MCNP Characterization of ^{239}Pu with PGAA for Minimum Detection Limits

C. Egozi[1], W.S. Charlton[2], Z. Beauvais[2], S. Landsberger[2]

[1]Los Alamos National Laboratory, P.O. Box 1663, 87545, Los Alamos New Mexico
[2]Nuclear Engineering Teaching Lab, University of Texas at Austin,
Pickle Research Campus, R-9000, Austin, Texas, USA 78712
Email: cegozi@lanl.gov

Abstract

As safeguards verification becomes a more pressing concern, with the increase in global special nuclear material production, enhanced nondestructive methods of detection are necessary. Prompt Gamma Activation Analysis (PGAA) is an effective process to generate, detect and measure characteristic gamma rays at high energies. These gamma rays are unlikely to be attenuated through glove boxes and therefore radionuclides would not need to be removed from the process line for quantification. While intense prompt gamma rays can be readily detected for PGAA using a high purity germanium detector, low intensity emissions may be lost in high background radiation due to the Compton continuum in the collected spectrum. When combined with Compton suppression to reduce the continuum, low intensity prompt gamma rays can be more easily identified. PGAA has only recently been applied for ^{239}Pu characterization experimentally at the University of Texas at Austin, but has not as yet been modelled in MCNP. This lack of characterization extends to the MCNP databases, as there are no tabulated prompt gamma rays for ^{239}Pu that can be referenced when running simulations. For verification purposes, an MCNP model of the Compton suppression system at the Nuclear Engineering Teaching Lab was created. The experimental system was a combined Compton suppression and PGAA setup. Therefore, the MCNP model was first created to model the decay gammas of ^{239}Pu with Compton suppression. Once this simulation matched the experimental results, the previously identified PGAA gammas were input instead of the ^{239}Pu decay gamma rays. With these results, it was possible to determine the minimum time required to detect the characteristic PGAA gamma rays for a 1.81E6 Bq 0.789 mg ^{239}Pu foil electrodeposited on nickel. The data produced from this simulation could be utilized to compare to experimental glovebox verifications to determine ^{239}Pu quantities in nuclear facilities. PGAA can also be used to detect very low levels of ^{239}Pu for on-site verification to determine whether the radionuclide is being produced.

Keywords

Prompt Gamma Activation Analysis, ^{239}Pu, MCNP, Compton suppression

1. Introduction

PGAA is a method applied to characterize radionuclides based on the gamma rays emitted in an (n,γ) reaction. The determination of these gamma rays is of particular importance in circumstances where decay gammas would normally be attenuated. This is due to the production of much higher energy prompt gamma rays that could still be detected through attenuating materials. Experimental PGAA facilities have produced spectra with high Compton scattering and noted the value of combining the PGAA detector with a NaI annulus for Compton suppression [1]. The prompt gamma rays of many isotopes, including ^{58}Ni(n,γ)^{59}Ni, have been well characterized with Compton suppression gamma spectrometry [2]. The IAEA has a database of prompt gamma rays for isotopes up to uranium [3], and others have performed independent characterizations of prompt gamma rays for 70 isotopes [4]. MCNP has been utilized for geometry optimization of an HPGe and a suppression annulus. It was determined through simulation that a coaxial system would be ideal for PGAA measurements [5]. Modeling of the ^{1}H(n,γ)^{2}H reaction with the use of Gaussian energy broadening and directional biasing has been performed on the Budapest PGAA facility for validation of the modeling [6]. MCNP simulation has been used to determine ways to optimize experimental systems, as well as for comparison to experimental results.

2. Experimental Procedure

The PGAA facility at the Nuclear Engineering Teaching Laboratory (NETL) at the University of Texas at Austin consists of a cold neutron beam incident on a sample at a 45^{o} angle and using an HPGe detector with NaI annulus to record Compton suppressed gamma spectra with an energy range from ~100 keV up to around 8-10 MeV [7]. Using the PGAA system, a ^{239}Pu sample consisting of 1.81E6 Bq of ^{239}Pu electrodeposited on a nickel foil was measured once using a passive measurement of the decay gamma rays from ^{239}Pu with the PGAA system neutron beam turned off and the HPGe detector operating in Compton suppression mode. The sample was then re-measured for 4 hours for an active measurement using the neutron beam turned on with the reactor at 900 kW. Results from the passive measurement are shown in Fig. 2 and results from the active measurement are shown in Fig. 5. The spectrum for the active measurement was analyzed using Peak Easy to determine net peak areas for key lines of interest and those are given in Table 1.

