Multiparticle Imaging of Weapons-Grade Plutonium Metal Using an Organic Glass-Based System

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ABSTRACT

Material control and accountability is a vital aspect of the modern nuclear safeguards regime. Special nuclear materials are among the most important substances to track and monitor, leading to various procedures in use today that are specifically meant to localize the materials in a timely manner. Generally, particle imaging systems, like gamma ray Compton cameras or neutron scatter cameras, can assist in these efforts by providing users with angular information that can be used to localize sources. Advanced imaging systems are engineered to simultaneously be sensitive to both gamma rays and neutrons, a beneficial function for scenarios where either particle type's signature is small or nonexistent. This is important due to several materials of interest emitting both fast neutrons and gamma rays, with shielding often being present along with the source. The University of Michigan has recently redesigned our dual-particle imager to feature organic glass scintillators, which were created and obtained from collaborators at Sandia National Laboratories. The composition of this relatively new detector material was designed to enable particle discrimination capabilities and has already shown promise in early imaging experiments. This work will present the first gamma ray and fast neutron images created with the system when measuring kg quantities of plutonium metal. Gamma spectrometry information resulting from single counts in the $CeBr_3$ and double scatter coincident events within the imager will also be reported. These results will demonstrate the capabilities of the new system in conditions more closely resembling monitoring or verification scenarios and will indicate how viable organic glass is for this specific application use.

INTRODUCTION

The Institute of Nuclear Materials Management places great emphasis on responsible stewardship of nuclear materials. This work contributes to the nonproliferation and arms control technical division of the Institution's overall material management domain by demonstrating a detection approach utilizing a novel glass-based dual-particle imaging system. In the realm of safeguards and verification, plutonium serves as a material of great significance due to its designation of special nuclear material (SNM) by the Nuclear Regulatory Commission [1]. Plutonium has both fast-neutron and gamma-ray signatures, allowing for the use of Compton imagers and neutron-scatter cameras in monitoring applications [2,3].

Our group at the University of Michigan has built and tested a new compact dual-particle imager using expertise from previous imaging work [4]. The imager uses an organic glass scintillator material of novel composition obtained from collaborators at Sandia National Laboratories [5]. This paper presents new results where kg quantities of plutonium were imaged with the glass-based imager using both fast neutron and gamma imaging techniques. These new results demonstrate the use of our imager for plutonium detection and localization.

MATERIALS AND METHODOLOGY

Organic Glass Scintillator (OGS)

The chemical composition of the OGS material in the imaging system used during this work is nominally a 90:10 mix of compounds $C_{42}H_{36}Si$ and $C_{51}H_{44}Si$. *Bis*-MSB ($C_{24}H_{22}$), a wavelength shifter, is also included at 0.2 wt%. Further discussion on the chemistry of the OGS, including synthetization methods, can be found in past work done by the Sandia group [5]. The composition of the OGS is significant to this work because it allows for particle discrimination capabilities and previous work has shown that it results in promising light output and timing properties [6,7]. The OGS components in the imager were cast at the University of Michigan using a molding process as shown in Figure 1.

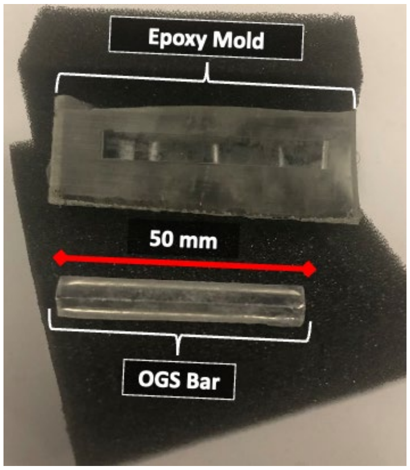

Figure 1: An OGS bar cast at the University of Michigan glass lab. Shown above the bar is the epoxy mold used during the casting process.

Dual-Particle Imaging Methodology

The active volume of the imager is based on an earlier design that was optimized for neutron and gamma-ray imaging using *trans*-stilbene scintillators [8]. The compact imaging system consists of 12 OGS bars (6x6x50 mm^3) and eight encapsulated $CeBr_3$ inorganic scintillator cylinders of 6 mm height and 6 mm diameter. Figure 2 shows a photograph of the assembled active volume. The OGS bars have been wrapped in Teflon to serve as a diffuse reflector and improve light collection. The $CeBr_3$ components are encased in aluminum structures at the corners and coupled to SiPM arrays using compression springs.

