Ultra-high energy resolution microcalorimeter gamma ray spectrometer for high precision nuclear fuel cycle research

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Abstract

Nuclear energy depends on the development of new reactor technologies which are increasingly safe, secure, and economical. In support of this mission, the Materials and Fuels Complex (MFC) Analytical Laboratory at Idaho National Laboratory provides sophisticated analysis of nuclear materials. An important component is nondestructive assay of fresh and spent fuel, reactor components, radioactive waste, and other materials from the nuclear fuel cycle. This assay includes gamma ray spectroscopy, which measures isotopic composition. For an accurate measurement, a gamma ray spectrum with high resolution and high counting statistics is desirable. However, such spectra can be difficult to obtain from the materials to be studied, which often consist of very dilute samples. Here we present a new gamma ray spectrometer, the High Efficiency and Resolution Microcalorimeter Spectrometer, 400 pixels (HERMES-400), developed at Los Alamos National Laboratory in collaboration with the National Institute of Standards and Technology and University of Colorado, installed in the MFC in 2022. The spectrometer uses a microcalorimeter array, a gamma ray detector with unparalleled energy resolution. Microcalorimeters are sensors which operate below 100 milliKelvin and respond sensitively to the absorption of a photon. This can give them energy resolution on the order of 60-80 eV photopeak FWHM (full width half maximum) in the 50-250 keV range. The detector is mounted within a dilution refrigerator, and can measure continuously, permitting multi-day measurements with thousands of counts per second. This gamma ray spectrometer permits precise, high statistics measurements on small amounts of novel nuclear fuel cycle materials. We will discuss the development of this detector and present spectra demonstrating its capabilities.

Introduction

As nuclear fuel cycle research expands more and more rapidly, the need for precise analysis of the composition of experimental nuclear fuel materials increases. This includes experimental types of fresh fuel, spent fuel, byproducts of new reprocessing techniques, and irradiated reactor infrastructure. In many cases, to satisfactorily characterize nuclear and radioactive material related to these processes, destructive analysis such as thermal ionization mass spectrometry or inductively coupled plasma mass spectrometry is necessary. These techniques can obtain

measurements of isotopic composition with relative uncertainty as low as 0.05% [1]. However, mass spectrometry requires time-consuming sample preparation and the loss of sample. Gamma-ray spectroscopy, on the other hand, is more rapid, and does not require extensive sample preparation, but relative uncertainty in a measurement of isotopic composition is typically limited to 1% [1].

One avenue towards lower uncertainty in gamma ray measurements is to build detectors with higher energy resolution. Los Alamos National Laboratory, the National Institute of Standards and Technology (NIST), and the University of Colorado Boulder have developed a gamma detector, the High Efficiency and Resolution Microcalorimeter Spectrometer, 400 pixels (HERMES-400), with about an order of magnitude higher energy resolving power than the conventional high purity germanium detector. This detector is installed at the Idaho National Laboratory (INL) Materials and Fuel Complex (MFC) Analytical Laboratory. It uses a technology called microcalorimetry to obtain very high resolution energy spectra, like the Spectrometer Optimized for Facility Integrated Applications (SOFIA), the current state of the art in microcalorimeter gamma detectors [2,3,4]. HERMES-400 can produce spectra with photopeak FWHM of about 100 eV near 100 keV. HERMES-400 is also optimized to make long, uninterrupted measurements with nearly twice as many microcalorimeter pixels as SOFIA. This detector demonstrates the capability for higher statistics in the 200-700 keV range than previously obtained with multiplexed microcalorimeter gamma detector arrays. The detector may distinguish gamma ray photopeaks of actinides from other radioactive isotopes such as fission products, which will improve characterization of materials of interest to the nuclear fuel research community, such as electrorefiner salt solutions.

