Radioactive Threat Vision via Quantitative Gamma-ray Imaging

Ethan Hull, Matthew Kiser

PHDS Corporation, 3011 Amherst Road, Knoxville, TN 37921, USA

Historically, radioactive material detection and localization have been accomplished through the semi-random movement of a detector in the presumed vicinity of the radioactive materials. This non-ideal search technique is subject to user perception errors, detector saturation and the radiation-dose and criticality hazards associated with close-proximity intervention. New compact high-resolution gamma-ray imagers provide the sensitivity and specificity to comprehensively assess the complete radiological situation from safe standoff distances *before* near-field intrusion. These portable high-resolution gamma-ray imaging detectors offer CBRNE teams an intuitive visual map of the radioactive material locations, identities and quantities. Both real-time on-screen information and reachback data transmission allow the team to determine the most effective threat mitigation strategy while minimizing risk to personnel. The semiconductor crystal, detector, electronic, software and imaging science are now at a manufacturing level to avail these benefits to the CBRNE responder in a portable footprint. In addition, the science and manufacture of these imagers has now been complemented by real-life measurements by CBRNE teams against relevant scenarios. Live CBRNE exercises have revealed multiple-source configurations, distributed-source shapes and quantitative information that could not have been discovered without the nuclear-visualization provided by gamma-ray imaging.

Introduction

Imaging is possibly the most useful source of information for numerous security applications. Much of the electromagnetic spectrum is well utilized by infrared and visible imaging to determine the presence, location and shapes of objects of interest. However, gamma-ray imaging is *not* conventionally used in the assay of radioactive threats because the technology has not been generally available. Usually the search for nuclear materials includes team members moving about with radiation detectors while watching for changes in gamma-ray counting rates to determine the most likely location(s) of the radioactive materials. Generally other visible observations must be relied upon to narrow the search. Fortunately, a new generation of gamma-ray detection *and imaging* technology now allows responders and interdiction teams to “see” the distributions of radioactive materials from a distance before approaching the region of interest (Kiser, et al. 2014, Burks and Dreyer 2014). As illustrated in Figure 1, the presence, locations and quantities of security-critical and nuisance radioactive materials can be quickly and comprehensively understood from a standoff distance by a single wide-field imaging measurement.