3. MCNP Simulation

The PGAA system modeled in MCNP was designed to replicate the experimental facility at the NETL as closely as possible including the sample, neutron beam incident on the sample, an HPGe detector, NaI annulus for Compton suppression, lead shielding, and Li carbonate neutron shielding [8]. The geometry in the MCNP model is shown in Figure 1 .

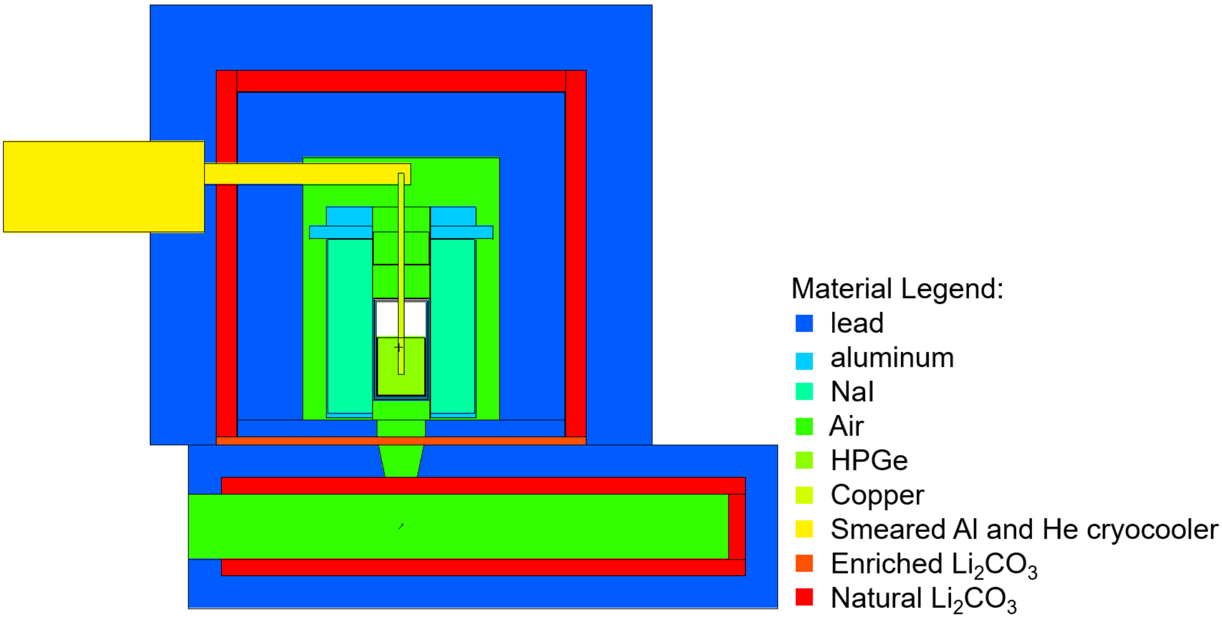

Fig. 1 Compton suppressed HPGe detector geometry setup for MCNP

A 1.81E6 Bq ^{239}Pu foil electrodeposited on nickel, with radius of 0.49149 cm was included in this simulation and placed 19.58 cm from the face of the HPGe detector. Compton suppression was simulated using an F8 Pulse Height Light (PHL) tally. This tally was specified to simulate the HPGe and NaI annulus operating in anti-coincidence mode. The spectrum produced was compared to the experimental result in Figure 2, and it was determined that the simulated results agreed well with the experimental results for gamma ray energies above about 80 keV. Gamma rays below 80 keV are generally not of interest in a PGAA measurement so no effort was expended to resolve the disagreement in that area. This verified that the detector geometry and materials in the MCNP model were sufficiently accurate to reproduce experimental results.