Transition Edge Sensor Microcalorimeters

HERMES-400 operates using highly sensitive detectors called transition edge sensor (TES) microcalorimeters [4,5]. A TES is a specially microfabricated superconducting sensor coupled to a small tin absorber element which absorbs the incident photons. The TES is cooled down to a temperature of less than 100 mK, close to its superconducting transition temperature. Its resistance has a very steep dependence on temperature in the superconducting transition. When a gamma ray is incident on the absorber of a TES detector, the temperature increases by a small amount, which is measurable as the current change in the TES circuit. The low operating temperature reduces the thermal fluctuations that contribute to Johnson noise, permitting a very low noise measurement, providing the exquisite energy resolution of a TES microcalorimeter.

Each TES has an area on the order of 2 mm by 2 mm and is coupled to a small tin absorber approximately 0.25 mm thick. The small size is necessary to reduce heat capacity, which is crucial to the ability of the TES to measure energy with high resolution. Therefore, to increase detector efficiency to a practical level, we build up detector arrays of tens to hundreds of pixels. Microwave multiplexing [6] permits the readout of several hundred pixels on a single microwave frequency line. Multiplexing necessitates microwave resonator and readout chips near the TES, which effectively convert each TES signal into a subtle change in the frequency of a resonator tone. These tones are then read out and converted to pulses by demultiplexing electronics and a field programmable gate array (FPGA). The pulses are saved in list mode and

processed into a spectrum using optimal filtering techniques [7,8]. The spectrum from each pixel is processed and then coadded into an overall spectrum.

Capabilities of HERMES-400

HERMES-400 implements several changes from the SOFIA detector, with the aim of increasing measurement statistics. First is the operation of the detector module within a dilution refrigerator instead of an adiabatic demagnetization refrigerator (ADR). The dilution refrigerator is a cryogen-free model SD from BlueFors. A pulse-tube compressor provides cooling of the system to 2.8 K. A mixture of ^{3}He and ^{4}He is then condensed within the unit. Below several hundred mK, the ^{3}He and ^{4}He phase separate. The process of ^{3}He crossing the phase boundary is a cooling process, absorbing heat and lowering the temperature of the system below 100 mK [9]. Unlike the ADR, which must be warmed from the operating temperature every few days in order to magnetize and demagnetize a paramagnetic salt, the dilution refrigerator may operate at the TES operating temperature of 85 mK indefinitely, as long as the ^{3}He/^{4}He may circulate. This permits long, uninterrupted measurements. Figure 1 depicts the dilution refrigerator at INL. A gas handling unit, at the right of the photograph, controls the flow of the ^{3}He/^{4}He. A magnetic shield of mu-metal around the refrigerator serves to protect the detector from stray magnetic fields which can trap magnetic flux in the superconducting elements, degrading resolution. A second, cryogenic magnetic shield made of A4K from the company Amuneal is within the dilution refrigerator. The detector modules point downward within the refrigerator, so samples are placed underneath for measurement. The footprint of HERMES-400 is necessarily bigger than that of the SOFIA system [2,3]. The cryostat (center of the left photograph), the gas handling unit (right), the pulse-tube compressor (not pictured), and the associated electronics and computers (left) take up approximately 6 m^2 and is about 2.5 m tall. As such, this detector is intended for permanent installation in the analytical laboratory.

