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Ability of non-destructive assay techniques to identify sophisticated material partial defects

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ABSTRACT

This study explores the ability of non-destructive assay techniques to detect a partial material defect in which 100 g of plutonium are diverted from the center of a 1000 g can of PuO_2 powder. Four safeguards measurements techniques: neutron multiplicity counting, calorimetry, gravimetry, and gamma ray spectroscopy are used in an attempt to detect the defect. Several materials are added to the partial defect PuO_2 can to replicate signatures of the diverted material. ^{252}Cf is used to compensate for the doubles neutron counts, ^{241}Am is used to compensate for the decay heat, and aluminum is used to compensate for the weight. Although, the doubles and triples difference before and after diversion are statistically indistinguishable with the AWCC in fast and thermal mode, the difference in the singles counts are statistically detectable in both modes. The relatively short half-life of ^{252}Cf leads to a decrease (three sigma uncertainty) in the doubles neutron counts after 161 days. Combining this with the precise quantity of ^{241}Am needed (10.7 g) to mimic the heat signature and the extreme precision in ^{252}Cf mass needed to defeat neutron multiplicity measurements gives reassurance in the International Atomic Energy Agency's ability to detect partial material defects.

1. Introduction

The International Atomic Energy Agency (IAEA) is responsible for ensuring nuclear technology is not used for weapons purposes by non-nuclear weapons states that are signatories to the Nuclear Nonproliferation Treaty [1]. The IAEA is not an enforcement organization but rather a verification organization that if need will report violations to the United Nations Security Council. In order to accomplish this task, the IAEA utilizes a vast array of techniques to implement nuclear safeguards across the world. These safeguards are intended to detect and deter diversions of nuclear materials by member states. Ensuring these techniques are effective against possible diversions is important.

This study will explore a specific diversion scenario in which a fraction of PuO_2 powder is removed and replaced with other materials that mimic commonly measured safeguards signatures. Plutonium is considered attractive for weapons purposes and diversion of such material possess a significant threat. Rather than explore a diversion of a significant amount of plutonium at once, this study will focus on a possible technique to divert a smaller amount of plutonium that may be repeated over time. A sphere of PuO_2 with a mass of 100 g of plutonium is taken from the center of a right circular cylindrical can which contained 1000 g of plutonium. A mixture of ^{252}Cf, ^{241}Am, and natural aluminum was added to the spherical hole in the center of the PuO_2 can to mimic the neutron doubles counts, decay heat generation, and mass of the can.

Four safeguards detection methods will be used to assess the effectiveness at detecting the partial defect. Monte-Carlo N Particle (MCNP) software simulation of passive neutron multiplicity measurements of an Active Well Coincidence Counter (AWCC) is used to compare singles, doubles, and triples counts of the samples. Calorimetry is used to compare the heat generation of the samples. Mass calculations are used to compare the weights of the samples. MCNP simulation of the gamma ray spectrum measured by a High-Purity Germanium (HPGe) detector is used to compare the gamma ray spectra of the samples.

2. Materials and methods

2.1. Neutron multiplicity counting utilizing the AWCC

2.1.1. Neutron multiplicity introduction

Neutron multiplicity counting is a commonly utilized non-

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destructive assay technique used to estimate the mass of fissile material, such as uranium or plutonium. When a fission event occurs, neutrons are emitted in multiples ranging from 0 to approximately 8. Since neutrons are not easily attenuated by high Z materials, the neutron multiplicity measurements can be used to approximate the mass of the fissioning material [2].

Neutron multiplicity counting is done by both active and passive interrogation. In the case of active interrogation, a neutron source is used to induce fission in the sample material. This technique is routinely used for measuring ^{235}U mass. In the case of passive interrogation, an interrogation neutron source isn't used, but rather the sample material spontaneously fissions. This technique is routinely used for measuring plutonium mass. Since the material in this study is PuO_2, passive interrogation will be used.