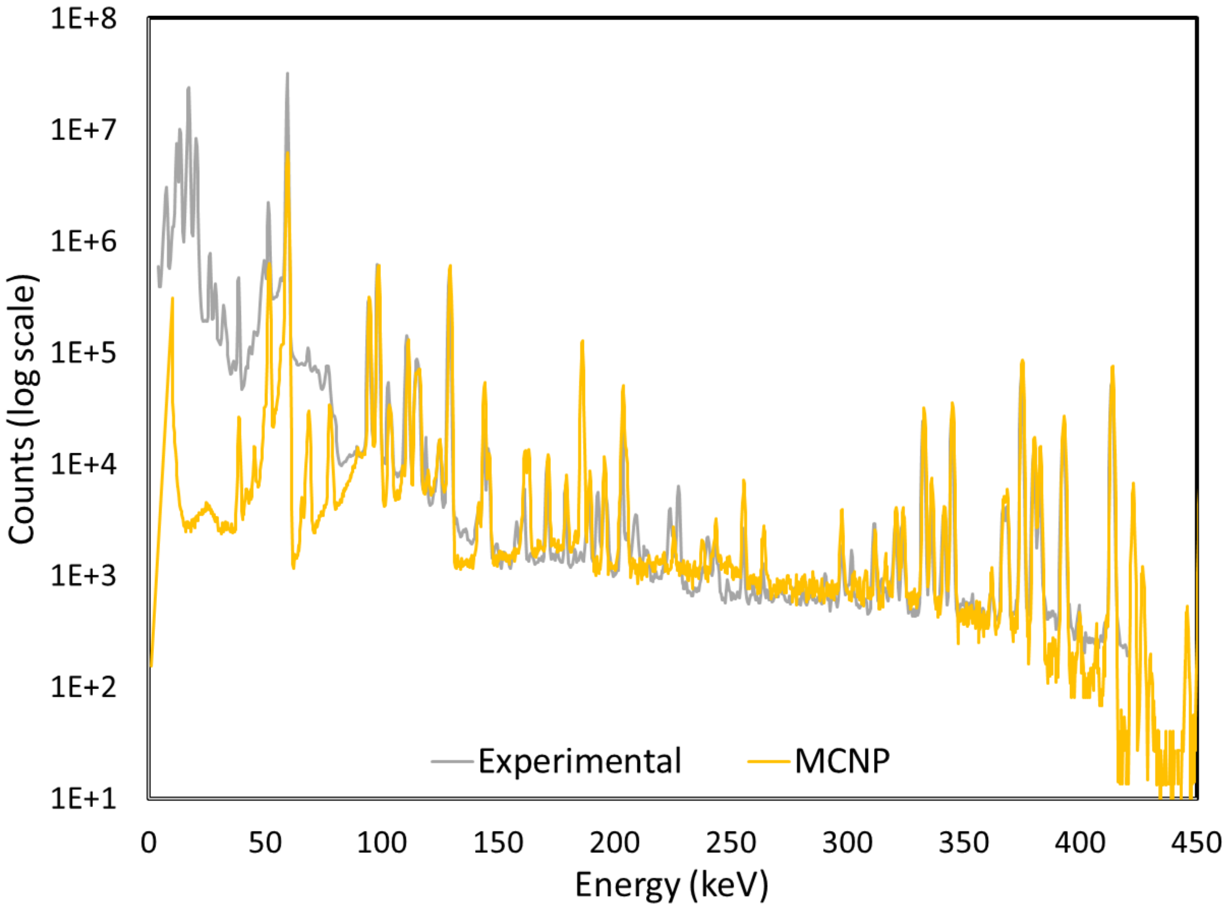

Fig. 2 Simulated (bottom) and experimental (top) Compton suppressed ^{239}Pu spectra

Once the geometry was verified, the MCNP model was adapted to perform an active PGAA measurement with Compton suppression which included adding the neutron beam to the simulation. However, the MCNP database does not contain PGAA lines for ^{239}Pu, so this simulation was performed in two distinct components. The first component was a simulation of an active, 4-hour, PGAA measurement for the ^{239}Pu electrodeposited on nickel where the PGAA lines were produced by neutron (n,γ) interactions only by the Ni. The nickel was comprised of ^{58}Ni and ^{60}Ni in their natural abundance ratios. The resultant spectrum for Ni PGAA is shown in Figure 3, and arrows indicate the high intensity gammas and their intensities, all of which are found in the IAEA database [3]. A second component of the simulation consisted of simulating PGAA lines born uniformly in the Pu sample and transported to the detector to record a spectrum. The ^{239}Pu prompt gamma yields were calculated relative to the 2554 keV ^{58}Ni prompt gamma. A spectrum for the expected PGAA lines from ^{239}Pu was then produced for a 4-hour irradiation. The two components were then summed using linear superposition to produce the expected spectrum from a 4 hour active PGAA measurement of Pu electrodeposited on Ni. With the resulting combined spectrum shown in Fig. 5. The input for ^{239}Pu was somewhat different, as the prompt gamma rays had to be provided manually. Therefore, the 7 gamma rays found experimentally, as well as their yields, were added to the SI and SP cards respectively. This setup had no neutron source, since the prompt gammas were already defined. The spectrum, Figure 4, shows the expected output of the 7 gamma rays.

