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Nuclear Facility Experience with the SOFIA Ultra-High-Resolution Microcalorimeter Gamma Spectrometer

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INTRODUCTION

The first ultra-high-resolution microcalorimeter gamma spectrometers are now being deployed thanks to extensive recent developments in superconducting transition-edge sensor arrays, readout electronics, cryogenics, and software for pulse processing and isotopic analysis. SOFIA (Spectrometer Optimized for Facility Integrated Applications) is a compact instrument that provides 5 to 10 times better energy resolution than high-purity germanium detectors. This technology has the potential to improve the economics, efficiency, and effectiveness of safeguards and nuclear material accounting by providing a nondestructive isotopic analysis method with precision and accuracy approaching that of destructive analysis. SOFIA has recently been demonstrated in the Los Alamos National Laboratory Plutonium Facility for a range of reference and process materials, in addition to measurements of safeguards-relevant plutonium and uranium isotopic standards. While improved energy resolution has a direct benefit in uncertainty of determined isotopic ratios, it has also become clear that improved nuclear and atomic data is needed to realize the potential of this next-generation nondestructive measurement technology. Our team is using available microcalorimeter data from well-characterized isotopic reference materials and gamma-ray energy standards such as Yb-169 to re-determine important values such as gamma-ray branching ratios and X-ray line widths, and incorporate these values into quantitative analysis software. We will discuss our experience with operating the SOFIA instrument in a nuclear facility environment, present quantitative analysis results, efforts to improve nuclear data, and discuss future applications.

DEVELOPMENT OF SOFIA

SOFIA (Spectrometer Optimized for Facility Integrated Applications) was developed by Los Alamos National Laboratory, University of Colorado, and the National Institute of Standards and Technology to make ultra-high-resolution microcalorimeter gamma spectroscopy practical for

routine use in nuclear facilities and analytical laboratories. Compared to other microcalorimeter instruments, SOFIA is a much more compact system based on a tabletop cryostat (Figure 1, left). Instead of using liquid cryogens or a large pulse tube refrigerator that requires three-phase electrical power and cooling water, SOFIA uses a low-power pulse tube that requires only single-phase 220V AC similar to a large window air conditioner. Its solid-state adiabatic demagnetization refrigerator eliminates the complexity of a dilution refrigerator. A 256-pixel superconducting transition-edge sensor microcalorimeter array (Figure 1, right) provides typical energy resolution of 60-80 eV FWHM at 129 keV and count rates up to 5000/s. Further details on the instrument are provided in [JNMM2020].

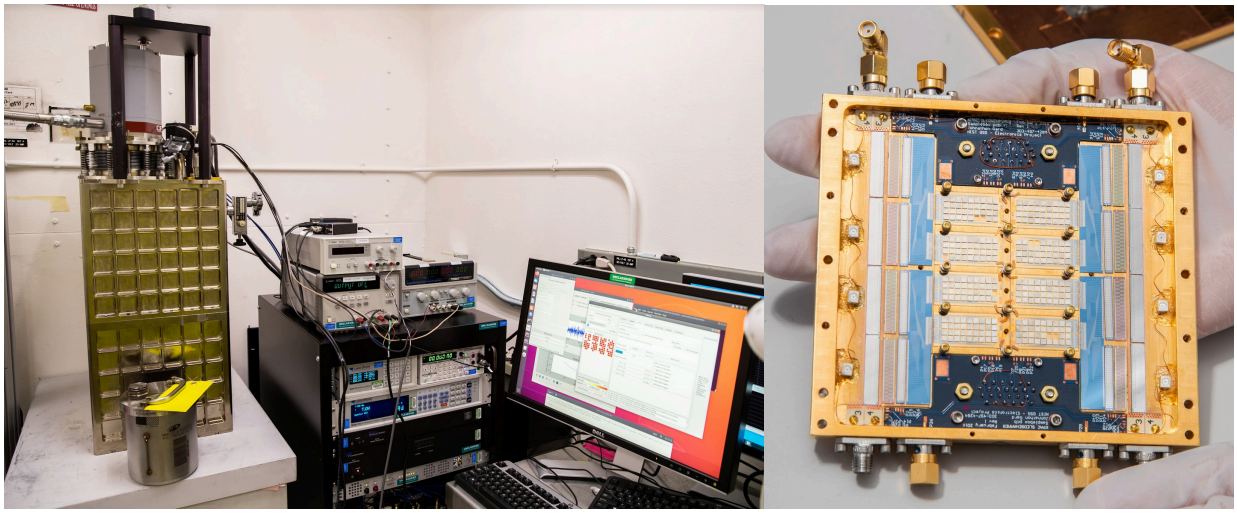

Figure 1: (Left) The SOFIA microcalorimeter gamma spectrometer uses a tabletop cryostat with a low-power pulse tube and adiabatic demagnetization refrigerator. (Right) An array of 256 superconducting transition-edge sensor microcalorimeters provides sufficient detection efficiency and count rate capability to measure nuclear material items in typical packaging.

