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RADIATION IMAGING TECHNOLOGY FOR NUCLEAR MATERIALS SAFEGUARDS

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Radiation Imaging Technology for Nuclear Materials Safeguards*

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Abstract

Gamma-ray and neutron imaging technology is emerging as a useful tool for nuclear materials safeguards. Principal applications include improvement in accuracy for nondestructive assay of heterogeneous material (e.g., residues) and wide-area imaging of nuclear material in facilities (e.g., holdup). Portable gamma cameras with gamma-ray spectroscopy are available commercially and are being applied to holdup measurements. The technology has the potential to significantly reduce effort and exposure in holdup campaigns; and, with imaging, some of the limiting assumptions required for conventional holdup analysis can be relaxed, resulting in a more general analysis. Methods to analyze spectroscopic-imaging data to assay plutonium and uranium in processing equipment are being developed. Results of holdup measurements using a commercial, portable gamma-cameras are presented. We are also developing fast neutron imaging techniques for NDA, search, and holdup. Fast neutron imaging provides a direct measurement of the source of neutrons and is relatively insensitive to surroundings when compared to thermal or epithermal neutron imaging. The technology is well-suited for in-process inventory measurements and verification of materials in interim storage, for which gamma-ray measurements may be inadequate due to self-shielding. Results of numerical simulations to predict the performance of fast-neutron telescopes for safeguards applications are presented.

Introduction

The need to assay distributed deposits of nuclear material in processing equipment (e.g., glove boxes, pipes, and tanks) arises in routine plant operations, in research, and in facility decontamination and decommissioning. To address this need, extensive effort has gone into the development of portable or semiportable radiation measurement systems, including hand-held sodium-iodide detectors and multichannel analyzers (such as the miniature, modular multichannel analyzer developed by Los Alamos). Despite the advanced state of portable equipment, the success of efforts to quantify nuclear material (e.g., holdup) depend largely on careful planning on the part of the facility. Implementation of a holdup campaign can require a significant effort by measurement technicians. The accuracy of the measurements depends on the validity of assumptions made about both the distribution of the nuclear material and the construction of the processing equipment. Although the track record for DOE facilities is excellent, the risk of injury to personnel or radiation exposure or contamination is nontrivial in these operations.

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Radiation imaging technology, particularly for gamma-ray imaging, is now widely available and could aid in measurements of nuclear deposits and in planning of measurement campaigns. Portable gamma-ray imaging systems are available commercially. In addition, compact medical cameras are available at low cost from a number of suppliers. Commercial gamma-ray cameras are largely based on scintillator technology (e.g., NaI, CsI, and BGO) and resolve position using a variety of methods, including position-sensitive photo-tubes and multiple photo-tubes with Anger electronics. However, a few solid-state cameras are available, and the emergence of CdZnTe, a material that is well suited for gamma-ray spectroscopy at room temperature, a new generation of portable gamma-ray imaging systems is anticipated. Nevertheless, with existing cameras, excellent position resolution is achieved with performance for spectroscopy equaling or rivaling established hand-held detectors. Consequently, the possibility for low-cost spectroscopic imaging of gamma-rays has been realized.

Neutron imaging technology is also mature; however, neutron imaging systems are not widely available commercially. Neutron imaging systems have been developed for many applications, including nondestructive evaluation (neutron radiography and computerized tomography), astrophysics, and nuclear physics research. Recently, an imaging system for thermal and epithermal neutrons (<100 keV) was developed by Brookhaven National Laboratory for safeguards and nuclear nonproliferation applications.[1] At Los Alamos, we are developing instrumentation to image fast neutrons for both nuclear nonproliferation and space applications (>1 MeV).

In principle, radiation imaging offers a number of advantages that could be exploited by facilities in holdup measurement campaigns or in decontamination and decommissioning. For example, in the planning stages, imaging technology could be used to perform wide-area surveys of equipment for which process information is inadequate or incomplete. The location of significant deposits could be identified. Because wide-areas can be covered in a single measurement, the effort required by personnel to assimilate the information needed to locate and characterize deposits could be reduced. This would reduce the dose received by personnel and their risk of injury. The use of imaging technology also has the potential to eliminate sampling errors associated with conventional, single-detector holdup measurements. In addition, assumptions regarding the distribution of material (e.g., point, line, or plane) can be relaxed because imaging systems measure this parameter directly.