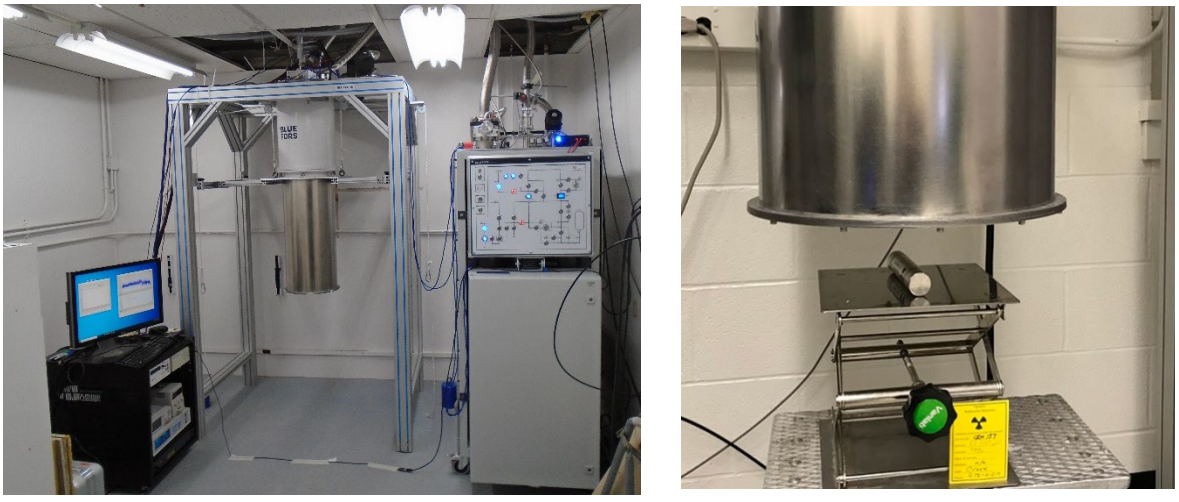

Figure 1: (Left) The INL gamma spectrometer, HERMES-400, in the basement of the INL MFC Analytical Laboratory. The cryostat is the suspended cylinder in the center with its lower half surrounded by a magnetic shield. To the right is the gas handling unit with pumps and valves for the helium gas mixture. To the left is a small electronics rack for data acquisition. (Right) The detector modules face downward, so sources are placed beneath the detector for measurement.

The design of the detector module itself also offers a unique advantage over previous designs [4]. The module is called a microsnout. It modularizes the detector array in such a way that the resonator and readout chips are perpendicular to the TES absorber array (figure 2, left). In this way, the readout chips remain sufficiently close to the TESs, but do not take up space in the active area of the detector. The active area of the microsnout is an array of 100 TESs with tin absorbers, seen on top of the microsnout. In this way, several microsnouts may be arranged close together, increasing the active area of the detector significantly. HERMES-400 will utilize four microsnouts in its final configuration, with a total of 400 pixels. The microsnouts are placed in a protective aluminum sleeve and mounted to a printed circuit board base (figure 2, center). At the time of this writing, up to three microsnouts at a time have been used, and the fourth is expected to be installed mid-2023.

One concern introduced by the cryogen-free dilution refrigerator is the coupling of vibrations from the cold head. Unlike the dense, compact ADR of SOFIA, the dilution refrigerator tends to transmit strong vibrations to the detector modules, severely degrading the performance. We developed a specialized spring mounted holder which isolates the detector modules from the dilution refrigerator body (figure 2, right). Small nylon cords (not pictured) help to damp remaining vibrations. The springs are affixed to an aluminum frame, and a copper mount with copper thermal straps allow the microsnout base at the bottom to have a good thermal connection with the 85 mK stage of the dilution refrigerator.

