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New and Novel Nondestructive Neutron and Gamma-Ray Technologies Applied to Safeguards

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TOPIC: New and Novel Safeguards Technologies

New and Novel Nondestructive Neutron and Gamma-Ray Technologies Applied to Safeguards

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The nuclear fuel cycle of the future will present us with new safeguards challenges. In this paper we will discuss several new technologies developed at LLNL that could be applied to safeguards. These are: fast neutron counting with list mode data acquisition, superconducting high resolution gamma spectrometer, compact solid state thermal neutron detectors, and a spectroscopic 3D gamma-ray imager with laser range scanner.

We will discuss a new segmented liquid scintillator multiplicity counter with nanosecond timing, which has 10^{-5} discrimination of neutrons and gamma-rays above 500 keV. Used passively, this fast neutron counter can detect isolated fission chain bursts in the presence of high backgrounds, such as in storage areas or where the signal is dominated by (alpha,n) or spontaneous fission backgrounds. It will be useful in quantifying total fissile mass. Used actively, in conjunction with 60-keV neutron interrogation to induce fission, it detects the high-energy fission neutrons and is blind to the interrogating beam. The 60keV beam energy selectively fissions ^{235}U and ^{239}Pu and not ^{238}U or ^{232}Th. This technology will be useful for higher count rate applications than traditional multiplicity counters.

We will also discuss a superconducting high resolution gamma spectrometer or microcalorimeter for ultra-precise analysis of low-energy gamma rays and X-rays. Superconducting gamma-ray spectrometers offer an order of magnitude higher energy resolution than conventional high-purity germanium detectors. This increases the precision of isotope ratio measurements in complicated mixtures that are affected by line overlap. We have developed gamma-ray detectors based on superconducting Mo/Cu multilayer sensors and attached bulk Sn absorbers. They have, depending on design, achieved an energy resolution between 50 and 100 eV FWHM at 100 keV, with count rate capabilities between up to 100 counts/s per pixel. We have also developed refrigeration technology for user-friendly detector operation at 0.1K. The detector can greatly improve the precision of destructive analysis (DA) as well as nondestructive analysis.

LLNL is currently develop configurable, real-time, low-power, compact, solid state neutron detection system for special nuclear materials neutron signature detection with 10% thermal neutron detection efficiency. This neutron detection system can be used for long term monitoring of nuclear material in storage, or it could be an added feature in a tag or seal. These neutron detection systems can also be used for detecting or monitoring neutron emissions in areas (e.g., under water, pipes, etc.) where there are complex scattering environments, no available power, and little or no human access. There are two configurations in the neutron detector, namely, ^{10}B for thermal neutron and $^{6,7}LiF$ for higher energy neutrons, the ^{7}Li is for normalization.

Finally, we will discuss a spectroscopic 3D gamma-ray imager that, combined with a laser range scanner could be used for design information verification system and nuclear material accountability applications. The Compact Compton Imager (CCI) currently being developed by LLNL has the potential to greatly improve Materials Accountability through enabling an automatic, efficient and more accurate real-time holdup and material accumulation measurements in bulk facilities across the nuclear fuel cycle (enrichment, fuel fabrication, and reprocessing) for international safeguards. With this instrument, we have recently demonstrated unprecedented imaging resolution, sensitivity and field of view, as well as 3D imaging capability. These features combined with an excellent spectroscopic resolution, create an instrument with unique capability for automatically mapping nuclear materials. This system can also enhance the current technology for Design Information Verification.

INTRODUCTION

The nuclear fuel cycle of the future presents many technological challenges both in the near term, to determine the expected characteristics of the spent fuel and process monitoring, and in the long term, safeguards. A specific challenge is how much fissionable material is present in a particular sample, production line or storage area. At different stages in the processing, how much SNM (^{235}U and ^{239}Pu/^{241}Pu) is left in the spent fuel from the mixed oxide fuels such as in GNEP? Is the expected amount of SNM in the production line, storage area, or waste lines? How can we prove that none of the SNM has been diverted? In this paper we will discuss two new technologies developed at LLNL that could be applied to safeguards as well as GNEP. These are: 1) fast neutron counting with automated analysis, and 2) superconducting high resolution gamma spectrometer, 3) compact solid-state thermal neutron detectors and 4) spectroscopic 3D gamma-ray imager. New analysis methods will be briefly discussed.

