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Review of Nondestructive Assay Methods for Safeguards Monitoring and Verification
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Sophie Weidenbenner[1], Camille Palmer[1], Mark Croce[2]
1) Oregon State University, Corvallis, OR, USA
2) Los Alamos National Laboratory, Los Alamos, NM, USA

Efficient and accurate verification techniques are necessary for a functional nonproliferation framework. Passive, nondestructive assay (NDA) methods are often more convenient but are limited to the inherent physical characteristics of the material, such as emitted radiation, to confirm the composition and amount of fissionable material present. Existing NDA instrumentation and methodologies are summarized in the context of specific materials and radiological signatures of interest to the International Atomic Energy Agency's (IAEA's) safeguarding framework. Additionally, a discussion and analysis of these NDA capabilities will highlight an ultra-high-resolution gamma microcalorimeter spectrometer. This microcalorimeter is an advanced, up-and-coming NDA technology with the potential to significantly improve the precision, accuracy, and confidence in safeguards NDA measurements. Technical details of this instrument are provided, along with results showcasing the capabilities of gamma microcalorimetry for nuclear material measurements.

1 INTRODUCTION

There are many passive NDA instruments approved for use by the International Atomic Energy Agency (IAEA) [1], each with their individual strengths and limitations dependent upon the application. One notable limitation governing all passive NDA techniques is the precision and accuracy of the measurements. For high precision isotopic analysis, samples are collected and shipped to an offsite laboratory where they must undergo chemical separations to be measured by a destructive assay technique such as mass spectrometry. Destructive assays are relatively costly and time consuming for an agency with a very limited budget and time-sensitive detection goals. Future, advanced NDA instruments should aim to achieve the high precision of destructive assays to reduce the reliance on such techniques while also minimizing measurement times.

Existing NDA instrumentation and methodologies are summarized in the context of specific materials and radiological signatures of interest to the IAEA's safeguarding framework. Additionally, a discussion and analysis of these NDA capabilities will highlight an ultra- high-resolution gamma microcalorimeter spectrometer known as SOFIA (Spectrometer Optimized for Facility Integrated Applications). SOFIA is an advanced, up-and-coming NDA technology with the potential to significantly improve safeguards NDA capabilities. Technical details of this instrument are provided, along with results showcasing the capabilities of gamma microcalorimetry for safeguards measurements.

2 BACKGROUND

2.1 Safeguards Goals and Materials of Interest

The IAEA states the goal of safeguards is the "timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons or for purposes unknown, and deterrence of such diversion by the risk of early detection." This definintion provides the basis of the IAEA's safeguarding framework, which describes the specfic materials of interest along with the significant quantity and timeliness detection goals associated with material type.

The materials of most interest to safegaurds are materials containing fissile isotopes. Only fissile material is capable of sustaining a nuclear fission chain reaction sufficient for a nuclear explosive device. Fissile isotopes include U-235, U-233, and many Pu isotopes. Thorium, though not directly fissile, can be used as a source of U-233 via bombardment with neutrons and thus is also considered a material of interest to safegaurds. For each material, the IAEA has defined significant quanitities (SQs) such that one SQ of nuclear material is the minimum amount of material for which the possibilty to manufacture a nuclear explosive device cannot be excluded. Materials are split into two categories: direct use nuclear material and indirect use nuclear material. Direct use materials are those which can be directly used to create a nuclear explosive, whereas indirect use nuclear materials need to undergo enrichment or transmutation before becoming usable. Table 1 summarizes the materials of interest to safeguards and provides the SQ guidelines for each.

Table 1: Summary of safegaurds materials of interest and corresponding significant quanitites [2]

Material	SQ
Direct use nuclear material	
Pu[a]	8 kg Pu
^{233}U	8 kg ^{233}U
HEU (^{235}U ≥ 20%)	25 kg ^{235}U
Indirect use nuclear material	
U (^{235}U < 20%)[b]	75 kg ^{235}U (or 10 t natural U or 20 t depleted U)
Th	20 t Th

[a] For Pu containing less than 80% ^{238}Pu.
[b] Including low enriched, natural and depleted uranium.