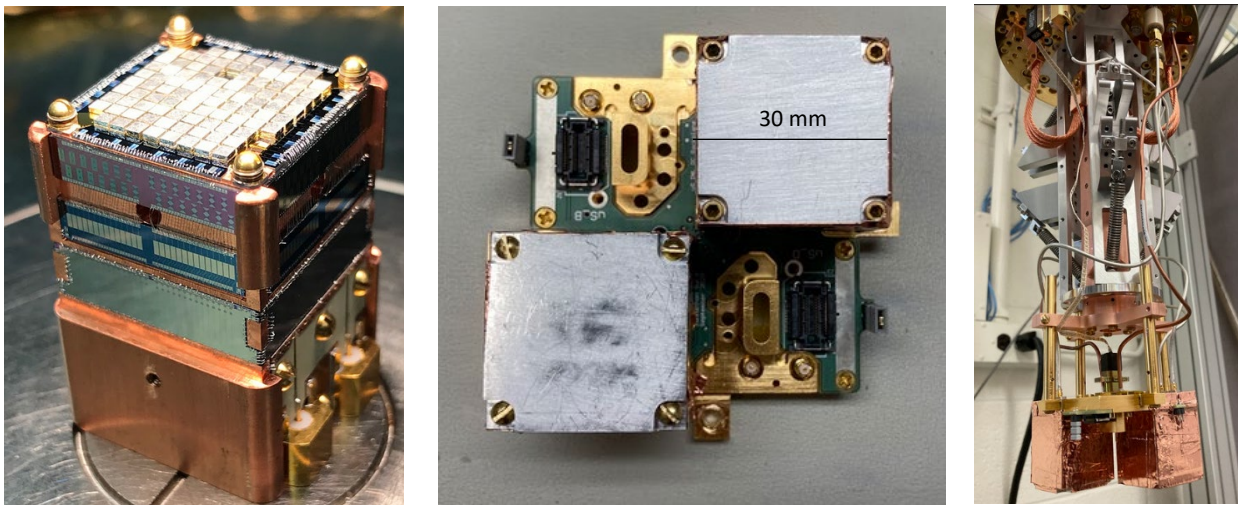

Figure 2: (Left) A microsnout without its protective aluminum shield, revealing the array of tin pixels on top and microwave multiplexing chips around each side. (Center) Two of four microsnouts mounted on the four-microsnout base. (Right) Two microsnouts wrapped in copper tape to enhance thermal conductivity on a cryostat mount suspended by springs. The mount is affixed at the top to the <100 mK stage of the dilution refrigerator.

Measurements on electrorefiner salt solution

Long measurements using one microsnout at Idaho National Laboratory have begun to show the capabilities of HERMES-400. First measurements demonstrated the high resolution of the microcalorimeters. Electrorefiner salts containing Am species and various fission products were measured. Figure 3 shows a measurement of about 3 hours on salt 109773, which was dissolved

and contained in a scintillator vial. Coadded spectra of 51 pixels gives a resolution of 116 eV at 59.54 keV. The highest resolution single pixel spectrum shown here demonstrates a FWHM of 73 eV at the 59.54 keV ^{241}Am peak (figure 3, inset). Figure 3 also shows Sn x-rays and escape peaks below 35 keV, and an ^{243}Am peak at 74.66 keV, which are discussed in greater detail later.