Having a combination of both organic and inorganic scintillators allows for dual-particle imaging capabilities. Fast neutrons are imaged using double elastic scatter events between different OGS bars using similar kinematics to traditional neutron scatter cameras. Gamma rays are imaged using sequential Compton scatter events in an OGS bar and photoelectric absorption events in a $CeBr_3$ cylinder. Both methods result an estimate for the particles' incident angle, which is projected onto the surface of a sphere using simple backprojection (SBP) in the form of a cone. A converging algorithm, LM-MLEM, may be applied to a SBP image to further converge on a source location. Equations on the particle kinematics, imaging process, and converging algorithm can be seen in more detail within the seminal paper of the glass imager [9].

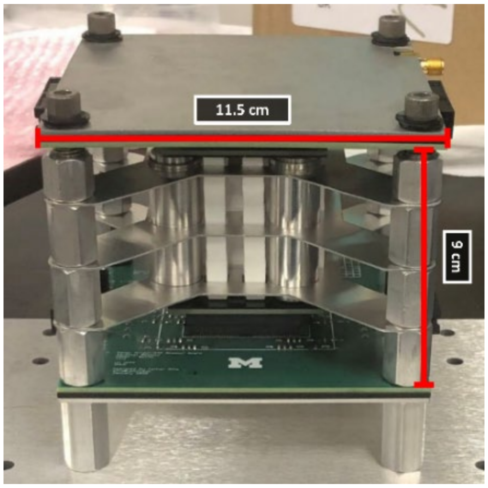

Figure 2: University of Michigan OGS dual-particle imager active volume. 12 OGS bars and 8 $CeBr_3$ cylinders are coupled between SiPM arrays.

Experimental Setup

This work reports on two experiments that were performed at the National Criticality Experiments Research Center. The first experimental setup focused on imaging the BeRP Ball, a ~4.5 kg sphere of α-phase plutonium metal composed of 93.3 wt% ^{239}Pu and 6 wt% ^{240}Pu. The BeRP ball was placed ~90 cm away from the imager at an azimuth and altitude angular direction of (11°, -5.7°) as shown in Figure 3 for 60 minutes. The second experimental setup focused on imaging the Thor core, a ~4 kg piece of δ-phase plutonium metal composed of 94.9 wt% ^{239}Pu and 5.1 wt% ^{240}Pu. The Thor core was placed ~100 cm away from the imager at angular direction of (11°, -0.6°) as shown in Figure 3 for 40 minutes.

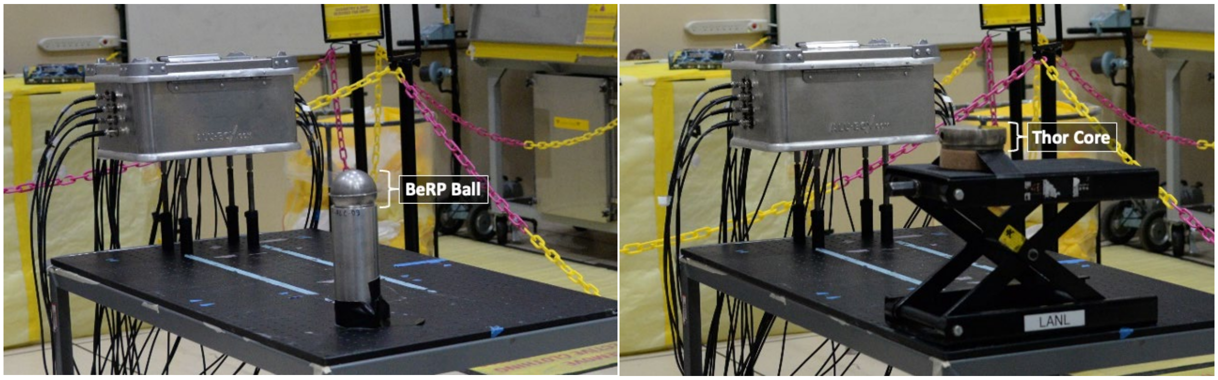

Figure 3: Experimental setup for the BeRP Ball experiment (left) and Thor core (right).