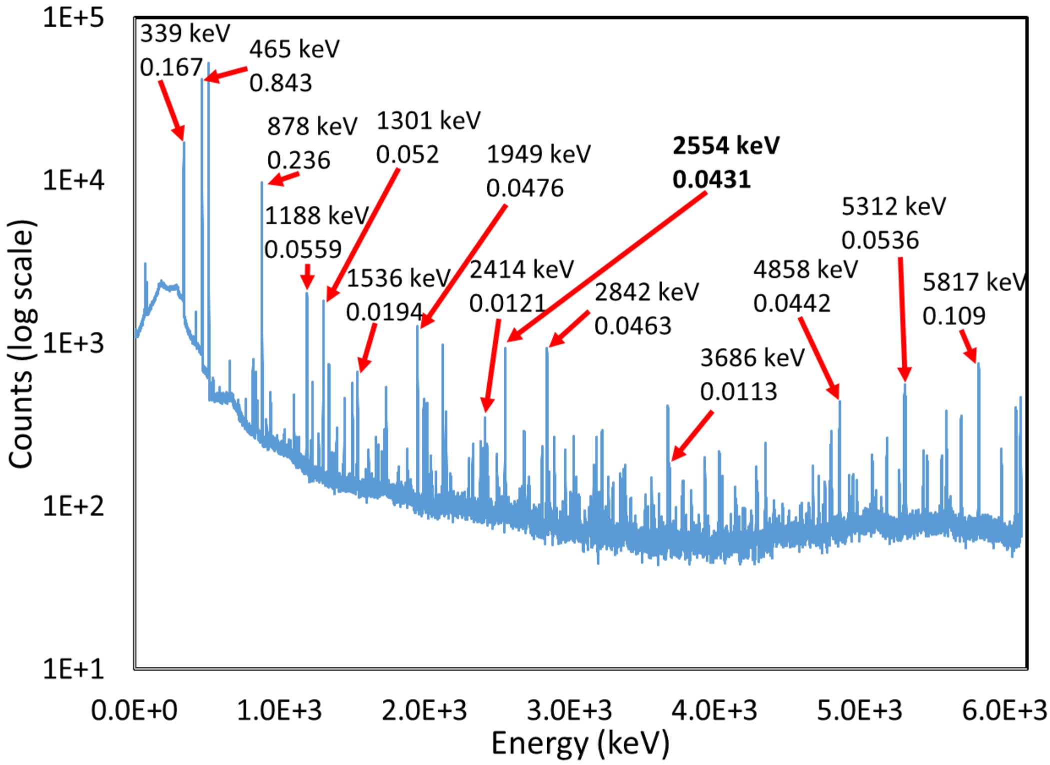

Fig. 3 Nickel PGAA spectrum from MCNP

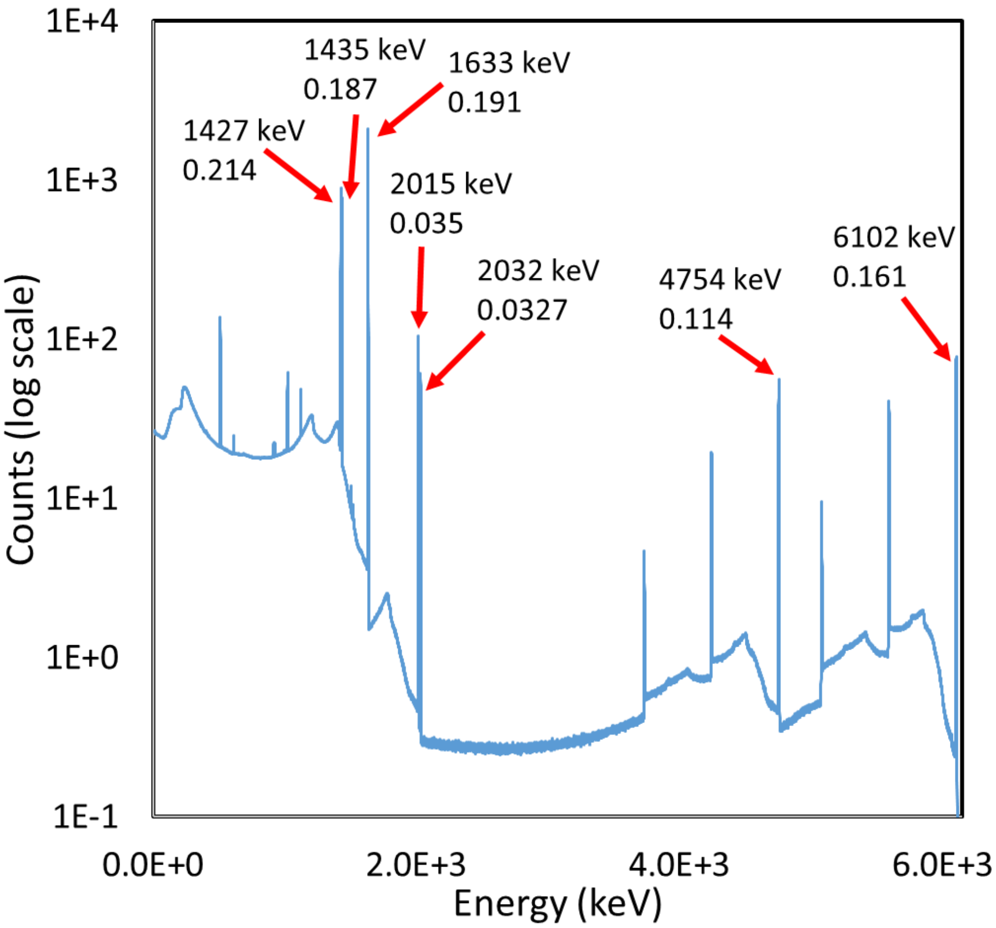

Fig. 4 ^{239}Pu PGAA spectrum from MCNP with experimental input