At Los Alamos, we are investigating the application of both gamma-ray and neutron-based imaging technology to nuclear material safeguards. We are currently evaluating commercially available gamma-ray imaging technology, including portable systems developed specifically for radiation surveys and medical cameras. Simultaneously, we are developing analytical methods for spectroscopic imaging that will provide accurate, position-dependent assays of nuclear material. We are also conducting research on fast-neutron imaging techniques. Fast neutrons are less affected by surrounding materials than thermal and epithermal neutrons and are more penetrating than gamma-rays. Consequently, assays based on fast-neutron measurements are less susceptible to bias due to attenuation by intervening material. In addition, the location of the source of fast neutrons can be determined accurately. A fast-neutron imaging system could be developed to image deep deposits of nuclear material or nuclear material with significant gamma-ray shielding.

Gamma-Ray Imaging

Gamma-ray imaging systems are available at a relatively low cost from a number of commercial suppliers. For example, we are currently evaluating a portable gamma camera from RMD, Inc., that is capable of combining gamma-ray and video images with a wide field of view (nominally 45°). Images are formed using a tungsten pinhole and a scintillator (BGO or CsI, 5- to 10-mm thick) coupled to a position-sensitive photomultiplier tube (~7.5 cm in diameter). The camera can resolve sources that are separated by less than 5°; however, because a pinhole is used, the efficiency of the camera is relatively low and varies appreciably as a function of position. The efficiency for 662 keV gamma-ray (from ^{137}Cs) was found to be 2×10^{-6} counts/gamma-ray at a distance of 1.5 m.

An image of an array of pipes containing plutonium (~6% ^{240}Pu) obtained using the RMD camera is shown in Fig. 1A. The total mass of plutonium in the array was 60 grams. The measurement was made using BGO (~25% FWHM at 662 keV) with a region of interest set about the 400-keV complex. The acquisition time was 30 minutes. The distance from the camera to the pipe array was 1.5 m.

The intensity of gamma-ray emission (contours) is overlaid on the video image. Figure 1B shows the gamma-ray emission map after it was corrected for a number of geometric factors (the pinhole acceptance function and the solid-angle of the pinhole subtended by the source) and for room background (measured separately by placing a shield in front of the pinhole). The source was assumed to lie in the plane of the pipes. The corrected image shows the location of the five plutonium standards used in the experiment. An assay of each source was obtained by summing the corrected counts in regions of interest containing the source (defined by boxes).

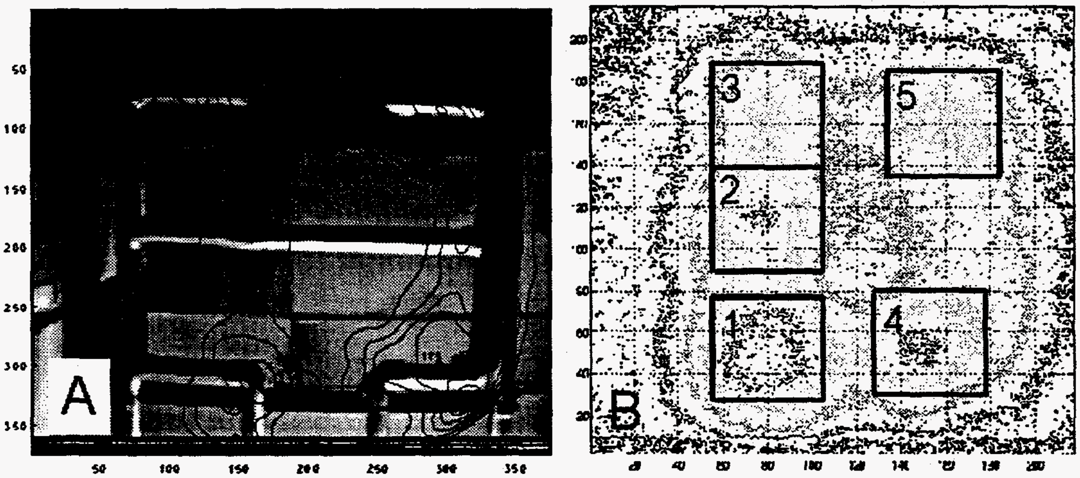

Fig. 1. Gamma-ray and video images of a pipe-array containing plutonium (A) and processed camera data (B).