RESULTS AND DISCUSSION

Extracting the single gamma ray events from the $CeBr_3$ in the BeRP Ball measurement results in the first plot shown in Figure 4. This spectrum features prominent gamma rays at 375 keV, 414 keV, and 646 keV, which strong indicators that the decay of ^{239}Pu is being observed. Although the gamma singles spectrum helps with isotope identification by indicating the most intense emissions observed, it also serves the dual purpose of providing insight towards what energy gates should be used during the gamma-ray imaging process in cases where the user may not know ahead of time what gamma rays are of interest.

Recording all the gamma ray OGS-$CeBr_3$ coincident events results in the second plot shown in Figure 4. This gamma coincidence spectrum is the distribution of all imageable gamma rays observed during a measurement. There is a notable difference in how well the peaks are visible in the coincidence spectrum and this is largely due to the organic scintillator having less energy resolution than the $CeBr_3$. Figure 5 has the same two plots found in Figure 4 but correspond to the Thor core data. Both coincident spectra from the sources have a particular energy gate highlighted to show the gamma ray energy regions used to create the images shown later in the results of this work. The BeRP measurement was gated using 646 ± 36 keV, corresponding to the 646 keV emission noted below, while the Thor measurement was gated using 350 – 425 keV, which encompasses the 375 and 414 keV emission.

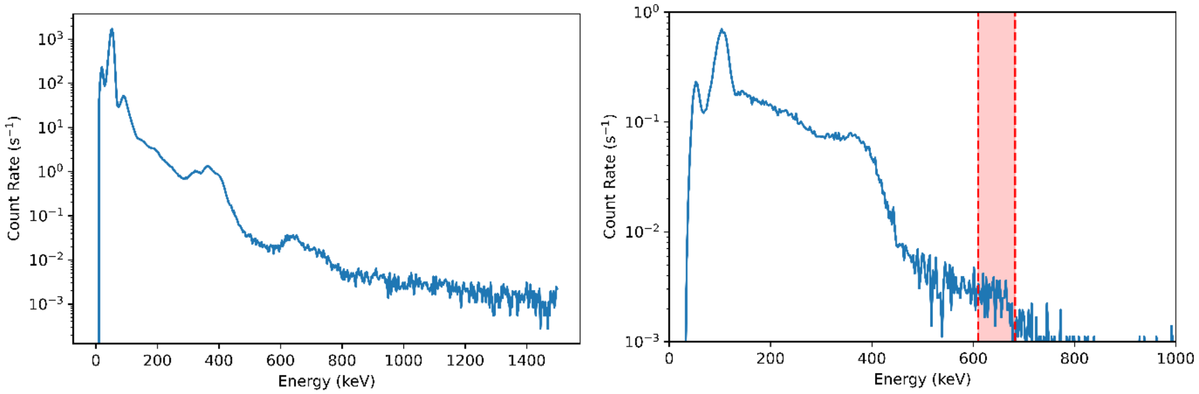

Figure 4: BeRP ball $CeBr_3$ gamma single counts spectrum (left) and gamma ray OGS – $CeBr_3$ coincidence spectrum (right) used for imaging. An energy gate of 646 ± 36 keV is noted on the coincident spectrum to indicate the region imaged.