2.2 Nuclear Material Accountancy

Nuclear material accountancy (NMA) aims to verify the presence or absence of special nuclear materials, as well as the identity and content of materials and any defects. The types of material defects can fall into three categories: gross, partial, or bias defects. Gross defects refer to entire item(s) missing, partial defects occur when smaller parts from a larger item are missing, and bias defects occur when small amounts of material are repeatedly diverted overtime. Verifying the identity of the material or object helps ensure operator declarations of item locations are accurate and no suspicious activity is occurring. Verifying the contents of the nuclear material requires knowledge of the isotopic information of the material to further ensure

operator declarations regarding item contents is accurate (e.g. enrichment levels in a UF_6 sample at an enrichment facility are within licensed limits). Other important aspects of NMA include the establishment of Material Balance Areas (MBAs) and the frequent measurement and calculation of Material Unaccounted For (MUF) as materials leave and enter these MBAs [4]. If no material has been diverted or added, MUF should be zero. Realistically, measurement systems have random and systematic error associated with their results, and this error must be propagated into the calculation of MUF expectancy. Because MUF is often non-zero, operators have a series of checks to determine whether they should sound the alarm. Radiological verification technologies are a vital aspect of NMA because they can detect and measure signatures of fissile material to determine isotopic composition and total material present. Gamma non-destructive assay (NDA) techniques are leveraged by inspectors to confirm the absences of gross and partial material defects with measurement accuracies on the order of a few percent. For bias defect detection, destructive analysis techniques with uncertainties less than one percent are typically required.

3 CURRENT GAMMA NDA INSTRUMENTS

The IAEA often prioritizes easily portable detection devices that allow for efficient inspections of entire facilities [1]. The most widely used gamma spectrometry detectors are NaI and high purity germanium (HPGe), which are scintillation and semiconductor type detectors, respectively.

The HM-5 field spectrometer, commercially available as the identiFINDER, is the bread and butter of small portable gamma detection instrumentation. This device typically employs a NaI detector as its basis, though the design is modular and the NaI crystal can be swapped if desired. The HM-5 is battery powered, easy to operate, and can measure dose rate, search for sources, identify isotopes, and determine the enrichment of non-irradiated uranium materials [1]. The device also has non-volatile memory enabling the post-processing of up to 50 gamma spectra, each with 1024 channels. The primary drawback of these devices is the notoriously poor energy resolution of NaI that limits its use for certain isotopic analysis. However, the versatility and convenience of this device is unparalleled, and it is typically carried by inspectors as part of their inspection toolkits [1].

Germanium detectors are a valuable tool for high resolution gamma spectrometry, used to determine precise isotopics of materials. HPGe detectors fall into the semiconductor category and require very low operating temperatures and very pure germanium to achieve the high-resolution gamma spectra for which they are known. The performance of these detectors suffers in presence of these impurities, and it also suffers if the device is not cooled to an appropriate operating temperature. At room temperature, electrons can receive enough energy to transfer to the conduction band through thermal excitation, which ultimately results in noise in the spectrum. Cooling of the system ensures only photons with sufficient energy are able to excite electrons across the band gap.

Conventionally, HPGe systems are bulky and require a supply of liquid nitrogen to provide adequately cool operating temperatures. ORTEC's Micro-trans-SPEC system eliminates the need for liquid nitrogen cooling and condenses the detector electronics into a handheld, electrically cooled spectrometer. This commercially available system also includes safeguard enabling software for IAEA inspectors and is known as the electrically cooled germanium system (ECGS) [1,5,6]. The ECGS design maintains resolution comparable to conventional HPGe systems but with a more convenient footprint [6]. The elimination of liquid nitrogen also opens the door for a continuous unattended monitoring, high-resolution gamma spectrometry

system, since the ECGS can stay cool as long as it has power. Overall, the ECGS is a valuable, versatile NDA instrument in an inspector's toolkit.