In used fuel, ^{240}Pu often has the largest abundance relative to the other even plutonium isotopes. Although the neutrons produced in a sample of plutonium are produced primarily by ^{238}Pu, ^{240}Pu, and ^{242}Pu, defining a^{240}Pu effective mass is more convenient. This ^{240}Pu effective mass represents the mass of ^{240}Pu that will give an equal signal to that of the plutonium sample [2]. Equation (1) shows the relationship between ^{240}Pu effective mass ($m_{240\ eff}$) to the mass of Pu^{238} (m_{238}), ^{240}Pu (m_{240}), and ^{242}Pu (m_{242}) in the sample.

$$m_{240\ eff} = 2.52m_{238} + m_{240} + 1.68m_{242} \tag{1}$$

This mass can be calculated from the singles, doubles, and triples counts resulting from a neutron multiplicity measurement. Equation (2) shows the relationship between the mass (m), the doubles counts (D), the efficiency of the detector (ε), the doubles gate fraction (f_d), the leakage multiplication (M_L), the first factorial induced fission multiplicity (ν_{i1}), the second factorial induced fission multiplicity (ν_{i2}), the singles counts (S), the spontaneous fission yield (F_0), and the second factorial spontaneous fission multiplicity (ν_{s2}) [3].

$$m = \frac{\frac{2D}{\varepsilon f_d} - \frac{M_L(M_L-1)\nu_{i2}S}{\nu_{i1}-1}}{\varepsilon F_0 M_L^2 \nu_{s2}} \tag{2}$$

The gate fraction can be calculated using the pre-delay gate width (P), the gate width (G), and the neutron die away time (τ) as shown in Equation (3).

$$f_d = e^{-\frac{P}{\tau}}\left(1 - e^{-\frac{G}{\tau}}\right) \tag{3}$$

Table 1 shows the spontaneous fission yields of ^{240}Pu and ^{252}Cf. Table 2 shows the prompt fission multiplicities of ^{239}Pu, ^{240}Pu, and ^{252}Cf. For plutonium measurements, the induced fission multiplicities of ^{239}Pu and the spontaneous fission multiplicity of ^{240}Pu are used.

In this study, MCNP is used to simulate neutron multiplicity measurements of the sample [4]. In plutonium dioxide, both spontaneous fissions and (alpha,n) reactions must be considered. The (alpha,n) reactions will mostly contribute to the singles counts but also may induce fission in the plutonium (primarily ^{239}Pu)

Table 1
Spontaneous fission yield [2].

Isotope	Fission Yield [fissions/s·g]
^{240}Pu	4.72E+02
^{252}Cf	6.23E+11

Table 2
Prompt fission multiplicities [2].

Isotope	ν_{i1}	ν_{i2}	ν_{s2}
^{239}Pu	2.88	6.773	-
^{240}Pu	-	-	3.794
^{252}Cf	-	-	11.962

Table 3
(alpha,n) yield in dioxide [2].

Isotope	(α,n) yield in oxide [n/s·g]
^{238}Pu	1.34E+04
^{239}Pu	3.81E+01
^{240}Pu	1.41E+02
^{241}Pu	1.30E+00
^{242}Pu	2.00E+00

which result in singles, doubles and triples counts. Sources 4C was used to calculate the neutron spectrum from the alpha-n reactions in the plutonium dioxide [5]. Table 3 shows the (alpha,n) yield in dioxide of the plutonium isotopes.