Assays of the sources derived from the gamma-camera data are compared to results obtained with a portable, collimated NaI detector (Table 1). The relative assay values are essentially equivalent. Only two of the measurements differ by more than 5% (i.e., sources 1 and 2). We estimate that it would take at least an hour for a skilled measurement technician to assay the pipe array using a hand-held instrument assuming the location of the plutonium was unknown. In making this estimate, we have considered the amount of time required for setup, locating the hot-spots, making the measurements, and analyzing the results.

Table 1. Assays of Plutonium Sources in the Pipe Array Using the RMD Camera (spectroscopic gamma-ray image) Compared to Results Obtained Using a Collimated Detector (collimated gamma-ray spectrum). The results for the Collimated Detector are used as a Reference.

Pu Source ID	^{239}Pu Assay Data (counts)		ASSAY ÷ REFERENCE*
	2-d Spectroscopic γ-Ray Image (TEST ASSAY)	Collimated NaI γ-Ray Spectrum (REFERENCE)	
1	19357	275	0.77
2	16039	148	1.18
3	12740	138	1.01
4	15869	171	1.01
5	13970	147	1.04
		mean =	1.00
		1σ =	0.15

* NORMALIZED

Neutron Imaging

At Los Alamos, we are investigating neutron imaging techniques that could be used in a variety of applications, including holdup measurements. Our focus is on fast-neutron imaging. Fast neutrons are highly penetrating and are less affected by surrounding materials than thermal or epithermal neutrons or gamma rays. Thermal or epithermal imaging, for example, is in many cases more sensitive to moisture in the surroundings (such as water in pipes) than the original source of the neutrons. In principle, fast neutron imaging can be applied to directly locate and assay neutrons produced by spontaneous or induced fission, even in deep, distributed deposits embedded in heterogeneous matrix material.

Because fast-neutrons have a large mean-free-path in most materials, they are difficult to collimate. As a result, physical collimation is very ineffective for fast neutron imaging. We are investigating several alternatives to achieve fast-neutron imaging with high spatial (or angular) resolution, including the neutron telescope technique, active collimation, and temporal multiplexing.

Our approach to investigating these techniques involves the development of detailed Monte Carlo simulations that accurately represent the performance of a full-scale, well-engineered system. Parameters used in the Monte Carlo simulation, such as the performance of photomultiplier tubes and scintillators, are determined from bench-top experiments. This approach enables us to determine the optimal parameters for a device well in advance of constructing a full-scale prototype. Because the simulations are accurate, the cost-savings that can be achieved using this approach are significant.

Most recently, we have begun to investigate the feasibility of applying fast-neutron telescopes to actinide imaging and assay. The concept is illustrated in Fig. 2. A fast neutron (e.g., from fission) suffers a single elastic collision with hydrogen in the first detector (a plastic scintillator, e.g., BC400) and drifts to the secondary detector, where it interacts again by elastic scattering. The light produced by the recoil proton in the primary detector is used to determine the energy lost by the incident neutron. The time-of-flight between the primary and secondary detectors provides the energy of the neutron after scattering. The position of both interactions is recorded. The information recorded by the instrument is sufficient to localize the direction of the incident neutron to the surface of a cone and to determine its energy.