DEPLOYMENT TO THE LANL PLUTONIUM FACILITY

In the summer of 2021, SOFIA was deployed to the Los Alamos National Laboratory Plutonium Facility (PF-4) for a demonstration of its nondestructive isotopic analysis capabilities on a range of items in a working nuclear facility. The major components of the system were easily loaded into a truck for transport (Figure 2). The electronics rack and pulse tube compressor are mounted on casters, and the cryostat can be placed onto a cart by two people. The pulse tube compressor was able to use an existing 220V AC receptacle and the supporting electronics used standard 120V 60 Hz AC power. Operation of the instrument was evaluated and determined to be within the existing work authorization for nondestructive assay measurements and there are no special hazards. Upon connecting the components of the system, cooling down to the detector operating temperature took approximately 32 hours. This relatively long cooling time is a consequence of using a low-power pulse tube cryostat. However, the system can remain cold essentially indefinitely as long as electrical power is supplied because items to be measured remain outside the cryostat. A two-hour regeneration cycle of the adiabatic demagnetization refrigerator is needed before up to 30 hours of detector operation. The regeneration cycle can be scheduled in software so that the system is ready for measurements at the beginning of a shift.

Figure 2: SOFIA loaded for transport to the LANL Plutonium Facility.

OPERATION IN THE FACILITY

Upon installation, SOFIA was tested to verify that it met the same performance specifications in the facility as in the development laboratory. Electrical noise levels and energy resolution were found to be as expected even in the presence of other nearby equipment. The development team trained plutonium facility staff during initial measurements, and the facility staff quickly became comfortable running the instrument independently. Detection efficiency proved to be more than adequate to measure typical items in standard packaging. As shown in Figure 3, items were most often plutonium oxides in SAVY containers. The measurement procedure was very similar to that used for germanium detectors. Items were placed on the table next to the instrument and in front of the side-looking detector window. The position of the item was adjusted to give a total count rate of 1000-2000 events per second. While the instrument is capable of higher count rates, energy resolution can degrade in high-rate measurements due to pulse pileup and crosstalk between individual sensor elements, similar to effects observed in germanium detectors. For some materials, cadmium filters were placed in front of the detector window to attenuate the intense 59.5 keV gamma ray from ^{241}Am and provide more uniform gamma peak intensities across the recorded spectrum.

Measurements have focused on low-burnup plutonium items and material with high ^{242}Pu content. An example spectrum is shown in Figure 4 for the Pu isotopic standard. This item consists of approximately 400 g of Pu oxide in a SAVY container. For materials with high ^{242}Pu content, observation of two gamma rays from the direct decay of ^{242}Pu were observed at 103 keV and 159 keV. A manuscript describing ^{242}Pu results is in preparation with a preprint available on arXiv [arXiv 2022]. Data analysis has typically been done after acquisition, although software for real-time processing of microcalorimeter data into an energy-calibrated spectrum is in development and has been demonstrated on microcalorimeter X-ray spectrometers at LANL. The SAPPY isotopic analysis code was used for determining ratios of the Pu isotopes and ^{241}Am. One challenge in gamma spectroscopy for items in the plutonium facility is the Compton scattering background

which is observed for large-mass Pu items. Work is in progress to use SOFIA data to improve analysis in the low-energy region (60-208 keV) where this background is significant, but is also where the most intense gamma rays from the major plutonium isotopes are found. Preliminary results suggest that if key nuclear data can be improved, especially gamma-ray branching ratios, the overall uncertainty on major Pu isotope ratios can be reduced to below 1% with SOFIA.

Figure 3: Items to be measured were placed next to the detector window on the side of the cryostat. Typical packaging consisted of SAVY containers.