COUNTING OF FAST NEUTRONS

Multiplicity counters are the workhorse of safeguards today. With the advent of mixed oxide fuels, the ability to measure plutonium in a direct measurement, rather than inferred from Cm/Pu ratios will be important. LLNL has developed a segmented liquid scintillator multiplicity counter with nanosecond timing, which has 10^5 discrimination of neutrons and gamma-rays above 500 keV. This detector is modular and scalable in size depending on overall efficiency requirements. It is based on NE-213 xylene-based liquid scintillator modules, now available as BC-501A from Bicron and EJ-301 from Eljen. Future detectors will be made from the EJ-309 to eliminate the flammability hazard of EJ-301. A photo of a 4” x 3” EJ-301 xylene liquid scintillator detector and PMT housing is shown in **Error! Reference source not found.**1. This type of liquid scintillator can also be used for neutron spectroscopy by unfolding the energy spectrum using a set of response functions. Historically liquid scintillator detectors have had relatively poor gamma ray – neutron discrimination from analogue pulse-shape discrimination (PSD). With fast digital electronics, far superior neutron – gamma ray identification and much lower energy thresholds yield much better overall neutron efficiency and identification than is possible with analogue PSD's. The fast digital electronic DAQ system can handle list mode data readout ~10KHz/Ch. (see Figure 2.)

Figure 1 Photograph of a 4" diameter x 3" Eljen EJ-301 xylene based liquid scintillator detector with integration PMT.

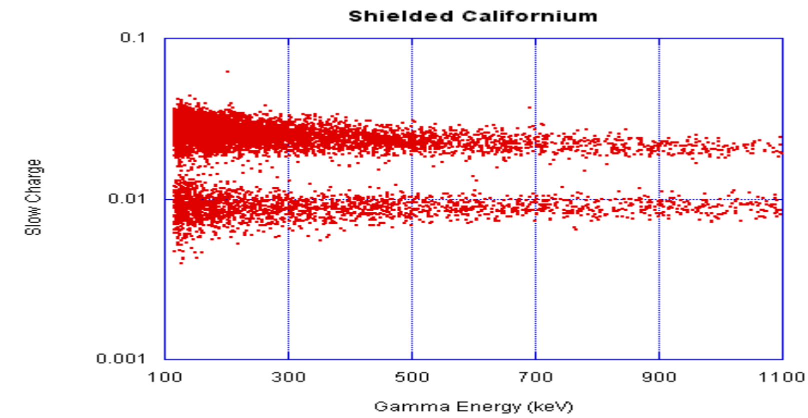


Figure 2 Separation between neutrons and gamma-rays in the DAQ system.

Used passively, this fast neutron counter can detect isolated fission chain bursts in the presence of high backgrounds. High background may occur in storage areas, or when the signal is dominated by (alpha,n) or additional spontaneous fission sources. With ^{3}He detectors, neutrons created in isolated bursts are spread in time by neutron diffusion times required to thermalize the neutrons. Consequently neutrons from different spontaneous fission events overlap in time, making it difficult to separate the contributions of multiple kinds of neutron sources. This is especially true when the different sources are qualitatively statistically similar, as they are for different spontaneous fission sources. With this liquid scintillator detector array we have recently demonstrated the isolation of individual fission chains in low-multiplying Pu. The liquid scintillator array can be useful in quantifying total fissile mass by isolating spontaneous fission bursts; statistically the different distributions of emitted neutrons (and gamma-rays) from Cm versus Pu spontaneous fission could be distinguished, and the absolute masses determined (from the rates of the separate kinds of spontaneous fission sources).