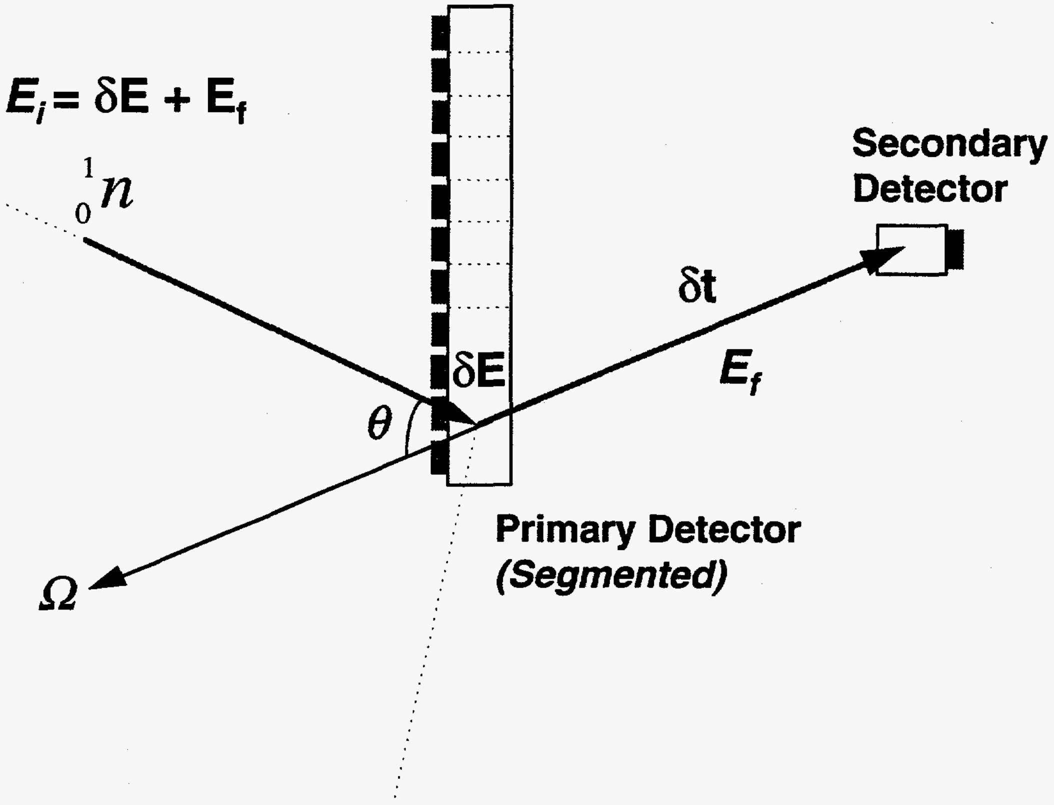

Fig. 2. Neutron telescope concept.

An image is formed by projecting the cones onto a hemispherical surface. An example of this process is shown in Fig. 3 for a point source of monoenergetic neutrons (2.45 MeV) located 2 m directly in front of a neutron telescope. Note that the coordinates of points in each image give direction. In principle, the telescope can form 2π images (a half-space). The superposition of curves formed by the intersection of the cones and the hemisphere is shown in Fig. 3A for a few events. The source is located at the point where the curves intersect. After many events, the source is well-resolved (Fig. 3B).