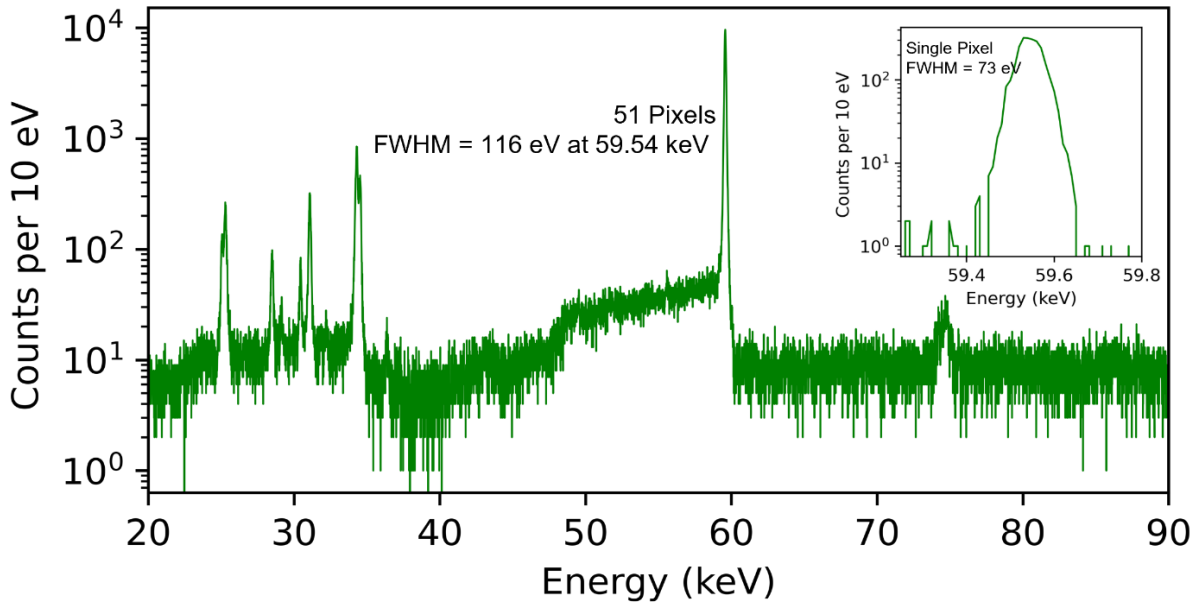

Figure 3: Spectrum of an electrorefiner salt, 109773, obtained at Idaho National Laboratory. The prominent 59.54 keV ^{241}Am peak has a FWHM of 116 eV from a coadded spectrum of 51 pixels. (Inset) The highest resolution single pixel spectrum has the 59.54 keV peak with a FWHM of 73 eV. There are also Sn x-rays and escape peaks below 35 keV, and an ^{243}Am peak at 74.66 keV in this figure.

HERMES-400 shows an exciting promise for high statistics spectra at energies from 200-700 keV, previously difficult to obtain with microcalorimeter gamma detectors [2-5]. The spectrum in figure 4 is from measurement of a second electrorefiner salt, 109762, which was also dissolved in solution in a scintillator vial. The spectrum, in purple, was taken for approximately 6 days, using the best 13 pixels from a single microsnout. A high purity germanium (HPGe) spectrum is also shown in orange. This salt was shown by previous analysis with the high purity germanium spectrum to contain ^{154}Eu, ^{155}Eu, ^{137}Cs, ^{134}Cs, ^{241}Am, ^{243}Am, and ^{239}Np. The ^{239}Np is present in this sample because it is the daughter decay product of ^{243}Am. In the microcalorimeter spectrum, there is a photopeak at 661.66 keV from ^{137}Cs, and a very large Compton background. We may still observe the ^{239}Np peaks at 228.18 keV and 277.6 keV above the Compton background. We also see the ^{154}Eu peak at 591 keV and ^{134}Cs peak at 604 keV. The 661.66 keV peak is used to calibrate the spectrum, and has a FWHM of about 200 eV. Notably, this peak is necessarily a convolution of the ^{137}Cs peak and the ^{241}Am peak at 662.4 keV. The calibration of the microcalorimeters is especially non-linear in this high energy region due to the nature of the superconducting TES response. Because of the worsened resolution, and still relatively low statistics, we do not resolve these peaks with confidence. In the near future we aim to improve

measurement conditions with increased numbers of pixels in maximize measurement capabilities at higher energies. Figure 5 and 6 provide details of the many low energy peaks.

This spectrum is significant because actinide photopeaks measured by microcalorimetry well above 200 keV may be used in characterization of isotopic percentages, even in the presence of a large Compton background. Observing such high energy peaks has previously been difficult in short microcalorimeter measurements limited by the regeneration requirements of an ADR. Using additional peaks at high energies will contribute to increased accuracy in such calculations. There are several smaller ^{239}Np in the high purity germanium spectrum in the Compton background region which are not easily visible in the microcalorimeter spectrum. At the time of this measurement, only one microsnout was operating with only a subset of pixels providing high quality spectra. It is likely that a similar measurement with the majority of the 400 pixels, with improved analysis, would reveal more peaks, and indeed this is a major goal of immediate future work.

Additionally, the presence of the Cs peaks in the microcalorimeter is valuable. It implies that the ^{137}Cs/^{134}Cs radiochronometry ratio can be used in microcalorimeter spectra, along with the other promising ratios ^{243}Am/^{241}Am, and ^{155}Eu/^{154}Eu. The three quantities together might be used towards decoupling and understanding both the burnup time of a material in a reactor and the time since irradiation.