Used actively in conjunction with 60 keV neutron interrogation, the liquid scintillators are threshold neutron detectors, blind to the interrogating beam, but responding to the induced high-energy fission neutrons. The 60 keV beam energy selectively fissions ^{235}U and ^{239}Pu and not ^{238}U or ^{232}Th. With fast timing to isolate induced fission events, this technology could be useful for high count rate applications.

<u>ANALYSIS METHODS.</u> We have developed at LLNL a complete analytic point model theory of neutron counting distributions and time interval distributions. The foundation of the theory is a complete analytic solution[1] of the Bohnel[2] fission chain model. The fission chain theory completes the Dierckx and Hage[3] and Hage and Cifarelli[4] theory of random time gate and triggered time gate counting distributions. We have also developed a theory of time interval distributions, including skipped intervals[5]. Using this theory we perform a multi-parameter maximum-likelihood analysis to determine the parameters values that characterize the distributions[6,7]. The parameters are: the system multiplication, a diffusion time constant (determined from the time-gate dependent Feynman variance to mean – *see Figure 3*), detection efficiency, spontaneous and (alpha, n) source strengths, external direct shine Poisson-distributed background count rate (that can also induce fission, as in active interrogation with an (alpha, n) source). A provisional model of cosmic ray background is also included. The theory has also been extended to include the neutron capture gamma-ray signal.

A random source used in active interrogation of HEU, and that dominates the count rate due to direct shine on the ^{3}He counters, is robustly separated in this analysis. This is because the Poisson contribution is qualitatively different from the fission signal, and its contribution is characterized by only a single parameter, the external count rate. This separation of sources capability can possibly be extended to the problem of separating the Pu contribution from a dominant Cm source, especially if this analysis is applied to the fast timing signal.

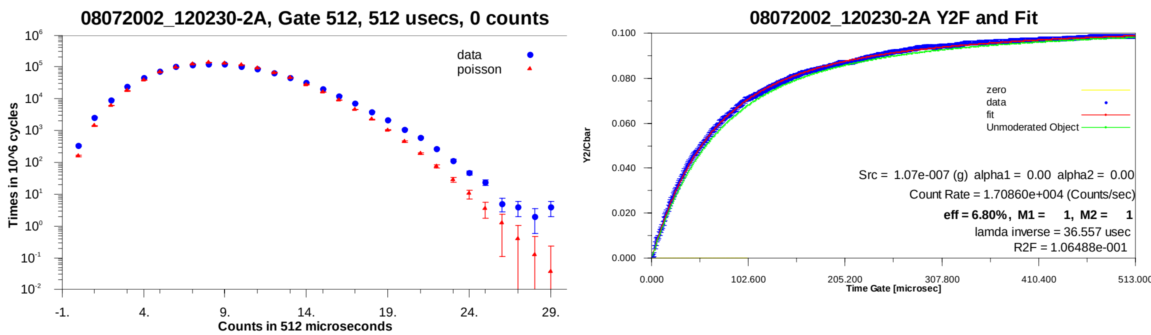


Figure 3 252Cf spontaneous fission count distribution (blue) compared to a Poisson distribution of the same count rate (red) for a 512 sec time gate. The right panel shows the time dependent Feynman variance to mean. The last point is determined by the increased variance of the count distribution in the left panel.

The liquid scintillator detectors also bring energy discrimination of the neutron flux which will inherently contain some information of the source (spontaneous fission vs. alpha induced neutrons) but also leads to our third area of investigation which is low energy neutron interrogation of a sample. SNM like ^{235}U or ^{239}Pu compared to Curium isotopes has a very different and specific ratio of spontaneous neutron emission versus thermal neutron induced fission neutrons. By actively interrogating the material with a low dose of thermal or thermalized 60 keV neutrons, one can well differentiate if the spontaneous neutron emission comes from curium or plutonium, for example. For an identical neutron emission rate, plutonium will react strongly to an external low energy neutron flux by emitting MeV neutrons, but curium does not.