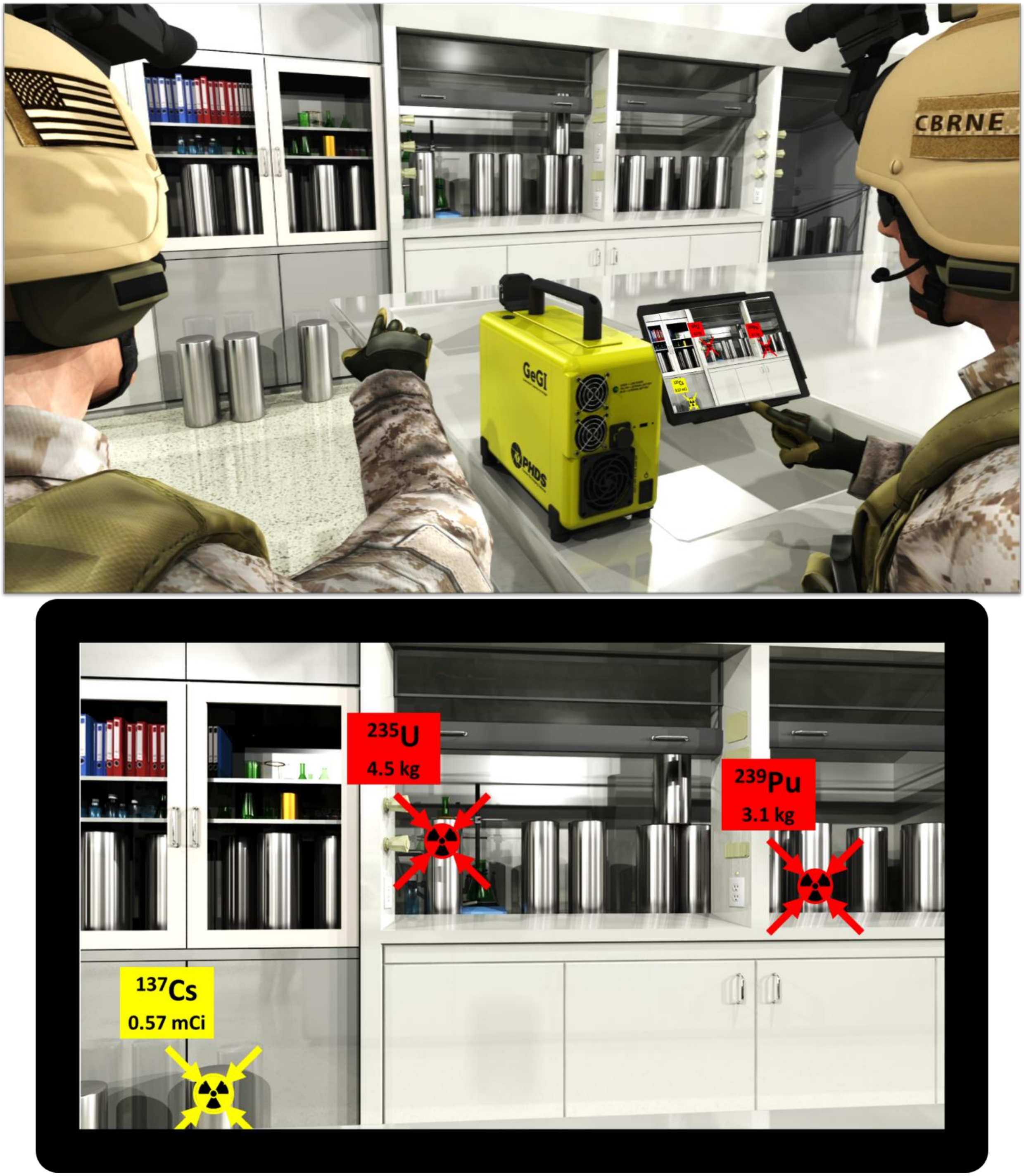

Figure 1. CBRNE team members analyze the radiological situation upon entering a nuclear weapons production site. The visual nuclear threat information from the imaging detector shows the identities, locations and quantities of the radiological materials on an intuitive visual display.

Germanium gamma-ray detectors (HPGe) are the largest-volume single-crystal semiconductor gamma-ray detectors available. The unparalleled energy resolution quality places germanium detectors in a class of their own with regard to radioisotope identification through gamma-ray detection and spectroscopy. Although germanium detectors have long served as the best quality gamma-ray detectors, the required cryogenic detector cooling (~77 K) and low-noise electronic instrumentation have historically limited the availability and utility of germanium detectors. This has been particularly true for innovative gamma-ray *imaging* germanium detectors, previously existing at only the laboratory-demonstration level. An example of a nuclear physics laboratory imaging detector is shown on the left side of Figure 2. The laboratory system relies on a large liquid-nitrogen dewar for cryogenic cooling, with long signal cables between the preamplifiers and the processing electronics. Considering the separate laptop serving as the data-acquisition device, the entire system weighs ~150 lbs., making it perfect for a laboratory but not useful for field work. On the right side of Figure 2, the 28 lb. yellow Germanium Gamma-Ray Imager (GeGI) has a far smaller footprint with the same active detector size (sensitivity) and performance as the laboratory system. An internal mechanical cooler provides cryogenic detector operation with no need for liquid nitrogen. All electronics, cabling and control systems are fully integrated and interlocked. A single click on the GeGI tablet collects data while algorithms alert the user to the presence, identity and location of radioisotopes on an intuitive visual threat assessment display. The combination of new large-diameter germanium-crystal growth techniques, pixelated detector fabrication, novel mechanical cooling solutions, compact electronics, and gamma-ray imaging advances allow the power of a nuclear-physics imaging laboratory to be carried in one hand.

Figure 2. On the left a laboratory imaging detector developed for nuclear-physics research relies on liquid nitrogen for cryogenic operation (~150 lbs.). On the right, the 28 lb. GeGI has a detector of the same size with all electronics and a Stirling cycle mechanical cooler contained inside the yellow box.

Germanium Semiconductor Manufacturing and Imaging Physics

The germanium semiconductor manufacturing required to fabricate an imaging germanium detector system resembles a distant cousin of the far more prevalent silicon semiconductor manufacturing for electronic components. The manufacturing process is summarized in Figure 3. Germanium is refined to the “high purity” level through successive melting and solidification in the zone-refinement process (Haller 1976). Fortuitously, the relevant electrically-active impurities in germanium segregate into the solid (k>1) or liquid (k<1) phases sufficiently to become highly concentrated into either the head or tail of a zone refined germanium ingot (~10 kg), leaving the middle region free of impurities. By the end of the zone refinement process, there is a sizeable region of high-purity germanium ($|N_a – N_d| \sim 10^{10}$ /cm^3) in the middle section of the polycrystalline zone-refined bar that can be used for high-purity crystal growth. A mass of high-purity germanium is loaded into a large-diameter Czochralski puller to be grown into a single crystal (Hansen and Haller 1983, Hall 1984). The entire mass (~10 kg) is fully melted before a small seed crystal is lowered onto the center of the germanium pool. The melt temperature is adjusted to the germanium melting point (937°C) and the molten germanium begins to freeze onto the seed crystal as it is drawn upward by the pull-shaft mechanism while rotating. The entire pool of molten germanium is grown into a solid single crystal and then allowed to cool.