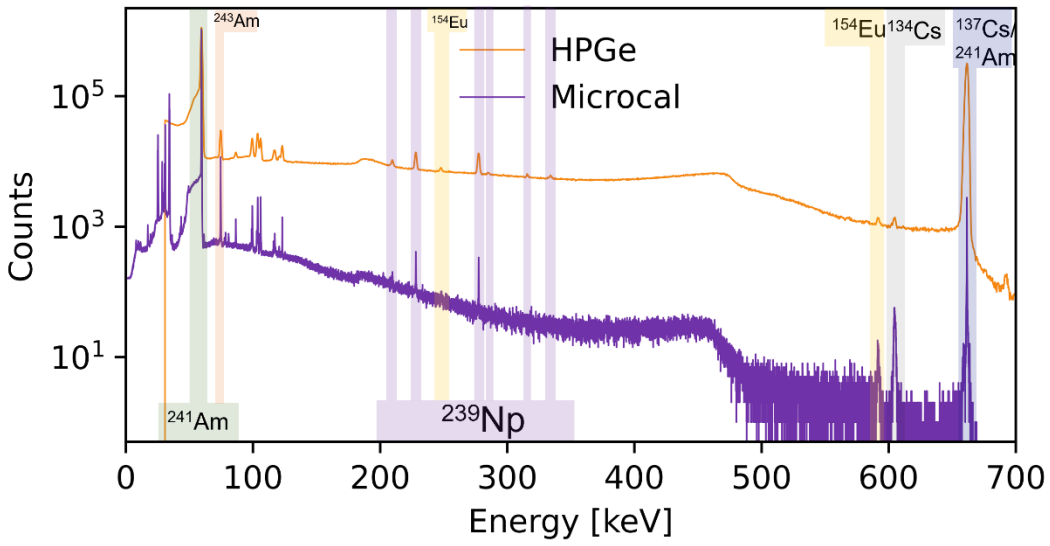

Figure 4: Spectrum of electrorefiner salt 109762. The ^{137}Cs and ^{241}Am convolved photopeak near 662 keV is the highest peak in the spectrum, and gives rise to a significant Compton background. The microcalorimeter spectrum is shown in purple, and the HPGe spectrum is in orange. Figure 5 and 6 show detail of the lower energy regions of the spectrum.

A practical and promising use of the detector is to resolve actinides from fission products and x-rays. In figure 5, we see the region in which ^{239}Np peak at 106.13 keV is very near the ^{155}Eu peak at 105.31 keV. These two peaks are resolvable with the spectrum obtained with the microcalorimeter detector. SOFIA has also previously demonstrated its ability to resolve these

two peaks, which are of interest in improving actinide quantification in spent fuels. We also see that in this region, we may resolve the ^{241}Am peaks from the nearby Pu x-rays at 98.97 keV and 102.98 keV, which can provide major constraints on the actinide quantification in this material. There are also Sn escape x-rays from the ^{154}Eu peak at 123 keV, resulting from the tin absorbers of the microcalorimeter array, which we can resolve from the lower ^{241}Am peak. The Pu x-rays result from the ^{239}Np beta decay. No Pu gamma rays are distinguishable; for example, the ^{239}Pu gamma ray at 129.3 keV cannot be discerned. The Pu gamma rays may not be apparent due to attenuation or interference with more prominent peaks. However, we identify what appear to be two weak U x-rays, which are not visible in the high purity germanium spectrum, indicating that there is some Pu in the solution. Understanding the actinide content of electrorefiner salts is crucial to validating the viability of the electrorefinery process, and this microcalorimeter spectrum shows that we may resolve actinide peaks not discernible by high purity germanium.