SUPERCONDUCTING HIGH RESOLUTION GAMMA SPECTROMETER

Gamma spectrometry is widely used to determine the isotopic analysis of radioactive materials in Safeguards. Cryogenic gamma-ray spectrometers operating at temperatures of T~ 0.1K offer an order of magnitude improvement in energy resolution over conventional high purity germanium detectors.[8] They typically consist of an absorber attached to a sensitive thermometer, both weakly thermally linked to a cold bath. We are developing gamma-ray detectors based on bulk superconducting Sn absorbers coupled to Mo/Cu superconducting-to-normal transition edge sensors (TESs)[9,10,11]

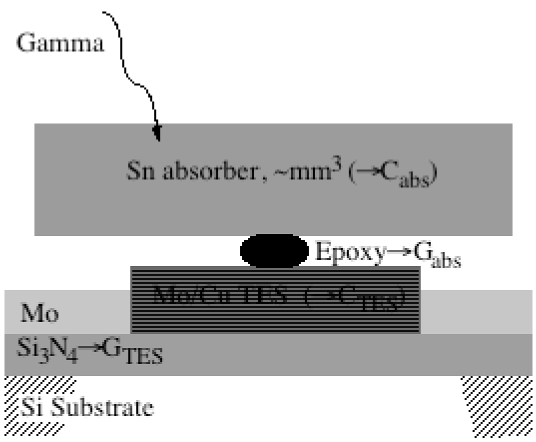
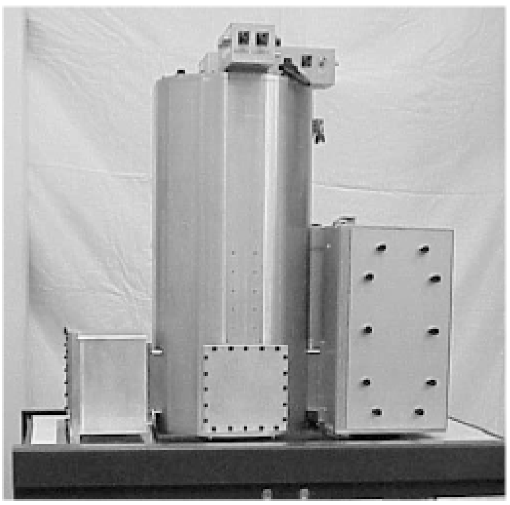


Figure 4 Schematic of superconducting transition edge sensor (not to scale)-

We are also developing refrigeration and readout technology for user-friendly detector operation at ~0.1 K. These detectors have achieved an energy resolution between ~50 and 90 eV FWHM for energies below 100 keV and are ideally suited for precise nondestructive analysis of nuclear samples. The spectrum of a low-enriched uranium sample (Figure 5) illustrates the advantage superconducting detectors offer for the analysis of nuclear samples. The 90 keV region of the spectrum is of interest for the analysis of uranium-containing samples, as it can be used for precise measurements of enrichment based on emission lines with very similar energies. (see Figure 2).



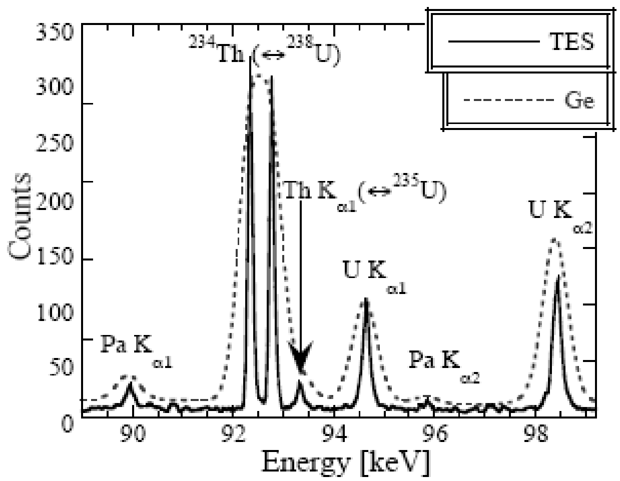


Figure 5 Cryogenic spectrometer with detector cold finger (left). Gamma spectrum of a low-enriched uranium sample in the 90 keV region used for precision measurements of uranium enrichment. A spectrum of the same sample from a planar Ge detector is included for comparison.

