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High-resolution microcalorimeter detectors as a tool in the future of nuclear safeguards

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Abstract

New measurements are presented from the LANL-NIST micorcalorimeter array for two standard plutonium sources. The results demonstrate substantially smaller error bars obtained from the spectral analysis program FRAM. Some areas of improvement to the analysis technique have been identified, indicating that the microcalorimeter results can be improved upon. These results support the viability of a device for performing real nuclear safeguards measurements in the near future.

1 Introduction

The challenge of providing reliably accurate and precise data is a critical component of any safeguards initiative. In the realm of nuclear safeguards, this is an especially daunting task since inaccurate and/or imprecise data could have very serious international consequences. As such, there is a constant drive within the community to establish better measurement and analysis techniques in order to further reduce the associated errors and uncertainties. Even with todays state of the art equipment, measurement uncertainties can extend to

several significant quantities worth of material over a relatively modest period of time [1, 2]. Furthermore, there is a strong desire for improved nondestructive analysis techniques in order to reduce both the cost, turnover rate, and inconvenience of destructive analyses [3].

One promising new technology that may help to realize these goals is that of gamma-ray microcalorimeter detectors. The hallmark quality of this new technique is the ability to achieve energy resolution nearly an order of magnitude better than typical planar high-purity germanium (HPGe) detectors. Such an improvement may help reduce uncertainties associated with, for instance, plutonium isotopics or uranium enrichment measurements. This may, in turn, help to reduce uncertainties in total plutonium and/or uranium content in a given sample without the need for destructive analysis. In this paper, we will describe this new detector technology as well as some recent measurements carried out with the LANL-NIST gamma-ray microcalorimeter (μcal) array. Discussion will focus on the capabilities of this technology as well as the progress toward a practical measurement device.

1.1 Microcalorimeter detector technology

Microcalorimeters operate on the principle that one can measure very precisely the temperature change of a superconducting material at exactly the point where the superconducting-to-normal transition occurs. At this point, the material resistance is a sensitive probe of temperature changes in the material. Thus, a small deviation in temperature will produce a comparatively large deviation in the resistance of the material and thus the current flowing through the device circuitry. The output signal is read out and amplified by a series of Superconducting Quantum Interference Devices (SQUIDs) and subsequently fed into room temperature electronics. The SQUIDs operate within a negative feedback flux-locked loop [4], which has been purported to improve the linearity of pulse amplification [5].

To reduce the heat load at operating temperatures near 100 mK, several sensors must be multiplexed into the same output channel. We use a time-domain multiplexing scheme which has so far been met with the most success in terms of the simultaneous readout of many detector pixels compared with other methods like frequency-division and code-division multiplexing [6]. Still, limitations do exist on the simultaneous readout of many pixels and this area continues to see substantial improvements.

To stop hard x-ray and soft gamma ray photons, a tin absorbing material

is attached to a superconducting Mo/Cu transition edge sensor (TES). The energy resolution achieved with these devices is proportional to $\sim 2T\sqrt{k_b C}$, where T is the temperature and C the heat capacity of the absorbing material. Because of this delicate relationship between energy resolution and absorber heat capacity, current gamma-ray microcalorimeter systems are generally limited to the detection of relatively low energy gamma-rays.

The latest installment of the LANL-NIST detector array presently consists of 256 individual pixels. Approximately half are designed for an energy range up to ~100 keV, and about half to ~200 keV. Several pixels were designed for even higher dynamic range, but for the purposes of this discussion they will be treated as 200-keV sensors.

2 Performance and recent measurements

With the recent system upgrade to a 256-pixel array, and the ability to simultaneously multiplex a large number of working sensors, it has become possible for the first time to carry out high-statistics measurements within a reasonable timescale. Recent measurement campaigns have focused on obtaining high-statistics plutonium data and making direct comparisons with the analytical capabilities of planar HPGe detectors.

Of the 256 pixels, 169 were operational, and 104 achieved energy resolution under 100 eV for the 103-keV gamma-ray peak in ^{153}Gd. For Pu data, the multiplexing capabilities are more limited, since the presence of large pulses provides a higher probability of disrupting the flux-locked loop. Hence, the Pu measurement campaign was carried out with the simultaneous operation of 64 pixels per run. Of these, typically ~30 produced spectroscopic-quality data.

In this paper, we describe a portion of the results obtained from two standard plutonium sources. The first, initially prepared for the Plutonium Isotopic Determination Intercomparison Exercise (PIDIE) [7], is denoted here as PIDIE-3. This source is ~0.4 g plutonium oxide contained inside a stainless steel casing with a thin window and collimator at its face. The second source was prepared as a mixed oxide source and will be denoted MOX-21 [8]. This source was doubly contained within welded stainless steel cans which measured on the outside 6.6 cm diameter × 16.4 cm high and was composed of 59.16 g plutonium, as well as a small amount of uranium. This source was substantially more intense than the PIDIE-3 source. The spectra obtained from each source is presented in Figure 1. Further details from the entire measurement

campaign will be published in Refs. [9, 10].