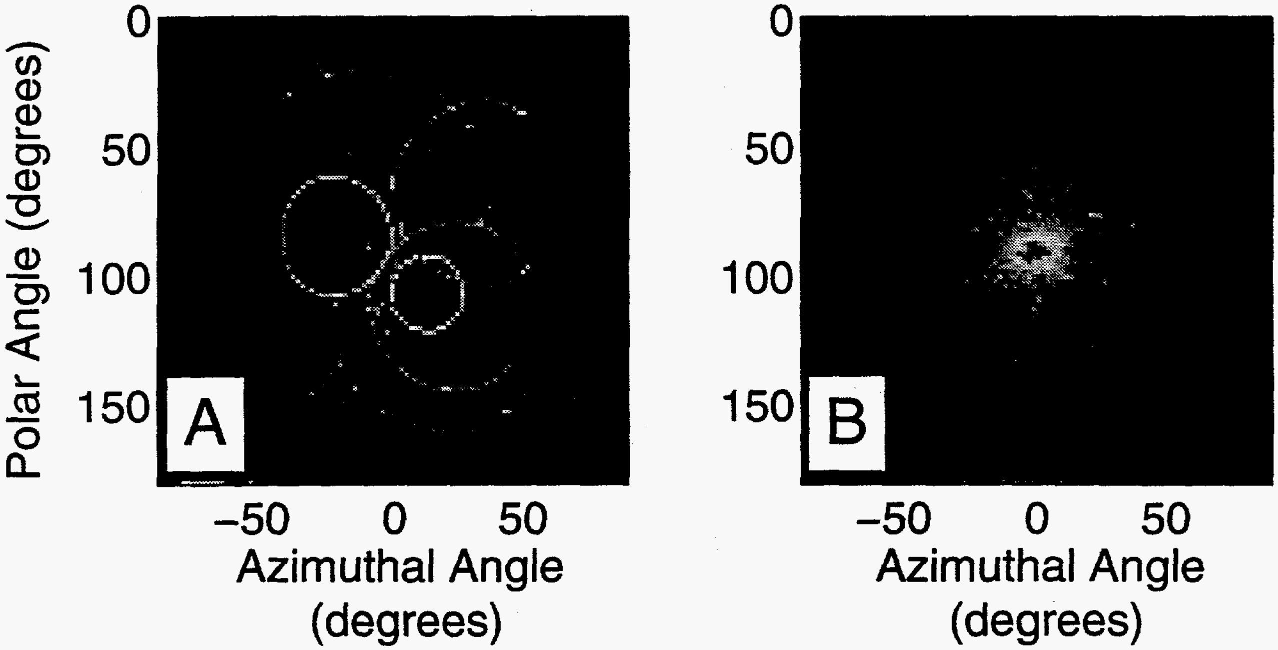

Fig. 3. Reconstruction of a point source with a few events (A) and many events (B).

Telescope Simulator

The performance of fast-neutron telescopes depends on the pulse-height resolution of the primary detector (approximately 18% full-width at half maximum (FWHM) at 1 MeV_{ee} for photomultiplier tubes (PMT) and 11% FWHM for avalanche photodiodes (APD)), the timing resolution of the system (better than 500 ps is possible for a detection threshold of 30 keV_{ee}), and the position resolution of the detectors. At present, we can simulate the response (light-output, position-, and timing-resolution) of individual detector elements, rods, and Anger cameras. Performance also depends on transport effects such as carbon scattering, which causes blurring. All of these parameters are simulated in detail by a specific-purpose Monte Carlo code which includes general three-dimensional geometry and a detailed treatment of neutron transport physics. The nuclear data used in the calculations are drawn from the evaluated nuclear data files (ENDF/B-VI).

A calculational study was initiated to explore the performance of a fast-neutron telescope for imaging neutrons from fission. The telescope geometry was restricted to parallel slabs consisting of arrays of rods or single-detector elements. The distance of separation between the primary and secondary detector slabs was varied to optimize telescope performance. Intervening materials such as tin or cadmium foil (for x-ray shielding) and the PMT or APD assemblies were included in the simulation.

Rods were initially eliminated from consideration because their position-sensitivity may not be sufficient given the light produced by fission neutrons. However, some experiments are being conducted to see if position sensitivity can be improved by roughening the surface to increase light-loss across the rod. The use of rods could be advantageous over arrays of single detector elements because fewer PMTs (or APDs) are required.[2] All of the simulations reported in this paper assume that position is determined using arrays of single-detector elements. A useful size for cylindrical detector elements was determined to be 5 cm in diameter by 2.5 cm thick. At this thickness, the detection efficiency is high and most of the coincidence events involve single-scatters with hydrogen, a necessary condition for imaging. A 900-cm^2 slab could be made using 45 elements packed in a hexagonal array.

Gamma-Ray Rejection Capability

An important characteristic of the neutron telescope is its ability to reject correlated events that are not caused by fast neutrons. Interference and blurring due to cosmic rays, accidental and correlated n–γ, and γ-γ events can be eliminated by time-of-flight discrimination. Fast, correlated events (such as gamma-ray double scattering) are eliminated using a lower level discriminator (at ~500 ps). Slow, correlated events (for example, caused by thermal neutron capture in hydrogen) are eliminated using an upper level discriminator (~20 ns). Coincidences caused by correlated fission neutrons and gamma-rays are expected to be minimal because the telescope is decoupled from the source. Because the event-pair resolving time of the telescope is very small (<50 ns) and gamma-ray coincidences can be rejected with high efficiency, very high count rates can be measured in a relatively high gamma-field. Consequently, the telescope may be effective for applications with high radiation fields (for example, remote handled transuranic waste). The major limitation of the telescope is dead-time caused by event processing (reconstruction in software or hardware).