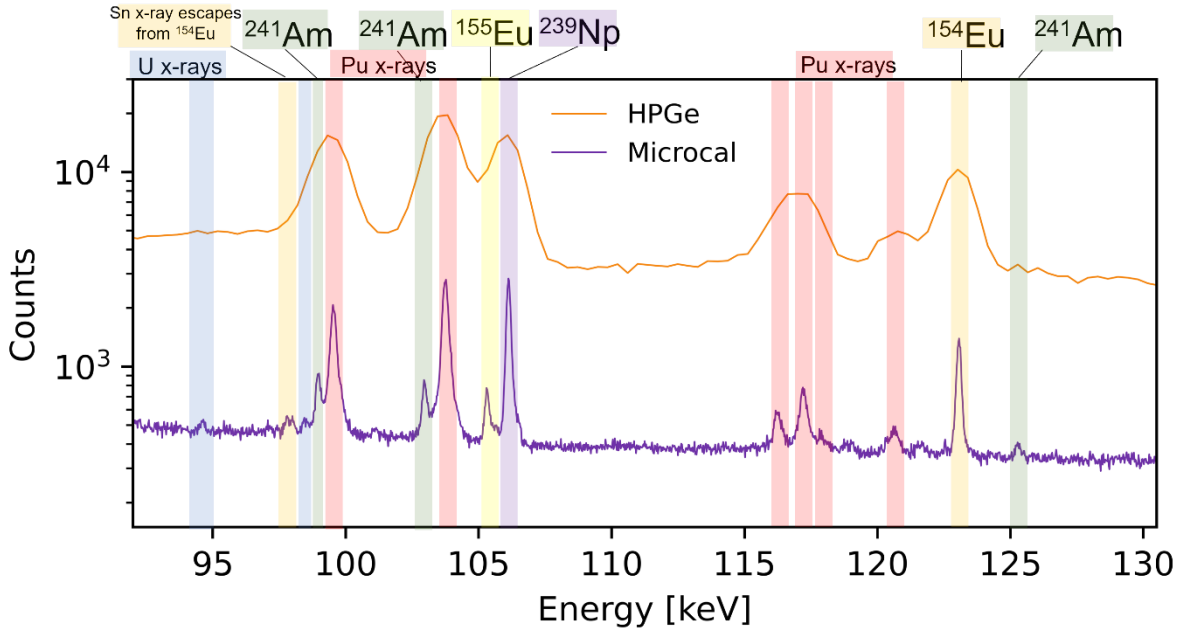

Figure 5: Spectrum of electrorefiner salt 109762. The microcalorimeter spectrum is shown in purple, and the HPGe spectrum is in orange. The gamma ray peak from ^{155}Eu at 105.31 keV is notably distinguishable from the ^{239}Np peak at 106.13 keV, and ^{241}Am peaks are distinguishable from adjacent x-rays.

Finally, microcalorimetry may also provide insight into the low energy region of the spectrum. Microcalorimeters may measure at lower energy than many high purity germanium detectors, as low as 15 keV. Figure 6 demonstrates the low energy region of the electrorefiner salt spectrum 109762. This high purity germanium spectrum ends around 30 keV, with very few features noticeable except the ^{241}Am peak, ^{243}Am peak, and ^{155}Eu peak. The microcalorimeter spectrum, on the other hand, reveals a wealth of features. Many are x-rays and x-ray escape peaks due to the tin absorbers of the microcalorimeters. Most prominent are the tin K_α and K_β x-rays and the corresponding escape peaks from the 59.54 keV ^{241}Am peak, which are mirrored about 29.77 keV. However, there are several real and valuable peaks originating from the electrorefiner salt.

Ba and Gd x-rays from decays of the Cs and Eu isotopes, respectively, are seen. A small ^{241}Am peak at 43.42 keV is observable near these x-rays. Below 20 keV, there is evidence of what is likely to be Pu L x-rays. Because many isotopes of interest have gamma rays below 50 keV, this region of the spectrum is of great interest. In HERMES-400, a 0.020 inch layer of cryogenic magnetic shielding inhibits the efficiency somewhat. We anticipate replacing this layer with an 0.004 inch layer, which will increase the transmission by nearly a factor of 10 below 40 keV.