The cooled crystal is sliced into wafers that are analyzed and then processed into imaging-capable planar germanium detectors. Using a non-standard segmented surface fabrication technique, the relatively large and thick wafers are processed into two-sided segmented imaging-capable detectors (Luke 1992, Hull and Pehl 2005). After fabrication, the detector is a diode that can be fully depleted of free charge carriers by a bias voltage (~1000 V) when cooled to ~80 K. In the depleted condition, there is a substantial electric field (~1000 V/cm) throughout the volume of the detector with little or no current flow. The only free (mobile) charge carriers in the depleted volume of the detector are created by ionizing radiation in the detector. Because germanium has excellent charge-carrier mobility and very little charge trapping, the charge carriers are collected efficiently on the segmented contacts to produce a highly accurate and uniform energy response. Germanium is the only physical system that truly functions in such a manner, and this crystal-detector property is the reason for the continued predominance of HPGe as the best gamma-ray spectrometer material. The finished detector is mounted in a mechanically-cooled vacuum cryostat incorporating interconnects and electronics to read out gamma-ray signals from the detector (Goulding 1972, Radeka 1968). This system also contains control electronics to monitor and interlock the detector temperature, cooler power, bias voltage and other critical system power supplies.

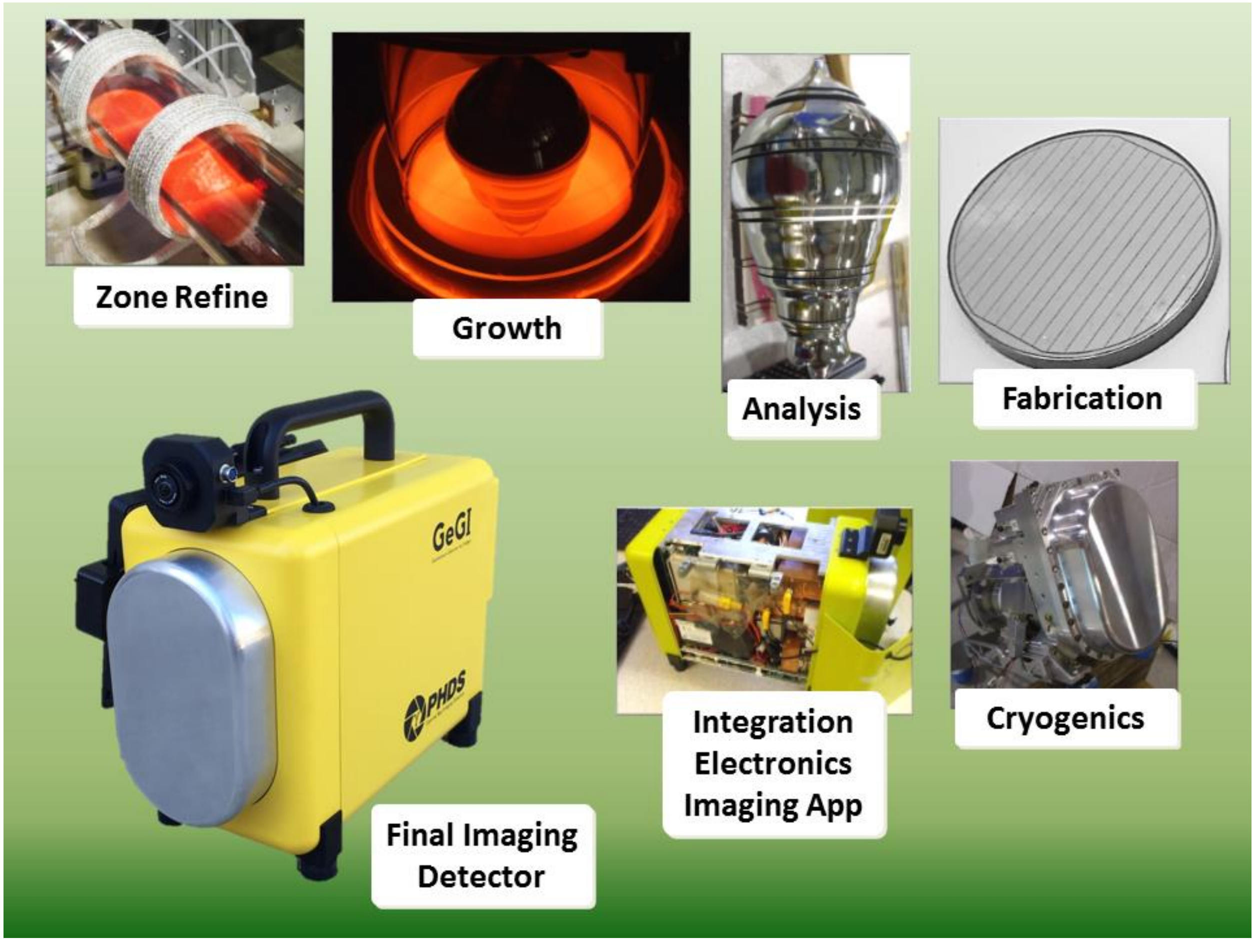

Figure 3. From raw materials through radiological answer, the manufacturing process starts with germanium material refinement, continuing with crystal growth, detector fabrication and system integration to complete the Germanium Gamma-ray Imaging (GeGI) detector system.

The fully assembled and instrumented GeGI detector system now provides a 90-mm diameter 10-mm thick HPGe detector with 3-dimensional spatial resolution of FWHM ~1.5 mm for individual gamma-ray interactions throughout the volume of the detector. The gamma rays incident upon the detector initially interact via photoelectric, Compton scattering or $e^{-}e^{+}$ pair production with varying probabilities depending on the energy of the incident gamma ray. For the purpose of this discussion, only photoelectric and Compton scattering are treated. At low energies, photoelectric interactions dominate, becoming equal to Compton scattering at ~140 keV. Because the GeGI detector can locate individual gamma-ray interactions, it can “image” and “locate” distributions of radioactive materials by analyzing the gamma rays from those materials in two different ways: pinhole imaging and Compton imaging.