Considering this study uses MCNP6 simulations, approximating the expected experimental uncertainties of the singles, doubles, and triples counts is necessary in order to understand how effective these measurements might be. These uncertainties are estimated using a technique developed by Dr. Stephen Croft [6]. Equation (4) shows the relationship between the singles count rate uncertainty (σ_S) and the singles count rate (S_r), the measurement time (t), the doubles count rate (D_r), and the doubles gate fraction (f_d).

$$\sigma_S = \sqrt{\frac{S_r}{t} + \frac{2D_r}{f_d t}} \tag{4}$$

Equation (5) shows the relationship between the doubles count rate uncertainty (σ_D) and the gate width (G), the doubles gate fraction (f_d), the neutron die away time (τ), the singles count rate (S_r), the doubles count rate (D_r), and the measurement time (t).

$$\sigma_D = \sqrt{\left(1 + 8\left(1 - \frac{1 - e^{-\frac{G}{\tau}}}{\frac{G}{\tau}}\right)\left(\frac{D_r}{S_r f_d}\right)\right)\frac{D_r + 2S_r^2 G}{t}} \tag{5}$$

Equation (6) shows the relationship between the triples count rate uncertainty (σ_T) and the gate width (G), the doubles gate fraction (f_d), the neutron die away time (τ), the singles count rate (S_r), the doubles count rate (D_r), the triples count rate (T_r), and the measurement time (t).

$$\sigma_T = \sqrt{\left(1 + 10\left(1 - \frac{1 - e^{-\frac{G}{\tau}}}{\frac{G}{\tau}}\right)\left(\frac{D}{f_d S}\right)\right)\left(\frac{1}{t}\right)\left(T + 2SG\left(\left(1 + \frac{D}{f_d}\left(1 - \frac{1 - e^{-\frac{G}{\tau}}}{\frac{G}{\tau}}\right)\right) + \frac{S^2 G}{2}\right)\right)} \tag{6}$$

Although the neutron die away time and efficiency is often known for a given detector configuration, they can also be calculated from the MCNP output. By utilizing two gate widths (one equal to the gate width mentioned previously (G) and the other equal to half that gate width), the neutron die away time can be calculated from the resulting normalized doubles counts from each gate. Equation (7) shows the relationship between the neutron die away time (τ) and the shorter gate width (G_1), the normalized doubles counts from the shorter gate (D_1), and the normalized doubles counts from the longer gate (D_2).

$$\tau = \frac{G_1}{\ln\left(\frac{D_2}{D_1} - 1\right)} \tag{7}$$

Equation (8) shows the relationship between the efficiency (ε) and the net multiplication (M_{net}) and the normalized singles counts (S_{norm}). Both the M_{net} and S_{norm} are obtained from the MCNP output.

$$\varepsilon = \frac{S_{norm}}{M_{net}} \tag{8}$$

2.1.2. The AWCC

The AWCC is a commonly utilized neutron multiplicity counter that the IAEA uses to make safeguards measurements on uranium and plutonium samples. This detector has 42 ^{3}He tubes embedded in high density polyethylene. The sample cavity is 20 cm in diameter and can be adjusted 23–35 cm in height. When measuring uranium, AmLi neutron sources are used to induce fission in the sample. However, when measuring plutonium, a neutron source is not needed as the even number plutonium isotopes spontaneously fission at a sufficient rate [7]. The AWCC can be operated in thermal mode or fast mode. In fast mode, there is a cadmium sleeve within the detector well that will remove thermal neutrons thus preventing them from scattering back into the plutonium and inducing fission. In thermal mode, the cadmium sleeve is not present [8]. This study models the AWCC in both fast and thermal mode. The initial simulations and analysis are done utilizing the fast mode. Fast mode was chosen as it has a lower sensitivity than thermal mode thus increasing the difficulty in detection. The results from the thermal mode are then compared to those from the fast mode.