Battery powered and portable multichannel analyzers (MCAs) are also leveraged to support detector mobility including the InSpector 2000 and MMCA (miniature multichannel analyzer). These devices can be paired with any type of detector as part of the readout electronics and data acquisition systems and are often used with $LaBr_3$, CdZnTe, or HPGe detectors [1]. $LaBr_3$ is a scintillation type detector similar to NaI but with better energy resolution. CdZnTe is another type of semiconductor detector that offers one significant advantage over HPGe detectors: room-temperature operation. The tradeoff of CdZnTe is inferior energy resolution relative to HPGe, however the resolution is still superior to that of scintillators. These portable MCAs paired with detectors are also easily combined with small laptop systems with analysis software such as the Multi-Group Analysis (MGA) software or the Fixed-Energy Response Function Analysis with Multiple Efficiency (FRAM), creating a cohesive and powerful measurement and analysis unit [3]. While these systems are relatively accurate and provide much needed mobile isotopic analysis of uranium and plutonium samples, they are still relatively bulky and lack the convenience of handheld systems.

4 ADVANCED GAMMA NDA: THE SOFIA MICROCALORIMETER

The Spectrometer Optimized for Facility Integrated Applications (SOFIA) is a gamma spectrometer based on low temperature microcalorimeters, developed to enable ultra-precise non-destructive nuclear material analysis. The design is centered around a tabletop cryostat system as shown in Figure 2, providing a compact system compared to previous cryostat systems [6]. SOFIA also overcomes the limitations of previous systems with modest infrastructure requirements and the incorporation of advancements in large, multiplexed transition-edge sensor microcalorimeters. The SOFIA cryostat system is cooled with a low-power, air-cooled pulse tube cryocooler requiring only single-phase 220 V power (~3kW) [7]. This infrastructure requirement is comparable to a large window air conditioner and is compatible with nearly any nuclear facility worldwide [7]. Notably, the design of the system eliminates the need for liquid cryogens as well. Detector cooling is facilitated by an adiabatic demagnetization refrigerator (ADR) which can sustain an operating temperature of 90 mK for up to 48 hours at a time. Following the completion of the holding time, the ADR enters an automated two-hour regeneration cycle after which it returns to operating temperature [7].

The detection module is a 256-pixel multiplexed array with a count rate potential of 5000 counts per second. The energy of individual gamma rays that interact with this pixel array creates a heat pulse which is converted into an electrical signal. Measurement times using SOFIA can be comparable to HPGe detectors. The detector is optimized in the 30-300 keV range, with a typical energy resolution of 70-90 eV FWHM for high activity Pu measurements with 15-20 counts per second per pixel. Energy resolution as good as 59 eV FWHM at 97 keV has been demonstrated. Resolution on this order represents five to ten times improvement compared to HPGe systems and approaches the precision and accuracy previously obtainable exclusively though destructive analysis techniques. [6]