This increase in energy resolution in the low-energy region can be very helpful for safeguards measurements where enhanced precision is needed. This is not only the case for measuring uranium enrichment at 92 keV, but also, for example, in precision measurements of plutonium isotopes at 100 keV. The method will help to detect weak lines above background and thus measuring minor isotopes such as ^{242}Pu directly, rather than inferring them through model-based correlations with other isotopes.[12] While mass spectrometry ultimately provides higher sensitivity, the automation and ease of operation makes gamma spectroscopy the preferred tool for initial inspection of a large number of samples.

SOLID STATE NEUTRON DETECTORS

LLNL is currently developing configurable, real-time, low-power, compact, solid state neutron detection system for special nuclear materials neutron signature detection with 10% thermal neutron detection efficiency. This neutron detection system can be used for long term monitoring of nuclear material in storage, or it can be an added feature in a tag or seal. These neutron detection systems can be also used for detecting or monitoring neutron emissions in areas (e.g., underwater, in pipes, etc.) where there are complex scattering environments, no available power, and little or no human access. There are two configurations in the neutron detector, namely ^{10}B for thermal neutrons and $^{6,7}LiF$ for higher energy neutrons.

LLNL originally developed novel inexpensive neutron detectors for the long-term monitoring of neutron activity from contraband nuclear material in areas that are inaccessible, such as a cargo-ship container during transport[13] Our design uses 5mm-diameter, self-biased ^{10}B-coated neutron detectors (tiny neutron tags) that have 2% efficiency for thermal neutron detection. These detectors have been demonstrated in the laboratory that they can detect neutrons emitted from plutonium in a complex scattering environment in a reasonable amount of time. These coin-sized detectors (when mass-produced) could be ~$20 a piece. For Safeguards, a simple detector that can be placed on pipes and casks to determine bulk measurement of neutrons (or lack thereof) for tracking processes and ensuring there is no diversion.

We have developed a prototype of a tiny tag with 15mW electronics (*See Figure 4*). Time information of each event is stored for further data analysis. These semi-conductor detectors are better than currently available ^{6}Li doped scintillation fibers and photomultiplier based scintillation detectors. They are also immune to gamma-rays and stable in comparison to ^{3}He tubes.

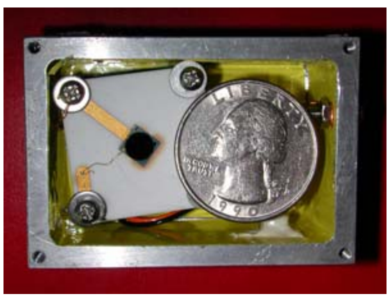


Figure 6. Solid state based neutron detection system with 15mW electronics.

More recently, we are developing next generation solid-state neutron detectors with thermal neutron detection efficiency beyond 25%. These high aspect ratio pillars[14] will be filled with CVD ^{10}B, $^{6,7}LiF$ for both thermal and low energy neutron detection. (Figure 5)

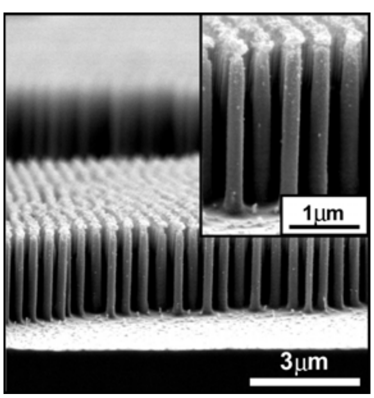


Figure 7. High aspect ratio silicon pillar structure developed at LLNL. When filled with 10B and 6,7LiF, the neutron detection efficiency for this system can be as high as 50%.

These inexpensive tiny tags can be used for ubiquitous neutron monitoring in the GNEP processes as "smart tags" for tracking material flow. In a centrifuge facility, they may be used as neutron imagers within the centrifuge hall to monitor for possible diversions (Neutron imagers have been suggested by Mark Pickrell.)[15] The information can be propagated with wireless options.