2.1.3. Activity decay calculations

Considering ^{252}Cf has a half-life of only 2.645 years [9], the decay of the sample must be considered when analyzing the doubles counts and associated uncertainty of multiple measurements taken over long periods of time. The uncertainty represents a threshold below which the difference in doubles counts is statistically detectable. Equation (9) shows the relationship between the number of atoms at some time (N) to the initial number (N_0), the time (t), and the decay constant (λ) [10].

$$N = N_0 e^{-\lambda t} \tag{9}$$

2.2. Calorimetry measurement

Since nuclear decay releases energy in the form of heat, measuring the heat produced by a material is a viable method to verify the material is as declared. Each radionuclide releases a particular amount of heat per mass, which allows for total mass determination if the radionuclide composition is known. This heat per mass can be calculated by combining the activity of the isotope in decays per time to the amount of energy released with each decay. In the case of alpha decay, the kinetic energy of the alpha particle makes up the vast majority of the energy released and will deposit its energy rapidly. However in the case of beta decay, kinetic energy of the electron is on average one third the energy released by the decay. The remaining energy escapes via a neutrino. The IAEA uses calorimeters to measure the heat produced from a sample [11]. The sample is placed within the body of a calorimeter. The body is surrounded by walls with some known thermal resistance. As the sample produces heat, this heat is transferred through the cavity and walls into the environment surrounding the calorimeter. The change in temperature of the body of the cavity is related to the heat produced in the sample. These measurements can be used on a variety of materials including samples of plutonium and a variety of geometries ranging from small to large sample. The calorimeter that will be considered here is the Mound Transportable Calorimeter. This calorimeter can measure plutonium masses from 200 g to 4.4 kg and has accuracy of 0.3%. It can measures samples up to 13 cm in diameter and 20 cm in height [2]. The sample being studied is within the mass and geometry range of the Mound Transportable Calorimeter.

Utilizing the data found in the Korean Atomic Energy Research Institute's Table of Nuclides [8], the heat generation of ^{241}Am is calculated. Table 4 shows the heat generation of ^{241}Am and the plutonium isotopes.

2.3. Mass measurement

Mass measurements are another important aspect of safeguards verification. Since most nuclear material is in sealed containers preventing the use of sight to determine the material is present, weighing containers and comparing the weights with verified records is a method in which to verify nuclear material has not been diverted from the container. Common commercial scales have relatively high precision as low as 0.001 g.

2.4. Gamma ray spectroscopy measurement

Gamma ray spectroscopy measurements are a commonly accepted non-destructive assay technique useful for IAEA safeguards applications. As radionuclides decay, the nucleus emit energy in the form of photons (gamma rays). The energy of these gamma ray emissions are specific to the decaying radionuclide and can be measured and used to identify the radionuclides presence and their concentrations. It is possible to measure the mass of a sample using gamma ray measurements, however, assumptions regarding the sample geometry and composition must be made [2]. Measured gamma ray spectra can be influenced from non-gamma ray radiation. Charged particles, such as alpha and beta particles can liberate electrons from nuclides in the sample and thus create characteristic x-rays. Beta particles also create Bremsstrahlung which can have a noticeable effect on the measure gamma ray spectrum. Both bremsstrahlung and the characteristic x-rays are included in the spectrum.

Table 4
Decay heat [15].

Isotope	Decay Heat [W/kg]
^{238}Pu	557.43*
^{239}Pu	1.88*
^{240}Pu	6.94*
^{241}Pu	3.22*
^{242}Pu	0.11*
^{241}Am	111.54

HPGe detectors are one of the most useful detector types for assaying plutonium materials. HPGe detectors have high energy resolution, which is important when attempting to analyze peaks in close energy proximity [12]. In terms of nuclear safeguards, resolving individual peaks is crucial to accurately identifying radionuclides and determining their relative concentrations. When measuring materials where multiple radionuclides are present, peaks may have similar energies and require high energy resolution to distinguish between them.

MCNP can be used to simulate detector responses from a variety of gamma ray sources. The gamma ray emission spectrum of the source can be calculated by RadSrc. RadSrc allows for the input of the isotopic concentrations of a sample and the amount of time the sample has decayed and will output the spectrum of the resulting decayed isotopics. The program can display the output spectrum in a format that can be directly used in a source definition of MCNP. RadSrc also calculates the source strength of the gamma ray emissions as well as the expected bremsstrahlung and characteristic x-rays [13].