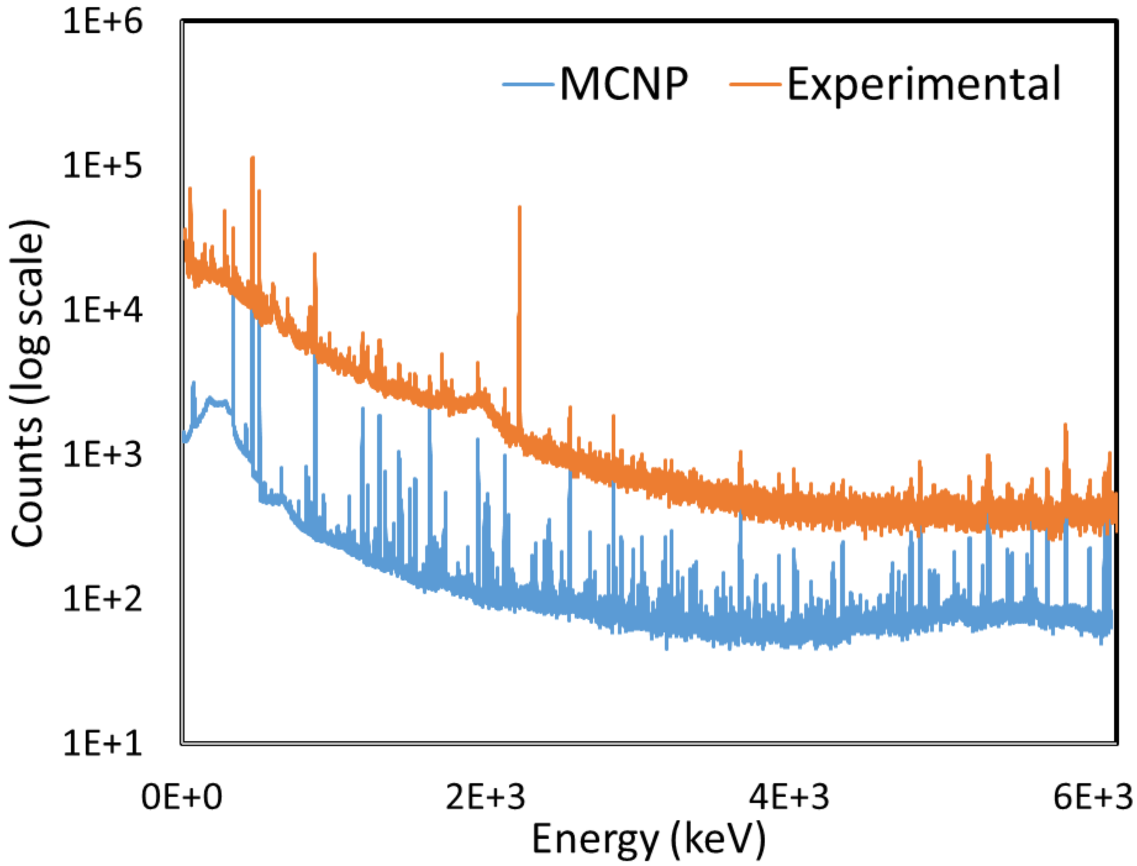

Fig. 5 Combined ^{239}Pu and nickel PGAA MCNP spectra compared to experimentally measured spectra