SPECTROSCOPIC 3D GAMMA-RAY IMAGER

LLNL is currently developing a new imaging technique able to provide 3D images of gamma-ray sources. Unlike standard 3D tomographic methods, this stand-off imaging technique does not require the field of view to be bounded within a predefined physical space.

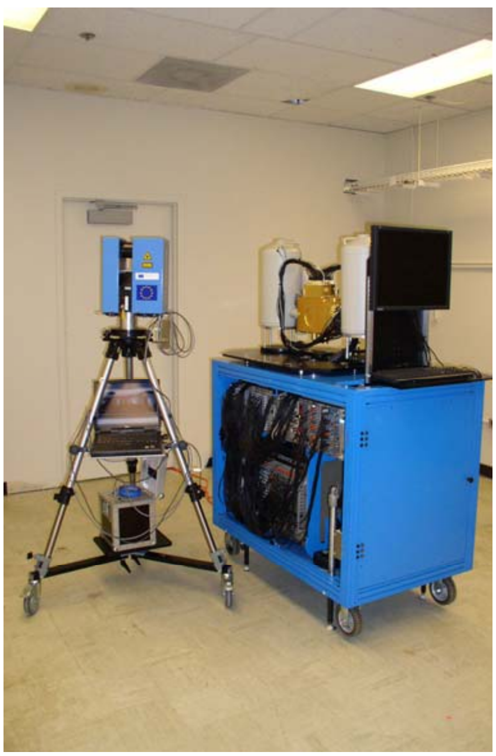


Figure 8 A 3D Design Information Verification (DIV) laser range scanner (left) and a Compact Compton Imager (CCI) gamma-ray camera on a mobile cart (right). This work is a collaboration with ORNL and Euratom.

In the present implementation, the gamma-ray imaging system is based on planar Si(Li) and HPGe double sided segmented detectors, which are used in a Compton camera configuration. This imaging system follows up on a previous imager made of a single Ge detector[16]. With this imaging system, unlike other gamma-ray imagers, the directionality of the photons is not obtained by using collimators, but by measuring the scattering angle and scattering direction of the incident photon in a Compton interaction inside a position sensitive detector. An added benefit of using these detectors is their outstanding spectroscopic capabilities (2keV at 662keV), which allows the system to create independent 3D images for each radioisotope of interest[17].

A lidar system is used in conjunction with the gamma-ray imaging system to confine the