Results

Telescopes are, in principle, excellent neutron spectrometers because the light-energy relation can be inverted for each event and the energy of the scattered neutron is well-known (via time-of-flight). The light-output (pulse-height) distribution of the primary detector is shown in Fig. 4 for a monoenergetic source of neutrons along with reconstructed neutron energy distributions (appearing as Gaussian-shaped functions) for two telescope configurations. The distance between the primary and secondary detectors was varied. The energy of the incident neutrons is inaccurate because a power law has been assumed for the light-energy relationship in the reconstruction algorithm. A more accurate model that treats the nonlinear variation of light output at low proton energy was used in the event simulator. The light output in the scintillator drops off very rapidly for low-energy protons and is inaccurately modeled by a power law. Inclusion of this effect in the reconstruction algorithm should improve both the resolution and accuracy of the telecscope, both for spectrometry and position.

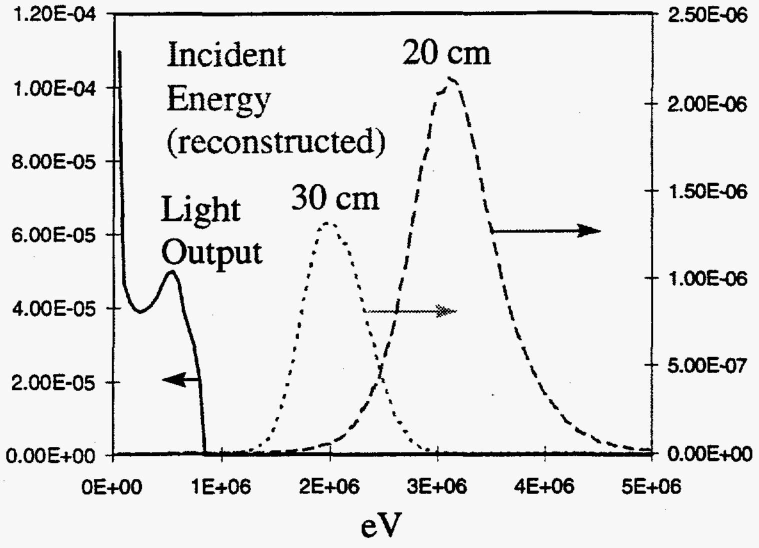

Fig. 4. Reconstructed pulse-height spectra for a monoenergetic (2.45 MeV) source of neutrons for two different telescope configurations (20- and 30-cm separations). The light-output spectrum for the primary detector is shown for comparison.

The variation of the efficiency of the telescope with energy is shown in Fig. 5. The efficiency is maximum between 3 and 4 MeV. Below 2 MeV, efficiency drops off rapidly due to decreased light output. A fission spectrum is shown for comparison. The variation of coincidence efficiency (coincident events per incident neutron) with direction for a fission spectrum is shown in Fig. 6. The coincidence efficiency is very uniform out to about 45°, where it drops off rapidly. Consequently, the useful field-of-view of the telescope is greater than 90°. The FWHM of a point fission source placed in the field-of-view was found to be less than 15°. Use of a more accurate model for the light-energy relationship in the reconstruction algorithm should improve resolution. Resolution is also a function of the separation between the primary and secondary detector (due to uncertainties in flight time), a parameter that can easily be adjusted in a well-engineered telescope.

