a neutron yield of 10^{11} n/s D-T is possible.

We are taking a systems approach to data fusion for nondestructive assay and unattended monitoring. Sensor and instrumentation selection is based on an analysis of the physical principles underlying their performance. This information is used in the overall analysis and design of the integrated system. With the systems approach, redundant information can be eliminated and the information needed to accomplish a task can be obtained while minimizing the complexity of the instrumentation. Whenever possible, algorithms based on physical principles are used to maximize the robustness and the range of applicability of the assay.

For example, the accuracy of active neutron assays (Cf-252 Shuffler) of drum-sized samples containing heterogeneous scrap and waste can be improved significantly if the location of the nuclear material is known. Bias as a function of source position for uniform matrices is well-known.[14] Consequently, the assay can be corrected if the position of the emitting material is known. Position information can be obtained in a number of ways, including passive neutron imaging.[15,16,17] Emission images from a tomographic gamma scanner can also be used directly by the position-correction algorithm. Tomographic gamma-scanning has the advantage of providing high-resolution position information as well as information about the geometry of the sample and matrix, such as fill-height, that could influence the result of the neutron assay. In addition, tomographic gamma scanning also provides an independent estimate of the amount of fissile material.

We are also developing data-fusion methods that work effectively when a direct approach to combine different data sources is not possible. For example, we used a statistical method called the Alternating Conditional Expectation (ACE) algorithm to combine data (from He-3 tubes and flux monitors) obtained by the californium shuffler. (ACE is in a class of algorithms called generalize additive models that are similar to neural nets.) The result was a significant improvement in the accuracy of the measurement.[17]

We are currently applying information analysis methods, developed by the Safeguards Systems group, to combine sensor and instrument data. Areas being investigated include neural networks, data mining techniques, pattern recognition, anomaly detection, and expert systems. Possible applications of the information analysis methods to nondestructive assay include integration of tomographic gamma scanning with neutron multiplicity counting for residue assay, integration of neutron imaging data with active neutron assay, and the use of prompt gamma-ray information to identify sources of matrix interference for neutron assay.

7. Summary

We have outlined four new areas of research being conducted by the Safeguards Program at Los Alamos National Laboratory:

1. Modeling of room-temperature semiconductors,
2. Investigation of fast-multiplicity methods,
3. Development of high-fluence, plasma-based neutron sources,
4. Data-fusion and information analysis.

A number of other new topics are currently being investigated, including neutron imaging and tomography and improved methods for calorimetric assay. Our objective is to improve safeguards instrumentation and practices through the development and implementation of new technology.

Because of their small size and ruggedness, room-temperature semiconductor detectors with medium energy-resolution show considerable promise for search and holdup applications. Fast-multiplicity methods are promising because they can provide increased sensitivity and throughput over existing multiplicity counters, enabling assays of materials for which the fission signature is obscured (for example, by (α,n) neutrons from Am-241 decay). New information will become available for safeguards measurements, including prompt gamma-ray signatures and fast-neutron images, if the work on high-fluence neutron sources is successful. Research on inertial electrostatic confinement could lead to rugged, low-cost neutron generators with very long operation lifetimes that output 10^{11} n/s. Finally, the development of data fusion and information analysis methods, including analytical methods for neutron and gamma-ray imaging, could lead to generalized assay systems that could be applied to safeguard the increasingly disparate materials encountered in facilities world-wide.

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