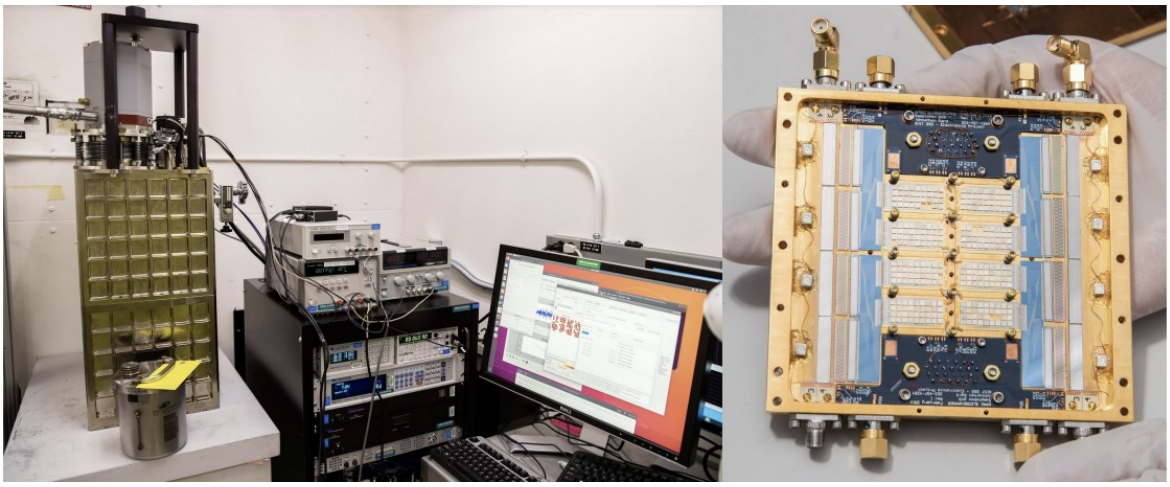

Figure 2: (Left) SOFIA spectrometer and accompanying electronics, with source cannister placed in front of detection window. (Right) Array of 256 superconducting transition edge sensor microcalorimeters used in the SOFIA instrument. [6]

With the ultra-precise NDA capabilities of SOFIA, several safeguards applications for this spectrometer are under evaluation, including: uranium enrichment determination in fresh fuel, spent fuel characterization, determination of actinides in aqueous and electrochemical spent fuel separation processes, and isotopic analysis of purified U, Pu, U/TRU products, and advanced reactor fuels [7].

A measurement campaign performed on varying enrichment samples using SOFIA demonstrates the viability of gamma microcalorimetry when traditional NDA technology is not feasible. Often, uranium enrichment measurements using gamma spectrometry use the Th-234 gamma rays and Th x-rays to determine the ratio of U-235/U-238. These measurements cannot be performed on chemically purified uranium samples until Th decay products reach secular equilibrium with U-238, which occurs approximately six months after purification. However, the energy resolution of SOFIA may provide a method for verifying uranium enrichment in freshly purified material prior to Th equilibrium being reached. Figure 3 shows a resolved U-238 peak at 113.5 keV. Typically, this peak is considered too weak to measure with HPGe. Each sample measured consisted of 200-230 g of U_3O_8 in aluminum cans. It is suspected that optimizing the samples could further reduce Compton scattering background which would enable accurate measurements at higher enrichment levels. [7]

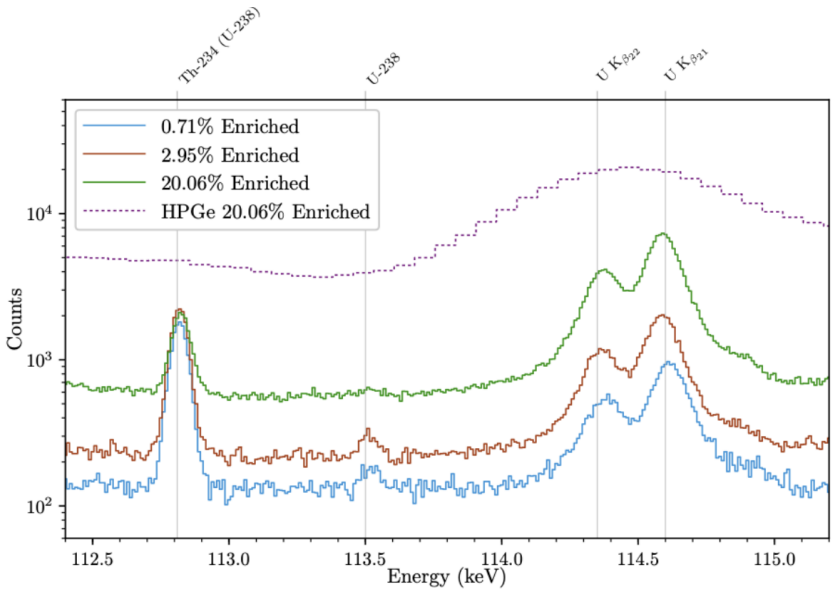

Figure 3: Enrichment measurement campaign using SOFIA, demonstrating the observation of the 113.5 keV peak from the direct decay of U-238. [7]

In a separate measurement campaign, the SOFIA spectrometer was deployed to the LANL Plutonium Facility (PF-4) for a demonstration of NDA capabilities on samples in a working nuclear facility [6]. One notable challenge of performing gamma spectroscopy within a plutonium facility is the Compton scattering background observed in large-mass Pu items. However, despite this challenge the SOFIA instrument performed well. Measurements on low-burnup plutonium with high Pu-242 content achieved well-resolved peaks for two gamma rays resulting from the direct decay of Pu-242 at 103 keV and 159 keV, which have been previously unobservable using gamma spectroscopy on un-modified samples. These results are available in a preprint manuscript through arXiv [8]. Figure 4 depicts the resolved 159 keV peak in comparison to the HPGe results on the same sample.