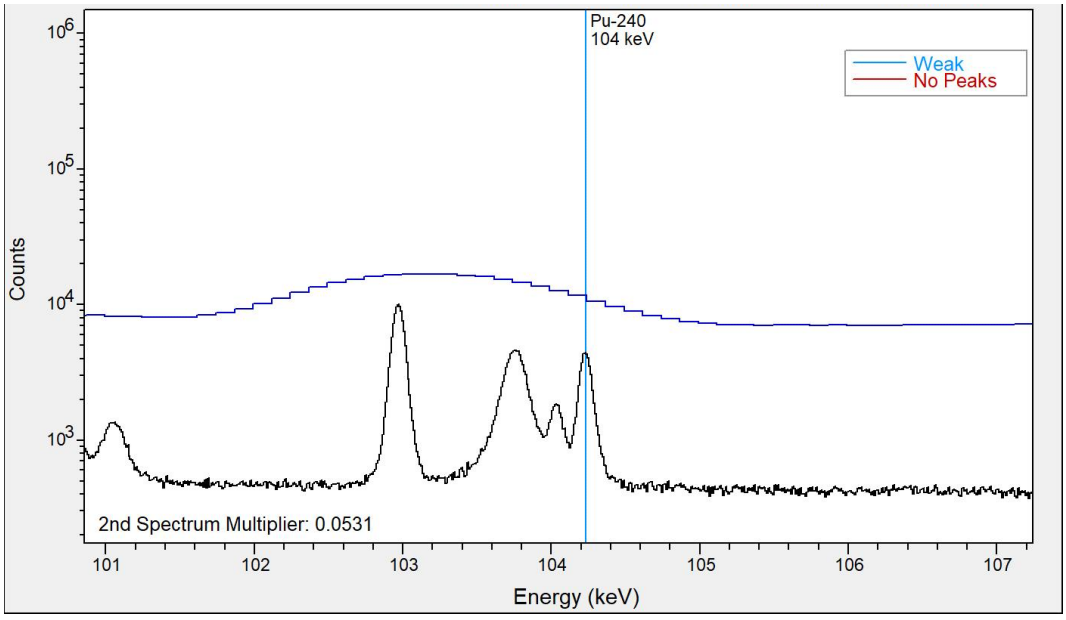

Figure 4: Spectrum of Pu isotopic standard measured with SOFIA, compared with results from germanium detector used for far-field gamma measurements.

DETECTOR CALIBRATION

While improved energy resolution is already reducing the uncertainty of isotopic ratios, it is becoming increasingly clear that improved nuclear and atomic data are needed to fully realize the potential of this next-generation nondestructive measurement technology. SOFIA will therefore also be used to improve the branching ratios of the emission signatures from the actinides of interest. The attainable uncertainties depend on the accuracy of the calibration lines that are used to extract the efficiency corrections of the detector in the region of interest. For U and Pu, the primary region of interest extends from 90 to 130 keV where the isotopes of interest have relatively closely spaced signature emissions (Figure 4). Yb-169 emerges as the best-suited isotope for detector calibration in this region because it has four strong gamma lines at 93.61447(7), 109.77924(3), 118.18940(14) and 130.52293(4) keV whose branching ratios have been characterized with very high accuracy [Helmer 2000, Morel 2000]. Additional gamma lines at 177.21307(4) and 197.95675(4) keV can be used to extend the calibration to higher energies if needed.

Three factors then contribute to the total detection efficiency ε_{total}: Self-absorption inside the source, transmission of the filter between source and detector, and quantum efficiency of the detector. In the ideal case, they can be described by the analytical expression

$$\varepsilon_{total} \propto \frac{1 - e^{-\mu_{Source}\rho_{Source}d_{Source}}}{\mu_{Source}\rho_{Source}d_{Source}} \cdot e^{-\mu_{Filter}\rho_{Filter}d_{Filter}} \cdot \left(1 - e^{-\mu_{Absorber}\rho_{Absorber}d_{Absorber}}\right)$$

Here μ_{Source}, ρ_{Source} and d_{Source} are the mass attenuation coefficient, density and thickness of the source. If the source is sufficiently thin so that self-absorption is negligible, the first term reduces to 1. The terms μ_{Filter}, ρ_{Filter}, d_{Filter} and $\mu_{Absorber}$, $\rho_{Absorber}$, $d_{Absorber}$ are the corresponding functions for the filter between source and detector and the absorber material. Note that this equation only contains products of μ, ρ and d for the material constants. Since neither dimensions nor densities are typically known with an accuracy below 0.1%, the calibration equation effectively contains (at most) three parameters that can be slightly adjusted to fit the observed efficiency curve, plus a common factor to set the overall scale.

Experiments with Yb-169 have shown that this analytical expression can be used to describe the efficiency of microcalorimeters in the region of interest between 90 and 130 keV within the statistical accuracy of the initial measurement. Under these circumstances, efficiency corrections will only add uncertainties of order 0.1% to isotopic ratios for sufficiently closely spaced lines [Friedrich 2021]. We expect to measure relative branching ratios with SOFIA with comparable uncertainties. These measurements with higher statistical accuracy will show the extent to which the analytical expression is valid before higher-order corrections will have to be included.

SUMMARY

The SOFIA microcalorimeter gamma spectrometer is the first instrument of its type successfully demonstrated in a working nuclear facility. Experience in the LANL Plutonium Facility has shown that microcalorimeter technology can be deployed for routine measurements of nuclear material items in typical packaging. The instrument has been running reliably in the facility since August 2021 and has obtained ultra-high-resolution gamma spectra of a wide range of items in typical

packaging. As microcalorimeter instruments like SOFIA become more widely available, they are expected to improve the efficiency of facility operations by providing a completely nondestructive, relatively rapid means of precise isotopic analysis to meet safeguards and material accounting goals.

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