Pinhole-imaging necessarily involves the placement of a high-Z (e.g., Pb, W, Ta) pinhole aperture in front of the detector. As shown on the right side of Figure 4, pinhole imaging is simply the reconstruction of the upside-down and backward projection of the radioactive-source configurations on the plane of the detector. Gamma-rays imaged and counted must come through the small pinhole aperture, so the efficiency of the pinhole-imaging aperture is relatively low. However, pinhole imaging produces an exact back projection of the radioactive-source configuration making it extremely useful for complex-source geometry measurement as will be shown later. The second gamma-ray imaging modality readily available in the GeGI detector is Compton imaging. Compton imaging necessarily relies on Compton scattering of the incident photon. The photon scatters through an angle θ that can be calculated using the energy of the incident photon (E_1) and the scattered photon (E_2) according to the Compton-scattering equation shown on the left side of Figure 4. GeGI measures these two energies, locates the two interaction sites and then calculates a narrow angular cone of acceptance that includes the direction of the initial incident photon. After a number (~20) Compton interactions in the detector, these cones begin to overlap in a common ray indicating the direction of the actual source of gamma rays. Compton imaging does not require a Pb aperture (pinhole) to function, so the entire volume of the detector is wide open and images all of 4π. Compton imaging is a very good way to initially assay the radiological situation – finding the highest intensity barrels or containers very quickly. Often a Compton-image assay is performed upon initial entry into an unknown environment as illustrated in Figure 1. The Compton image quickly reveals the locations, identities and quantities of the radiological materials. The Compton image is commonly followed by pinhole-aperture imaging at closer proximity to better characterize the shapes and substructures of the gamma-ray emitting object(s). In both imaging modalities, a comprehensive radiological threat assessment can be obtained before committing to a particular physical intervention strategy. All of this information appears in real-time on the screen of the GeGI tablet. The data is conveniently relayed to intelligence assets for further detailed analysis by pressing a single reachback button. There is no better tool for situational awareness of the radiological environment.