2.5. Diversion scenario

The diversion scenario analyzed in this study challenges the four safeguards technologies mentioned above: neutron multiplicity counting, calorimetry, gravimetry, and gamma ray spectroscopy. The diversion material is plutonium dioxide (PuO_2). In order to address the neutron multiplicity counting, the doubles counts of the diverted material is reproduced by the addition of ^{252}Cf. For calorimetry, the heat generation of the diverted material is reproduced by the addition of ^{241}Am. The weight of the diverted material is made up by the addition of aluminum. The PuO_2 which is diverted is removed from the center of the sample, thus minimizing the effects in the gamma ray spectrum measured from the exterior of the can.

Neutron multiplicity simulations using the AWCC in MCNP before and after diversion are compared. The mass of ^{252}Cf was increased until the doubles counts from both simulations matched. The total heat generation within the diverted sample is calculated and the mass of ^{241}Am metal that will produce an equal amount of heat is determined. The weight of the diverted PuO_2 sample is calculated and the mass of aluminum equal to that is determined, minus the mass of ^{241}Am which has already been added. Gamma ray measurements before and after diversion are simulated in MCNP. A 2″x2″ coaxial P-type HPGe detector model is used. The gamma ray emissions of the material is calculated using RadSrc. The two spectrum are compared to determine if there is a detectable difference between them.

Table 5 shows the densities of the materials used in this analysis and the MCNP simulations.

2.6. Initial can model

The total mass of plutonium in the can is 1000 g (1133 g of PuO_2). The can is a right circular cylinder with height and diameter of the PuO_2 equal to 8.23 cm. The steel can casing is 0.185 cm thick. The plutonium vector used in this study is shown in Table 6. This isotopic concentration was chosen because it represents typical reactor grade plutonium.

Table 5
Mass density [14].

Material	Density [g/cm^3]
PuO_2	2.5
Al	2.8
^{241}Am	12
^{252}Cf	15

Table 6
Plutonium vector.

Isotope	wt. %
^{238}Pu	1.6
^{239}Pu	58.8
^{240}Pu	20.8
^{241}Pu	13.8
^{242}Pu	5

3. Results and discussion

3.1. Post-diversion can model

The diverted material was taken from the center of the can as to minimize the probability of detection from gamma ray spectroscopy. In order to maintain isotropy, the material is diverted as a sphere. The density of the PuO_2 powder shown in Table 5 and the weight percent of the plutonium in the powder (88.2 wt %) are used to calculate the radius of the diverted sphere (2.212 cm). The aluminum and americium are added as shells within the cavity left by the diverted material. The californium is a small sphere within the center of the cavity.

Fig. 1 shows the MCNP geometry of the can. The blue is the can casing (steel). The olive color is the PuO_2 powder. The green is the aluminum. The thin yellow shell is the ^{241}Am. The orange is air. Considering the small scale of the ^{252}Cf mass, it cannot be seen in the figure but is directly in the center of the innermost sphere where the small crosshair is located in the figure.

Although the aluminum is intended to compensate for the mass diverted, the mass of americium must first be calculated as to not overcompensate in mass. Considering the ^{252}Cf mass will be well below the detectable sensitivity of the scale, its mass will be neglected.

Using the mass of diverted plutonium, the vector in Table 6, and the decay heat generation in Table 4, the heat generation from the decay of the whole can is 11.918 W. The heat generation from the decay of the diverted plutonium is 1.192 W. The fractional uncertainty in the heat decay measurement is 0.003, which corresponds to an uncertainty of 0.036 W in the measurement of the decay heat of the whole can. Thus the decay heat of the ^{241}Am must be