2.1 Spectral analysis

The spectra obtained from the Pu measurements were analyzed for Pu isotopic content with FRAM [11]. The HPGe data were analyzed with previously defined parameter sets *upu60-210solidx* and *planar-widerange3*, where the former utilizes the more intense but convoluted low-energy region of the spectrum, and the latter utilizes only higher-energy peaks above the k-edge. In general, the low-energy region is thought to yield superior results at shorter measurement time, whereas the higher-energy region is better suited for longer measurements. The microcalorimeter data were analyzed with a custom parameter set that resembled closely *upu60-210solidx*. The results are presented in Table 1. Note that the primary result depicted in the table is $R_{240/239}$, the ratio of ^{240}Pu content to ^{239}Pu content. The number of counts in the 104-keV peak from ^{240}Pu, determined from a peak fit reported by FRAM, is used as a measure of spectrum statistics.

Whereas the measurement time required to obtain identical statistics is considerably higher for the microcalorimeter system, it is clear that the single-measurement error bar obtained with these spectra is far superior to what is achieved with the HPGe detector at a comparable level of statistics for both sources. Furthermore, at HPGe measurement time of 227 hours on the PIDIE-3 source, the reported error bar from spectral analysis with the *upu60-210solidx* parameter set is comparable to that achieved with μcal, and with the *planar-widerange3* parameter set the reported value is better. However, one should note that the value for $R_{240/239}$ obtained with *planar-widerange3* exhibits a considerable offset, 6σ from the reference value as reported in Ref. [7]. On the contrary, the μcal result is reported to within 1σ of the reference value.

The results obtained with the MOX-21 source were similar to those obtained with the PIDIE-3 source in the sense that the reported error bar on the μcal measurement is considerably smaller for an identical level of statistics. However, in this case the μcal result deviates from the reference value by 3.4σ. The HPGe results, on the other hand, both agree to within 1σ of the reference value.

It should be emphasized here that the FRAM code has been tailored specifically for the application to HPGe data. Since the photopeaks observed in the μcal data tend to exhibit different peak shapes than what is typically seen in HPGe spectra, the peak fit functions often yield poor fits when applied to the

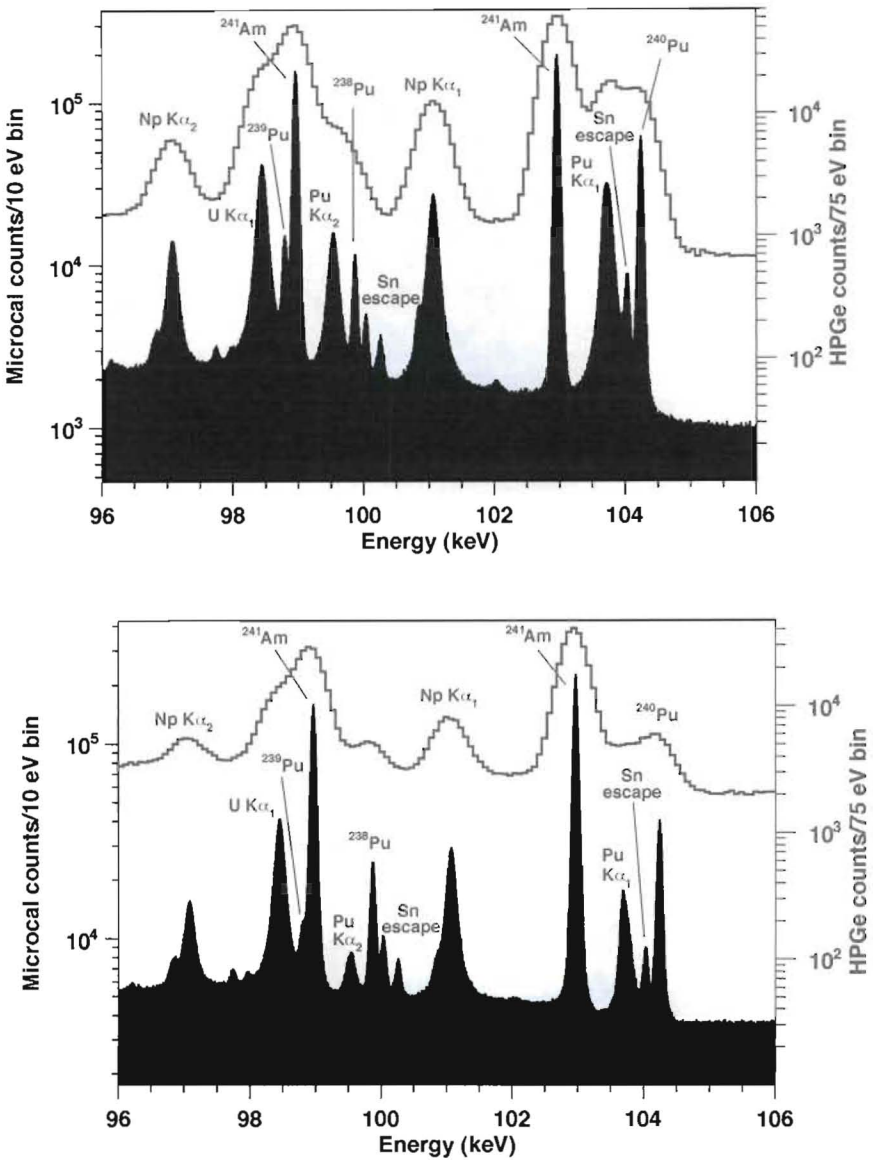

Figure 1: A portion of the spectra from PIDIE-3 (top) and MOX-21 (bottom) plutonium sources. Spectra obtained with HPGe is also presented for comparison.