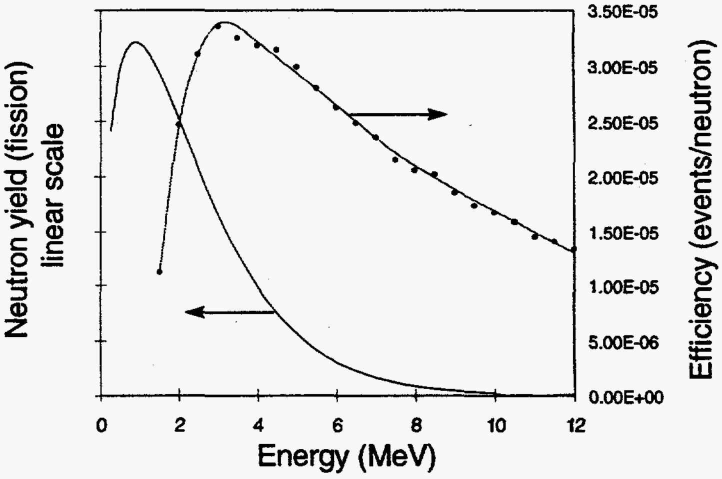

Fig. 5. Efficiency as a function of energy for an optimized telescope.

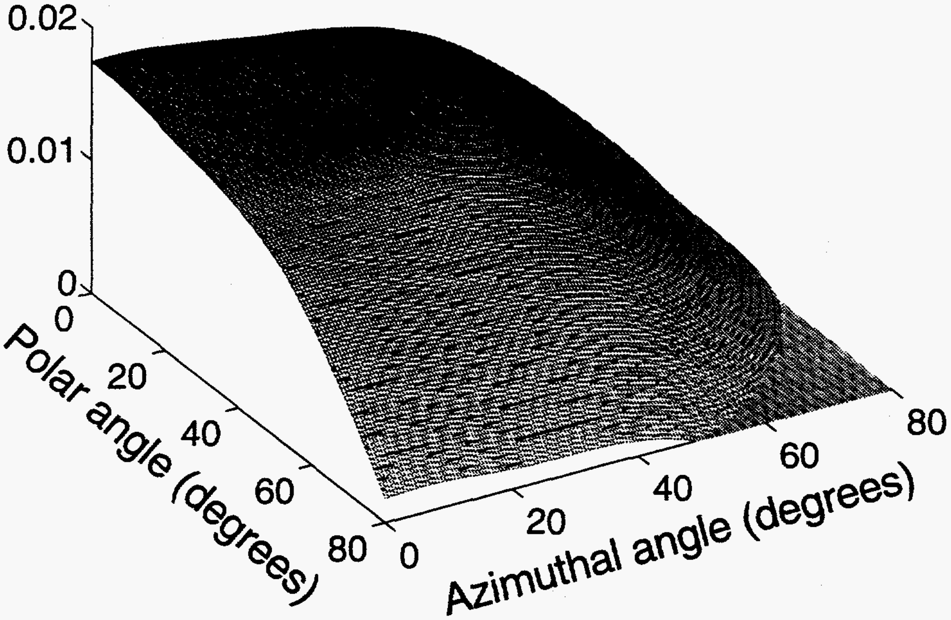

Fig. 6. Coincidence efficiency as a function of angle.

To test the ability of the telescope to image deep material deposits, we simulated two fission sources embedded in a concrete wall with 10% moisture content. The geometry of the experiment is shown in Fig. 7A. The source closest to the telescope (1) was embedded 10 cm within the concrete. The other source was 4 cm deep. The image of the sources formed by the telescope is shown in Fig. 7B. Both sources are clearly resolved. The second source appears less intense because it is more distant from the telescope.