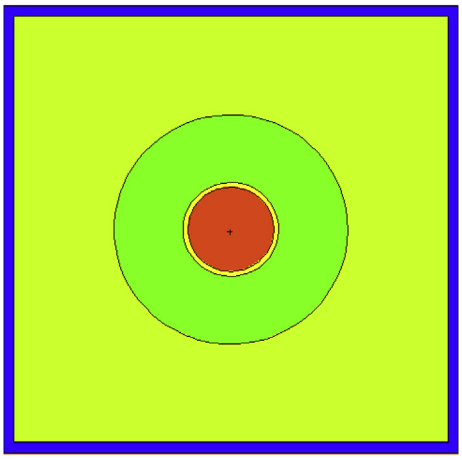

Fig. 1. Side view of the MCNP model of the can geometry with the inner sphere of PuO_2 replaced with the replacement material.

between 1.156 W and 1.228 W so that the total decay heat will be within the measurement uncertainty. This range corresponds to a ^{241}Am mass between 10.363 and 11.005 with the median mass being 10.684 g. Subtracting this mass from the total diverted mass (113.340 g), yields a mass difference of 102.656 g. Therefor 102.656 g of aluminum is required.

In order to calculate the mass of ^{252}Cf required, the singles, doubles, and triples counts of the whole can as a result of fission and (alpha,n) reactions are found using MCNP. Then, the singles, doubles, and triples counts from the remaining plutonium dioxide after diversion are found. The measurement time is assumed to be 300 s and the AWCC is in fast mode. Using Equation (7), the neutron die away time was calculated to be 50 μs Using Equation (8), the efficiency was calculated to be 31.63%. These values agree with known values for each [8]. The multiplicity counts are shown in Table 7.

Initially approximating the mass of ^{252}Cf is done using Equation (2). However, this approximation is less than the actual amount needed because the absence of the diverted material causes less induced fission overall. The mass can be better approximated by using the doubles counts given by the MCNP output and the spontaneous fission neutron generation rate of ^{252}Cf. The ^{252}Cf source is small enough to appear as a point source to the detector. Thus the output from the MCNP simulations remain unchanged with slight changes in the ^{252}Cf mass. The amount of mass is determined by diving the doubles counts required by the normalized doubles count from the MCNP simulation of the spontaneous fission of ^{252}Cf and the spontaneous fission neutron generation rate of ^{252}Cf. This method produced a ^{252}Cf mass of 14,201 ng. Table 8 shows the singles, double, and triples counts including the ^{252}Cf source. The uncertainties were calculated using Equations (4)–(6). The singles counts uncertainty is significantly less than the difference in the pre-diversion and post-diversion case so the difference would be detectable. However, the doubles and triples counts different wouldn't be detectable.

Using Equation (5), the three sigma uncertainty of the post-diversion doubles counts is found to be 148,777 counts. This is approximately a 1.62% uncertainty. Therefore, the difference in doubles counts before and after diversion would be undetectable.

In addition to comparing the doubles counts, Equation (2) can be used to calculate the $^{240}Pu_{eff}$ mass which can be compared with the known value that can be obtained from Equation (2). The calculated $^{240}Pu_{eff}$ mass is 334.99 g. The known value is 332.32 g. This is a percent difference of 0.80% and would be practically undetectable as the sensitivity of the mass calculation from a passive measurement is 1% [16].

Fig. 2 shows the simulations results of the gamma ray spectra collected from an HPGe detector pre-diversion and post-diversion. From inspection of the spectra, two prominent ^{241}Am peaks at 662 keV and 722 keV are clearly visible in the post diversion spectrum. The 662 keV peak in the post diversion spectrum has $[4.28 \pm 0.02] \cdot 10^5$ counts. The number of counts found in the same area of the pre diversion spectrum is $[9.91 \pm 0.05] \cdot 10^4$ counts. Considering the difference between the numbers of counts is significantly greater than the uncertainties, the peak would be detectable. However, this technique assumes the plutonium is fresh. ^{241}Am will be present in older plutonium and this analysis would be comparing the difference in the ^{241}Am peaks rather than comparing spectra with and without the peaks. The intensities of these peaks will be noticeable different as the mass of ^{241}Am in older plutonium will be significantly less.