Table 1: A partial list of the results obtained from plutonium measurements. N_{240} indicates the number of counts in the 104-keV peak from ^{240}Pu. $R_{240/239}$ is the ratio of ^{240}Pu content to ^{239}Pu.

Source		time (hrs.)	N_{240}	$R_{240/239}$	err (%)
PIDIE-3					
	Reference[1]			16.6939	
	HPGe - low	3.75	460968	16.6596	0.58
	HPGe - high	3.75	460968	16.5979	1.03
	μcal	227	449810	16.7123	0.31
	HPGe - low	227	28212207	16.7195	0.32
	HPGe - high	227	28212207	16.5118	0.18
MOX-21					
	Reference[2]			21.2114	
	HPGe - low	6.75	321156	21.3526	1.38
	HPGe - high	6.75	321156	21.0152	1.33
	μcal	242	320982	21.4769	0.37

[1]Reference value obtained from Ref. [7] and dated to 2/16/2010.
[2]Reference value obtained from Ref. [8] and dated to 4/01/2010.

μcal data. This means that, with some improvement to the fit functions the μcal results may improve substantially. Similarly, the shape of the efficiency curve used in FRAM is tailored specifically for the application to HPGe spectra. Modification of this shape may also result in further improvement of the results.

2.2 Toward a practical measurement device

It is clear from the data presented here that the superior energy resolution exhibited by the μcal system yields substantially higher quality spectral analyses. These measurements utilized spectra from ~30 pixels spanning 220+ hours for two standard plutonium sources. Current plans for system upgrades include improved detector design so as to facilitate higher pixel yield, and improvements to electronics and system operation which are expected to improve sensor throughput capabilities. These upgrades may improve readout by nearly an order of magnitude. Additionally, improvements to pixel design may decrease pulse time constants, which are currently of the order several ms. This should allow for higher individual pixel count rates prior to resolution

degradation.

It is difficult to predict exactly how many pixels will be needed to reduce the required measurement time to levels comparable to HPGe detectors. Even under the assumption of static pixel quality (ie: no improvement to pixel design), one must consider the fact that similar or better results can be achieved with the μcal system with considerably fewer statistics, so that it is not necessary to match the count rate capabilities of HPGe detectors. A more appropriate question might posit what is needed to achieve uncertainty limits below what is achievable with HPGe detectors. This question is, similarly, more complex than how it might be initially perceived. Recall that the parameter set *planar-widerange3* produced an uncertainty 0.18% in $R_{240/239}$ after 227 hours of data collection with the HPGe detector and the PIDIE-3 source, but the value $R_{240/239}$ deviated by 6σ from the reference value. An observation of empirical trends indicates that this value will continue to decrease with measurement time, clearly inconsistent with the measurement error. Therefore, it is difficult to establish a lower limit on the uncertainty achievable with HPGe. The μcal results were accurate to within 1σ for this source, but not for the more intense MOX source, and it is unknown whether measurement value and reported uncertainty will be consistent for sources with different isotopic content. For this reason, further investigation is needed to better understand the error analysis reported by FRAM. Work on this topic is ongoing.

Another potential impediment to the implementation of a practical measurement device is the interference of the flux-locked loop caused by large pulses. Preliminary investigations appear to indicate only a small contribution to system dead time due to loss-of-lock events during the plutonium measurements, but the limitations this imposes on the number of sensors which can be simultaneously read out was certainly an issue during this measurement campaign, as it necessitated 64-pixel readout instead of 128, which was easily achieved with the ^{153}Gd source from which the maximum significant gamma-ray contribution is 103 keV. Planned upgrades described above are expected reduce this problem, but it is not known what the effect of potentially high background flux of 300+ keV radiation will have on measurement capabilities.

3 Conclusions

We have presented new data from the recently-upgraded LANL-NIST mircalorimeter array. These data show considerable promise for improved spec-

tral analysis capabilities compared to the current industry standard, high purity germanium. This comes from the superior energy resolution and thus better peak separation in the complex 100-keV region of the plutonium spectrum. Some of the results for $R_{240/239}$ from both HPGe and μcal appeared to deviate from accepted values by several σ, which indicates a possible issue with how uncertainties are reported for high-statistics data in FRAM, or with the parameters used in the analysis. Whereas measurement time is still an impediment to the present μcal system, several improvements are planned for both the hardware and analysis, which should help to bring this technology closer to possible implementation in realistic measurement scenarios.

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