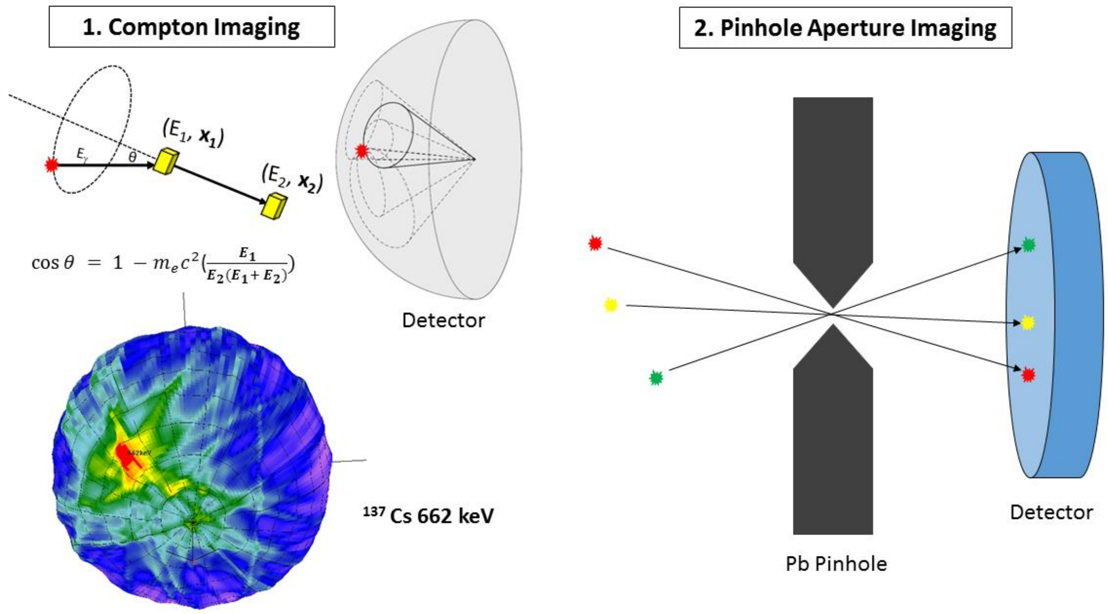

Figure 4. Compton Imaging and Pinhole Aperture Imaging are both simultaneously accommodated by the Germanium Gamma-ray Imaging Detector. Compton Imaging is well suited to high sensitivity search while pinhole imaging makes highly detailed object images including shape and substructure information.

CBRNE and International Safeguards Imaging of Special Nuclear Materials

The following examples are all real measurements made using a standard COTS GeGI gamma-ray imaging detector system in radiological situations that were *not* known to the operator. In this CBRNE team-training demonstration, GeGI was used in Compton imaging mode to initially locate and identify sources at a distance. The barrel shown in Figure 5 was immediately identified as containing a source of ^{137}Cs. This barrel contained a relatively intense (500 µCi) ^{137}Cs nuisance source shielded in a 1.5-inch thick Pb pig. Such a source configuration produces excessive Compton down-scatter that purposely confounds the measurement of Special Nuclear Materials (SNM) because all of the important SNM gamma rays happen to fall in the Compton continuum of ^{137}Cs. Even though a substantial ^{137}Cs Compton continuum exists, the excellent gamma-ray energy resolution and sensitivity of the GeGI detector unambiguously identified a very small ^{239}Pu source using the 375 keV, 414 keV and 129 keV gamma ray peaks from a distance of ~1 meter. Compton images of the 375 keV and 414 keV peaks revealed the location of the ^{239}Pu object in the barrel to the right of the ^{137}Cs source. The quantitative Compton-imaging feature of the GeGI determined a mass of 1.4 g of ^{239}Pu. Other detectors present at the scene were unable to identify the presence of ^{239}Pu in this situation, much less locate it.