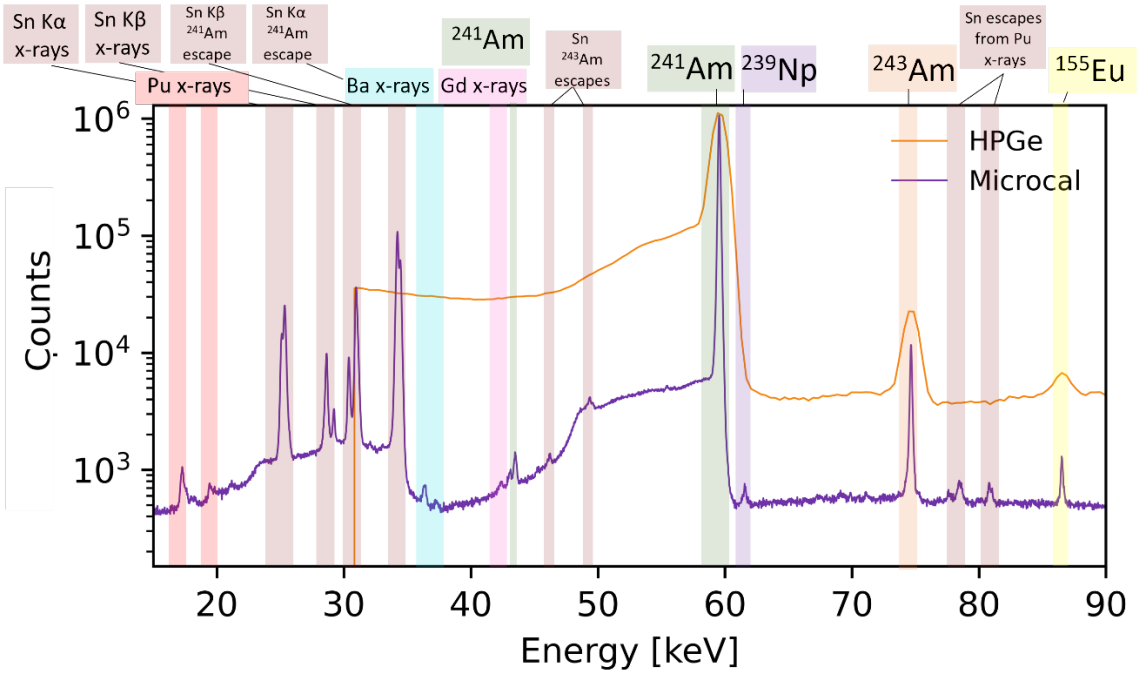

Figure 6: Spectrum of electrorefiner salt 109762 below 90 keV. The microcalorimeter spectrum is shown in purple, and the HPGe spectrum is in orange. Sn x-rays and x-ray escape peaks, due to the tin absorbers on the TES pixels, make up many features in this energy region, shown in light brown. Several actinide peaks as well as x-rays from decay products Ba and Gd are also visible.

Future Work

HERMES-400 has performed exciting preliminary measurements, and there are several planned improvements. First, all four microsnouts with 400 total pixels will be installed, which will allow HERMES-400 to fulfill its full capabilities for high efficiency measurements. Second, we intend to reduce the amount of cryogenic magnetic shielding within the cryostat from 0.020 inches to 0.004 inches in front of the detector face, which as discussed will improve the efficiency below 40 keV by nearly a factor of 10. Although the energy resolution may suffer slightly due to the reduced magnetic shielding, many gamma rays of interest are in this energy region. Third, tests to optimize the vibrational and magnetic field environment are ongoing, and are necessary to improve the energy resolution of the detector to a more desirable level. Current possibilities for this improvement may entail the replacement of fasteners and springs near the microsnouts with less magnetic components, the implementation of a Kevlar suspension system for the

microsnouts to reduce both magnetic and vibrational issues, and the addition of mass to the cryostat frame to reduce the vibrational coupling to the microsnouts.

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