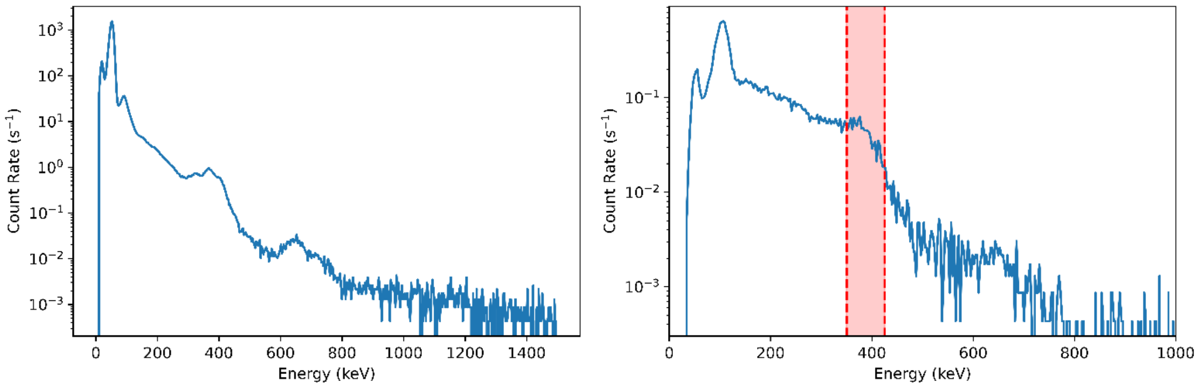

Figure 5: Thor core $CeBr_3$ gamma single counts spectrum (left) and gamma ray OGS – $CeBr_3$ coincidence spectrum (right) used for imaging. An energy gate of 350 – 425 keV is noted on the coincident spectrum to indicate the region imaged.

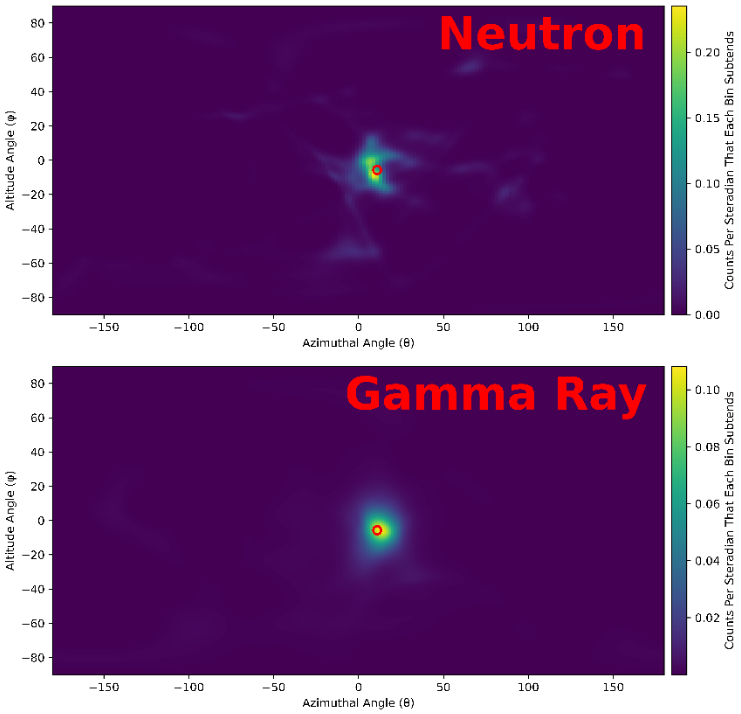

Figure 6: Converged images of BeRP Ball from neutrons (top) and gamma rays with energy gating of 646 ± 36 keV (bottom).

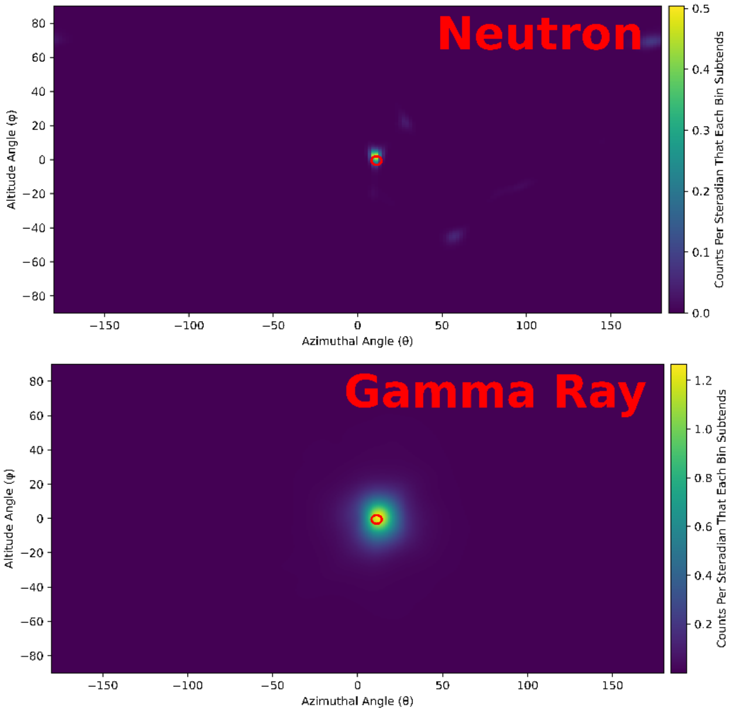

Figure 7: Converged images of Thor core piece from neutrons (top) and gamma rays with energy gating of 350 – 425 keV (bottom).