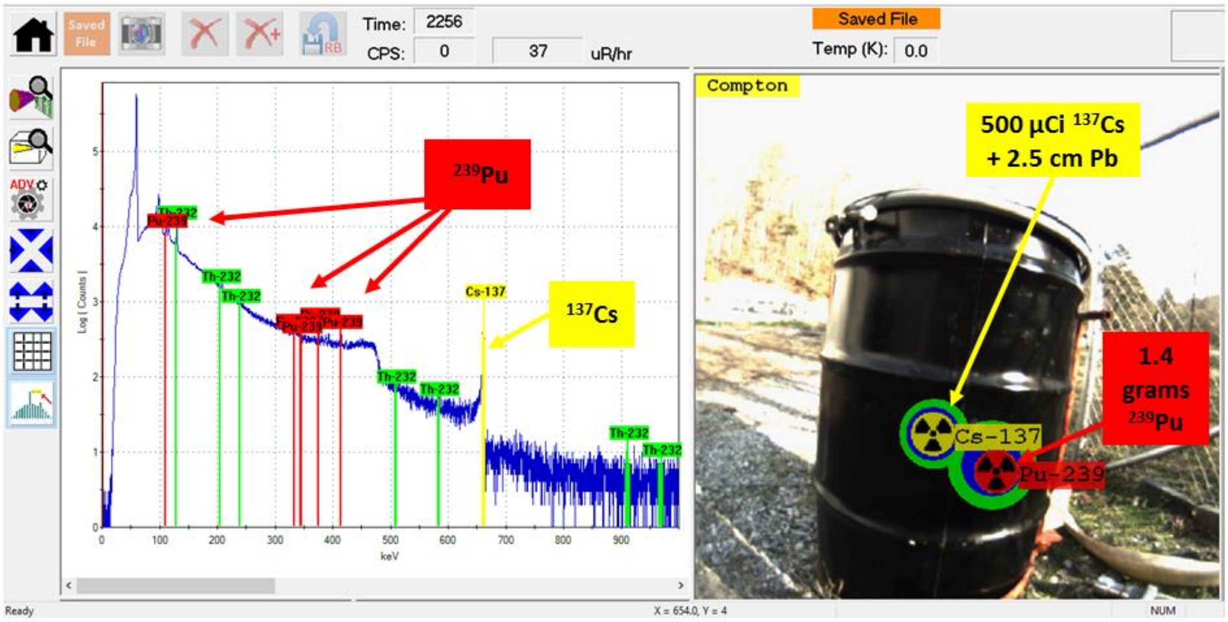

Figure 5. A CBRNE training exercise demonstrates the capability to detect, locate and quantify a very small ^{239}Pu source in the presence of a much larger ^{137}Cs source. The excellent energy resolution from the germanium detector detects the relatively weak ^{239}Pu gamma-ray lines in the energy spectrum while the Compton imaging locates the object and quantifies it at as having a mass of 1.4 g.

After identifying, locating and quantifying the ^{239}Pu object inside the barrel, the object was removed from the barrel for further characterization. The GeGI was moved in to very close proximity (~10 cm) to the object and a detailed quantitative pinhole image was acquired. The outer case of the ^{239}Pu object was a 5-cm equilateral triangular shape. No photograph of the object was available. However, the near-field quantitative pinhole-aperture image of the object showed that the larger triangular shape was actually made up of four smaller triangular sections or chambers, as shown in Figure 6. Each of the smaller chambers included a relatively uniform planar distribution of ^{239}Pu amounting to ~0.3-0.5 grams in each chamber. The gamma-ray energy spectrum also contains ^{241}Am and ^{237}Np gamma rays. The combination of GeGI imaging and spectroscopy clearly separated the ^{239}Pu in this measurement. This first-ever measurement of the triangular (and sub-triangular) special-forms object represents a clear example of the power of isotope-specific gamma-ray imaging to characterize shapes of special nuclear materials.

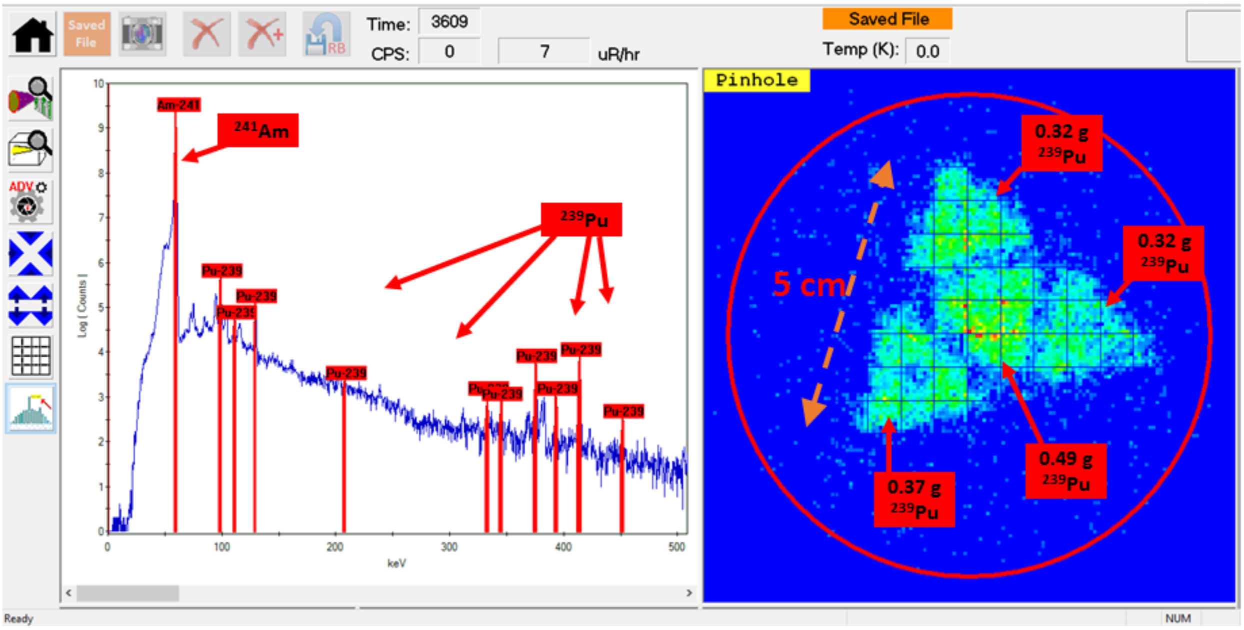

Figure 6. The energy spectrum and pinhole-aperture image from the triangular ^{239}Pu object. The gamma-ray image reveals the object to be made up of four smaller triangular chambers each containing a relatively uniform planar distribution of ^{239}Pu quantified as shown.