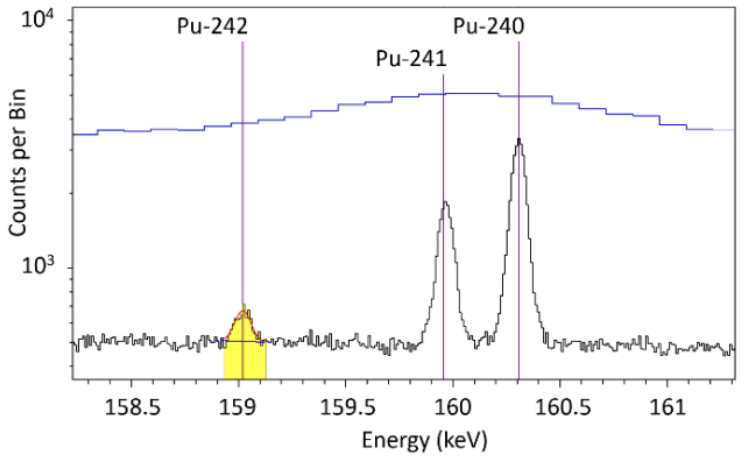

Figure 4: SOFIA spectrum highlighting the 159 keV region resulting from the direct decay of Pu-242, previously unobservable using gamma NDA. Top spectrum in blue shows the HPGe spectrum from the same sample. [8]

The ultra-precise measurements provided by SOFIA have the potential to improve nuclear data, improve confidence in NDA measurements, and reduce the reliance on destructive analysis. SOFIA measurements have demonstrated an improvement in the precision of key nuclear data necessary for non-destructive isotopic analysis such as branching ratios, x-ray line widths, and gamma ray energies [7]. This increase in precision demonstrates SOFIA's potential to improve the performance of HPGe-based isotopic analysis which relies on precise nuclear data. With the high-resolution gamma spectra obtained using SOFIA, weak or closely spaced gamma ray peaks, previously unresolvable and therefore unused, can now be resolved and quantified as well. By resolving closely spaced gamma peaks, SOFIA measurements have the potential to reduce measurement bias and increase confidence in results. The precision and accuracy of these measurements is on par with destructive analysis techniques such as mass spectrometry. By rivaling the precision of destructive analysis, SOFIA can reduce the reliance on techniques that are relatively costly and time consuming. [9]

CONCLUSION

Passive gamma non-destructive analysis technologies are invaluable for safeguards monitoring and verification. NDA has historically been used by the IAEA to perform verification inspections and provides quantifiable assurances regarding the nonproliferation of nuclear materials. The accuracy (on the order of a few percent) offered by current gamma NDA technologies enables verification of gross and partial defects of materials. For bias defects of repeated small-quantity diversions over time, destructive analysis techniques are typically needed due to their measurement uncertainties of less than one percent.

As discussed, the IAEA has a robust gamma NDA capability, especially when it comes to portable, handheld devices. The HM-5 device, centered around a NaI detector, provides convenient and efficient quantification of uranium enrichment levels, but does not have the resolution to obtain precise Pu isotopics. HPGe devices such as the ECGS have much better energy resolution while still maintaining portability. However, the current fleet of gamma NDA systems do not have the level of precision and accuracy of destructive analysis techniques and therefore leave the IAEA reliant on these time-consuming and costly analyses. Gamma microcalorimetry was highlighted as an advanced addition to current gamma NDA technologies. Specifically, the SOFIA spectrometer demonstrates a 5-10x improvement in energy resolutions compared to HPGe and accuracy previously only obtainable though destructive analysis. The ultra-precise measurements provided by SOFIA have the potential to improve key nuclear data for non-destructive isotopic analysis, increase confidence in NDA measurements, and reduce the reliance on destructive analysis.

ACKNOWLEDGEMENTS

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