Figure 6 shows the resulting fast-neutron and energy-gated gamma-ray images from the BeRP ball measurement. The location of the source in the angular space is shown as a red outline in this work. All images shown in this work are the results of applying the LM-MLEM converging algorithm on the SBP images. We also note that in this work, neutron images were generated using a light output threshold of 75 keVee per event. This corresponds to imageable neutrons needing to surpass this threshold twice, once for each scatter. Gamma-ray images were generated using a 25 keVee threshold, resulting in an effective minimum imageable gamma ray energy of 50 keVee. Overall, we note that both the neutron and gamma images for the BeRP ball had good agreement with the true source location.

Figure 7 shows the converged neutron and gamma-ray images for the Thor core piece. We note that despite having fewer imageable neutron double-scatter events than the BeRP ball experiment, the converged source location using neutron data was still accurate. Both images agreed with the Thor core location during the experiment. Finally, we note that in cases where energy is not necessary (i.e., when simply confirming presence of a gamma source in general) gamma-ray images can typically be produced with data from very short measurement times with reasonable accuracy.

CONCLUSIONS

We presented gamma-ray and fast-neutron images created with an organic glass-based system when measuring kg quantities of plutonium metal for the first time. These sources had masses of 4+ kg and ^{239}Pu compositions of 90+ wt%. Plutonium material management continues to be a prominent goal for international safeguards due its potential for nuclear proliferation. Successful dual-particle imaging of plutonium metal demonstrates that the technology in this work can be utilized in this sector of nuclear safeguards to great effect. The ability to localize plutonium using either the fast-neutron or gamma-ray signatures is impactful because a wide variety of scenarios can be addressed. Viable imaging can be achieved since cases where one signature is hidden or suppressed means that the other can be used instead. Localization, while important on its own, is further strengthened when coupled with spectroscopy capabilities, another useful aspect of this imaging system. Future work on characterizing the system design and improving the electronics can help push this work towards a more field-deployable level. This technology serves to further promote nonproliferation and arms control, aligning with INMM's overall goal of material management.

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REFERENCES

[1]. U.S. NRC, Material Control and Accounting of Special Nuclear Material, 10 C.F.R. 74.4 (2015).

[2]. Hamel, M. C., Polack, J. K., Poitrasson-Rivière, A., Clarke, S. D., & Pozzi, S. A. (2017). Localization and spectral isolation of special nuclear material using stochastic image reconstruction. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 841, 24-33.

[3]. Goldsmith, J. E., Gerling, M. D., & Brennan, J. S. (2016). A compact neutron scatter camera for field deployment. Review of Scientific Instruments, 87(8), 083307.

[4]. W. M. Steinberger, M. L. Ruch, N. Giha, et al., "Imaging special nuclear material using a handheld dual particle imager," *Scientific Reports*, vol. 10, no. 1, Feb. 2020. doi: 10.1038/ s41598-020-58857-z.

[5]. Carlson, J. S., Marleau, P., Zarkesh, R. A., Feng, P. L. "Taking Advantage of Disorder: Small-Molecule Organic Glasses for Radiation Detection and Particle Discrimination" J. Am. Chem. Soc. 2017, 139, 9261–9626.

[6]. Carlson, J. S., and Feng, P. L. "Melt-cast organic glasses as high-efficiency fast neutron scintillators." *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 832 (2016): 152-157.

[7]. Giha, N. P., et al. "Organic glass scintillator bars with dual-ended readout." *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment* 1014 (2021): 165676.

[8]. Steinberger, W. M., et al. "Optimizing the Position of Inorganic Scintillators in a Handheld Dual Particle Imager." 2020 IEEE Nuclear Science Symposium and Medical Imaging Conference (NSS/MIC). IEEE, 2020.

[9]. Lopez, R., Steinberger, W. M., Giha, N., Marleau, P., Clarke, S. D., & Pozzi, S. A. (2022). Neutron and gamma imaging using an organic glass scintillator handheld dual particle imager. *Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment*, 1042, 167407.