Having described separate examples of Compton and pinhole gamma-ray imaging of the ^{239}Pu object, it is informative to note that *both* imaging modalities can be used at the same time with the pinhole aperture in place. On more than one occasion, identification, location and quantification of radioisotope distributions have been demonstrated with *both* Compton and pinhole imaging modalities *at the same time*. The measurement shown in Figure 7 highlights a result from an IAEA nuclear-safeguards workshop held in October 2015 in Seibersdorf, Austria. During the workshop, different radioactive material configurations were hidden behind a screen. Data were then collected and analyzed by GeGI. The particular measurement in Figure 7 is an example of a simultaneous Compton/Pinhole image. The Compton imaging clearly resolved and quantified ^{137}Cs and ^{60}Co sources while the Pinhole image measured three distinct distributions of enriched ^{235}U. Analysis of the Compton image (accounting for the 2.54 cm thick pinhole shield) indicates 65 µCi of ^{137}Cs and two sources of ^{60}Co of about 10 µCi each. Analysis of the selected regions of interest in the pinhole image indicates 116 grams of ^{235}U contained in three distinct regions of 64, 32, and 20 grams. The quantitative image processing software allowed all of this information to be obtained with a single measurement from a single standoff position 2.5 meters from the target. PHDS personnel operating the GeGI had no *a priori* knowledge of the source configuration during these measurements (IAEA 2015).