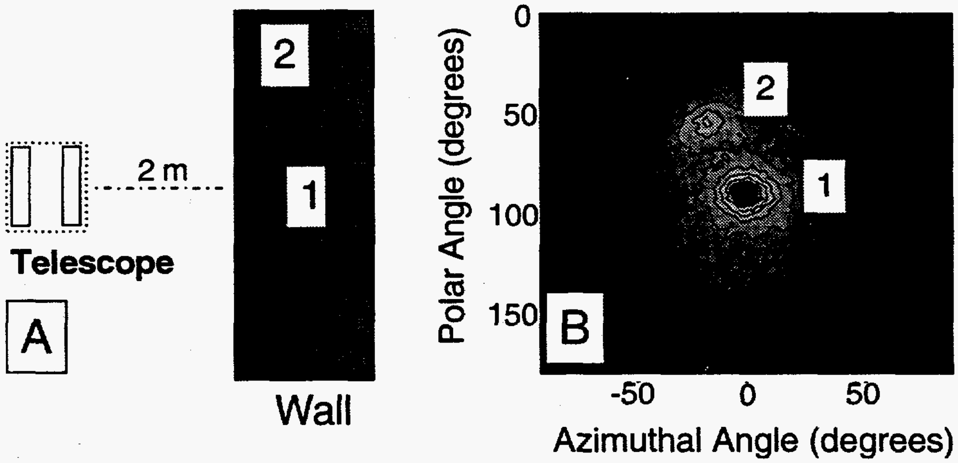

Fig. 7. Reconstruction of a distributed fission source in a concrete wall.

To summarize, the detection efficiency for sources in the field of view of the telescope is on the order of $3x10^{-5}$ events per source neutron for fission neutrons at 2 m from the telescope. This means that 1 g of ^{240}Pu could be imaged by the telescope in about 60 minutes (> 100 events). This corresponds to 20 g of weapons-grade plutonium. Imaging of uranium is also possible. The telescope is insensitive to neutrons below 1 MeV. Consequently, an AmLi source could be used to illuminate uranium deposits. The directional resolution achieved by the telescopes simulated in this study was better than 15° over a field-of-view that exceeds 90°.

An engineering design study will be conducted to further optimize telescope performance. Physical parameters such as the separation between the primary and secondary detectors and the size, geometry, and composition of the detector elements influence telescope performance (spatial-resolution, energy-resolution, field-of-view, and efficiency). These parameters will be examined using the telescope simulator code. The validity of the code will be tested against neutron double-scatter experiments. Improvements in the reconstruction algorithm, such as the addition of a model that accounts for the nonlinear variation of the energy-light relation for low-energy protons, will also be investigated.

Summary and Conclusions

The results obtained by the RMD camera for the pipe array serve to illustrate potential advantages of gamma-ray imaging technology over the hand-held detector technology currently employed at all major nuclear material facilities. The advantages include the following:

- with gamma-ray imaging, sampling errors are eliminated. It is difficult to overlook a nuclear material deposit, provided it is in the field-of-view of the camera and is unshielded;
- unless the entire field-of-view is filled with nuclear material, room-background can be obtained directly from regions of the gamma-ray image that do not contain sources. This feature should significantly reduce the setup time required for single detectors; and
- assumptions regarding the distribution of nuclear material are not needed; however, as with conventional methods, knowledge of the distance to the source, the attenuation coefficient and geometry of the intervening material, and the thickness and composition of the deposit is required for accurate results.

At present, a truly portable camera with all of the features needed for nondestructive assay of distributed deposits of nuclear material do not exist; however, all of the necessary technology, including video cameras and range finders, are available off-the-shelf. Some research is needed to optimize the design of commercial cameras (efficiency, field-of-view, and resolution) for plutonium and uranium measurements. In addition, efficient algorithms that can accurately assay nuclear material deposits using spectroscopic images need to be developed.

Fast neutron imaging, using the neutron telescope technique, appears to be a reasonable alternative to gamma-ray imaging technology for nuclear material measurements. Detailed, Monte Carlo simulations of neutron telescope measurements show that deposits of nuclear material can be imaged using fast neutrons. In addition, telescopes are excellent neutron spectrometers. The efficiency and resolution of the telescope are sufficient for a wide range of applications. In addition, fast neutron imaging provides a direct measurement of the source of neutrons and is relatively insensitive to surroundings. The technology appears to be well-suited for in-process inventory measurements and verification of materials in interim storage, for which gamma-ray measurements may be inadequate due to self-shielding.

References

1. P. E. Vanier and L. Forman, "Advances in Imaging with Thermal Neutrons," *Institute of Nucl. Mater. Manage., Nucl. Mater. Manage.* **XXV**—CD ROM (Proceedings Issue) 1151-1155 (1996).

2. R. C. Byrd, A. Gavron, M. J. Leitch, et al., "An Imaging Detector for Neutron or Gamma Sources," Los Alamos National Laboratory report LA-11612-MS (1989).