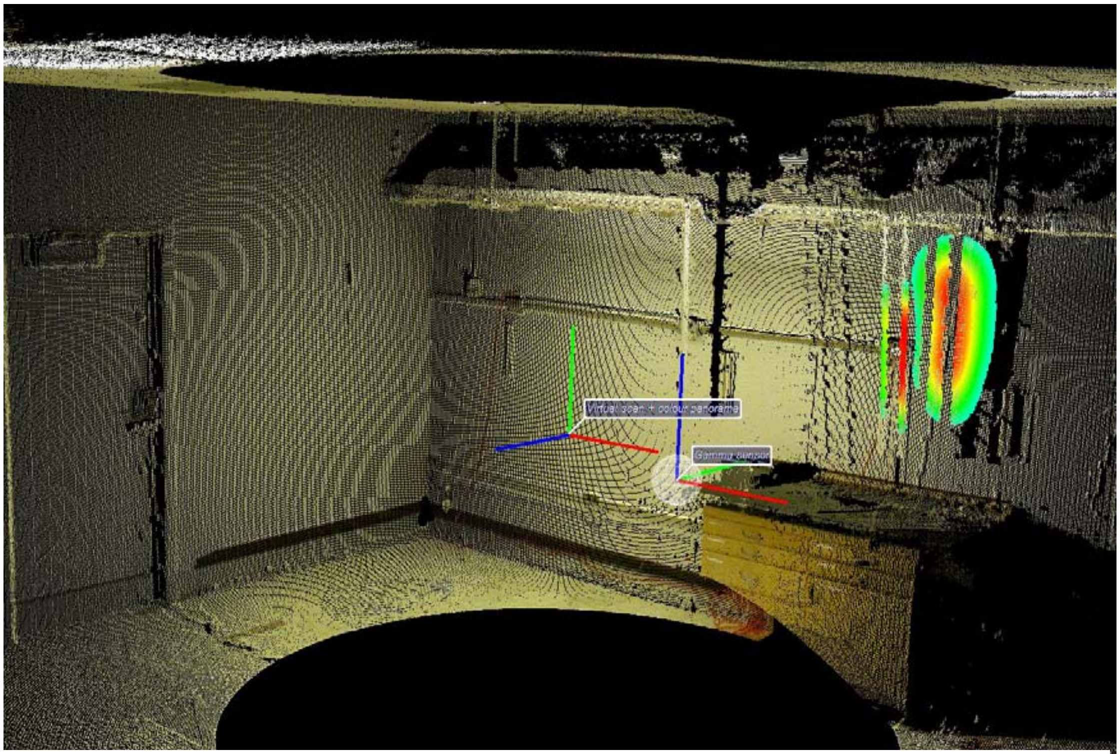


Figure 9 Snapshot of the 3D integrated model of the room. Backprojected hot spots of radioactivity are represented as surface plots superposed onto the virtual scan. The position of the gamma-ray imager is represented by the inner coordinate system on the right side.

gamma-ray image space to the interior of physical objects situated within the vicinity of the gamma-ray imager. This approach results in superior image contrast and much reduced image reconstruction processing times. Figure 10 shows an example of such a 3D gamma-ray image of a ^{152}Eu source. In Figure 9, a snapshot of the virtual model of the laboratory room as obtained from the lidar scans is shown along with a backprojected gamma-ray distribution as obtained from a single gamma-ray projection. The whole 3D gamma-ray image showed in figure 10 required gamma-ray projections taken from 3 different positions.

The new imaging technique can potentially increase the sensitivity and automation level for inspection, search, monitoring and diagnostics of special nuclear materials in reprocessing facilities and glove boxes.

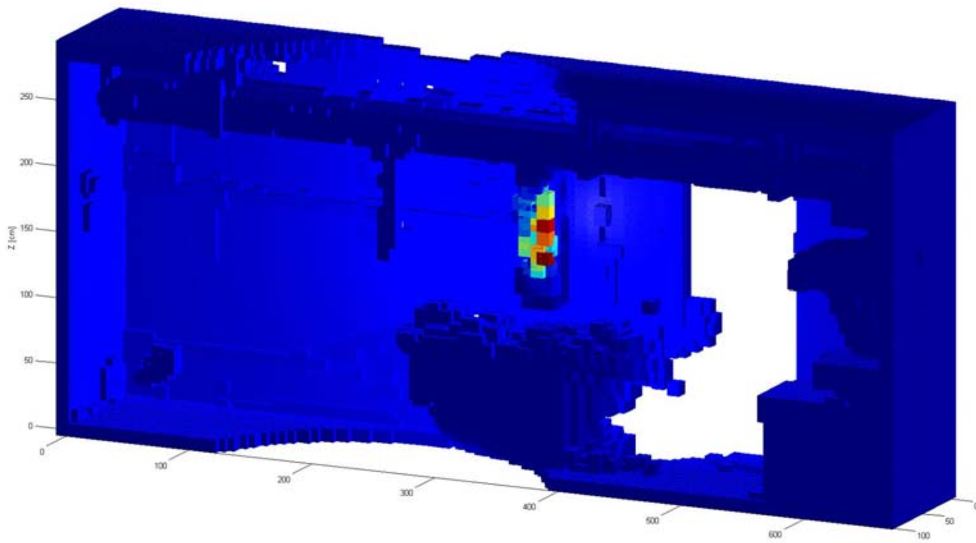


Figure 10: Example of a 3D gamma-ray image (a linear 1m long Eu-152 gamma-ray source was used. The voxalized image space was determined by the lidar scans.

SUMMARY

We have discussed two technologies developed at LLNL for Homeland Security applications. We have given some reasons why these techniques can be brought to bear on the Safeguards problem.

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