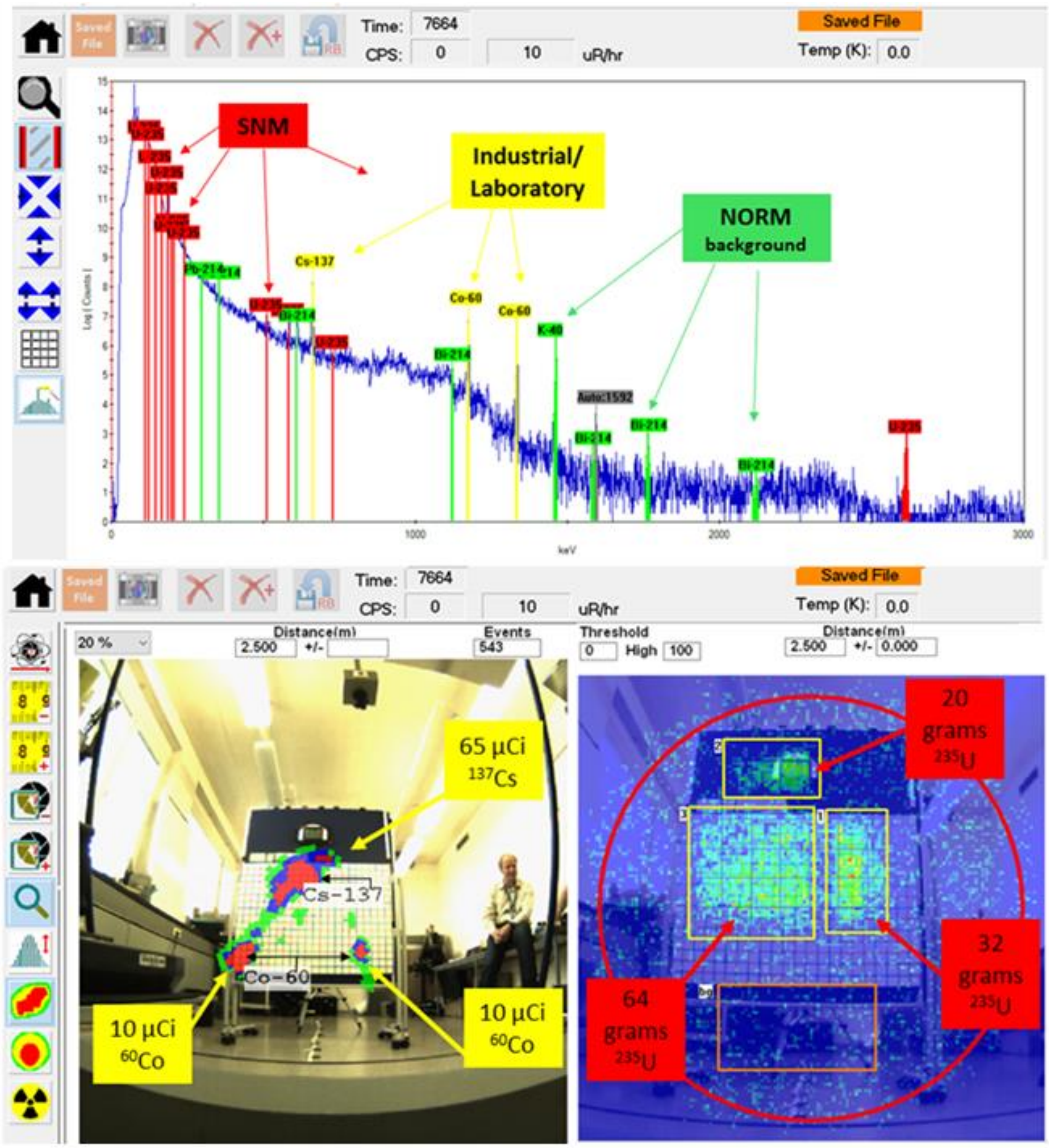

Figure 7. A simultaneous Compton and Pinhole Image. The Compton imaging locates, identifies and quantifies ^{60}Co and ^{137}Cs sources while pinhole imaging showed detailed regions (and missing regions!) of an enriched ^{235}U fuel assembly.

Conclusion

Thanks to improvements in germanium semiconductor detector technology, detection systems are now available that can also provide gamma-ray imaging to lend vision to nuclear threat detection. Although the examples presented here focus specifically on Special Nuclear Materials in CBRNE and Safeguards applications, imaging detector systems like GeGI are now finding application in numerous other fields including nuclear materials management, decontamination & decommissioning, health physics, nuclear medicine and nuclear nonproliferation. The notion of an investigator walking around watching the count rate on an ordinary detector will be increasingly replaced by the use of versatile imaging detectors like GeGI. Imaging detectors provide nuclear-threat vision information on the screen of a phone or tablet. Rather than having to collect multiple measurements to infer the radiological source characteristics, the CBRNE team will have a comprehensive assay of the exact nuclear-material assembly in question. The single-button reachback capacity and real time displayed information result in improved situational awareness and, of course, a safer intervention.

References

Burks, M., and J. Dreyer. 2014. *GeGI: Chicago field test report (trip #2).* Technical Report, LLNL-TR-656851 https://e-reports-ext.llnl.gov/pdf/777627.pdf.

Goulding, Fred S. 1972. "Pulse-shaping in Low-noise Nuclear Amplifiers: A Physical Approach to Noise Analysis." *Nuclear Instruments and Methods* 100: 493.

Hall, R.N. 1984. "HPGe: Purification, Crystal Growth, and Annealing Properties." *IEEE Transactions on Nuclear Science* 31 (1): 320.

Haller, E.E., Hansen W.L., Hubbard, G.S., Goulding, F.S. 1976. "Origin and Control of the dominant Impurities in High-Purity Germanium." *IEEE Transactions on Nuclear Science, NS-23, No. 1* 81.

Hansen, W.L., and E.E. Haller. 1983. "High-Purity Germanium Crystal Growing." *Materials Research Society Symposium Proceedings* 16: 1.

Hull, E.L., and R.H. Pehl. 2005. "Amorphous germanium contacts on germanium detectors." *Nucl. Inst. and Meth. A538 No. 1-3* 651.

IAEA. 2015. *Technology Demonstration Workshop on Gamma Imaging: Preliminary External Technical Report.* IAEA Headquarters, Vienna; IAEA Laboratories, Seibersdorf: International Atomic Energy Agency.

Kiser, Matthew, Seth Henshaw, Nathan Hoteling, John Goldsmith, Mark Gerling, Morgan Burks, Jonathan Dreyer, et al. 2014. "Evaluation of Gamma and Neutron Imagers for Radiological/Nuclear Search." OUO Technical Report, DNN R&D.

Luke, P.N., C.P. Cork, N.W. Madded, C.S. Rossington, M.F. Wesela. 1992. "Amorphous Ge Bipolar Blocking Contacts on Ge Detectors." *IEEE Trans. Nucl. Sci. Vol. 39, No. 4* 590.

Radeka, V. 1968. "State of the art of low-noise amplifiers for semiconductor radiation detectors." *International Symposium on Nuclear Electronics.*