4. Results and Discussion

The MCNP net counts for the high intensity ^{58}Ni and ^{239}Pu gammas were compared to the experimental net counts from Figure 5 to determine whether the simulation was comparable to experiment. The resulting values, as well as their percent errors, are displayed in Table 1.

Table 1 ^{58}Ni and ^{239}Pu experimental and MCNP comparison of high intensity gamma rays

Gamma Ray (keV)	Isotope	Experimental net area	MCNP net area	% Difference
339	^{58}Ni	106918 ± 574	132464 ± 433	-23.9
465	^{58}Ni	490126 ± 825	366529 ± 627	25.2
1427	^{239}Pu	9446 ± 250	9623 ± 127	-1.87
1435	^{239}Pu	7998 ± 255	8042 ± 123	-0.55
1633	^{239}Pu	54417 ± 316	23964 ± 173	56.0
1949	^{58}Ni	15040 ± 229	13520 ± 135	10.1
1992	^{58}Ni	4343 ± 203	3900 ± 92	10.2
2015	^{239}Pu	5039 ± 198	5272 ± 101	-4.62

2032	^{239}Pu	1322 ± 192	1368 ± 79	-3.48
2554	^{58}Ni	10845 ± 190	11042 ± 125	-1.82
2842	^{58}Ni	10685 ± 183	11550 ± 125	-8.10
3025	^{58}Ni	3128 ± 146	2961 ± 83	5.34
4754	^{239}Pu	2371 ± 149	2211 ± 95	6.75
6102	^{239}Pu	9676 ± 162	9331 ± 124	3.57

The ^{239}Pu gamma rays of interest, as well as the high energy ^{58}Ni gammas, are below 11 percent error relative to the experiment. This holds true for all the gamma rays aside from the 1633 keV. This is due to the experimental setup containing a significant amount of PFA. The fluorine in the PFA emits a 1633 keV gamma ray, so there is little confidence in the ^{239}Pu gamma ray at that energy. The background continuum does not match the experimental spectrum due to the high energy lead prompt gamma rays at 6738 keV and 7368 keV that are not in the MCNP database, and therefore not in the simulated spectrum. This likely contributes to the high error in the low energy ^{58}Ni gamma rays. In addition, the hydrogen presence experimentally at 2223 keV has a much higher content than the simulated hydrogen. Despite these considerations, within the range of 1000-6200 keV, the net areas of all ^{58}Ni and ^{239}Pu are determined accurately.

The counting time was then varied to determine the shortest time in which all net area uncertainties for the ^{239}Pu peaks remained below 10%. The geometry, flux (900 kW), and source dimensions remained unchanged. This optimal time was found to be 5000 seconds, with the net areas and uncertainties shown in Table 2 below.

Table 2 ^{239}Pu MCNP high intensity gamma rays for 5000 seconds counting time

Gamma Ray (keV)	Net area	% uncertainty
1427	3360 ± 75	2.23
1435	2792 ± 73	2.61
1633	8319 ± 102	1.23
2015	1837 ± 59	3.21
2032	474 ± 46	9.70
4754	770 ± 56	7.27
6102	3255 ± 73	2.24

This is therefore the minimum counting time required experimentally to ensure net area uncertainties remain below 10%.

5. Conclusions

MCNP can be applied with Prompt Gamma Activation Analysis to determine ^{239}Pu of a known mass electrodeposited on nickel. This method can be used to evaluate the minimum detectable activity of ^{239}Pu in a specific system before acquiring a source and performing experimental measurements. An approximate counting time can be determined theoretically in order to limit excess experimental irradiation of the source. The attenuation of these gamma rays through nuclear material storage containers can be determined, and due to their high energy, they are far less likely to be attenuated. The use of MCNP with PGAA in a nuclear facility can be extremely useful, where it is not always practical to eliminate containment and perform experimental measurements to evaluate ^{239}Pu content. This is a preliminary study to determine viability of the detection system, further work is needed to characterize the neutron source, moderator, and shielding for implementation in a safeguards context.

Acknowledgements

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