3.2. Detecting the diversion

Although initially the diversion would be undetectable when considering only the doubles counts, after some time the ^{252}Cf will decay enough to produce doubles counts that will not be large enough to reach the three sigma uncertainty threshold. In order to detect the diversion, the doubles counts must be 148,777 counts less than the doubles counts from the can before diversion. The ^{252}Cf must produce less than 1,141,804 counts. Using the same method used to approximate the initial mass of ^{252}Cf, the mass that will produce 1,141,804 counts is determined to be 12.564 ng. Converting the masses to number of atoms and using Equation (9), the amount of time required to decay is 161 days. Table 9 shows the resulting singles, doubles, and triples counts pre-diversion and post-diversion after 161 days has passed. This doubles counts percent difference is equal to the three sigma uncertainty mentioned previously.

In addition to the decay of ^{252}Cf affecting measurement, the decay of ^{241}Am will affect the calorimetry measurement. Considering the calorimeter can measurement the ^{241}Am mass within 0.321 g, once the ^{241}Am mass reaches 0.321 g below its initial mass, the calorimeter will be able to detect the difference. However, the time required for this decay to occur is 19 years, therefore this detection will not be timely.

The diversion may also be detectable if the AWCC is switched to thermal mode (the cadmium sleeve is removed). Using Equation (7), the neutron die away time for this configuration was calculated to be 57 μs Using Equation (8), the efficiency for this configuration was calculated to be 36.24%. Both are slightly larger than in the fast configuration, as would be expected. Table 10 shows the resulting pre-diversion and post-diversion singles, doubles, and triples counts from the MCNP simulations of the AWCC in thermal mode. As with the case of fast mode, the singles uncertainty is lower than the difference between pre-diversion and post-diversion. Also, the uncertainty for the doubles and triples are larger than the difference thus the difference would be undetectable.

Using the method described in section 3.1, the $^{240}Pu_{eff}$ mass can be calculated using the multiplicity counts in Table 10. The calculated value post-diversion is 340.79 g. The known value is 332.32 g. The difference should be detectable as the difference is beyond 1%.

Another major obstacle in using this diversion technique is the difficulty in obtaining the replacement materials. Although obtaining aluminum is easy, obtaining ^{252}Cf and ^{241}Am are not as straightforward. 14.201 ng of ^{252}Cf is commercially available for purchase, in limited quantities [17], but the mass would need to be precise which would be difficult. Although not as difficult, obtaining a precise amount of ^{241}Am would also be difficult. However, diversion only yields 100 g of plutonium and would need to be repeated 80 times in order to obtain one significant quantity of plutonium (8 kg) [18]. In order to divert one significant quantity of plutonium, approximately 855 g of ^{241}Am would be required. Obtaining this much ^{241}Am commercially is not practical. Since

Table 7
Multiplicity counts without 252Cf [counts].

	Singles	Doubles	Triples
Pre-diversion	58,135,203 ± 9268	9,156,926 ± 51,569	1,716,938 ± 137,917
Post-diversion	51,879,577 ± 8702	7,866,345 ± 45,639	1,380,293 ± 115,338
Difference	6,255,625 ± 12,713	1,290,581 ± 68,864	336,645 ± 179,789

Table 8
Multiplicity Counts in fast mode with 252Cf [counts].

	Singles	Doubles	Triples
Pre-diversion	58,135,203 ± 9268	9,156,926 ± 51,569	1,716,938 ± 137,917
Post-diversion	55,232,769 ± 9110	9,156,926 ± 49,592	1,715,103 ± 129,703
Difference	2,902,434 ± 12,996	0 ± 71,545	1835 ± 189,325
Percent Difference	4.99 ± 0.03%	0.00 ± 0.78%	0.11 ± 11.03%

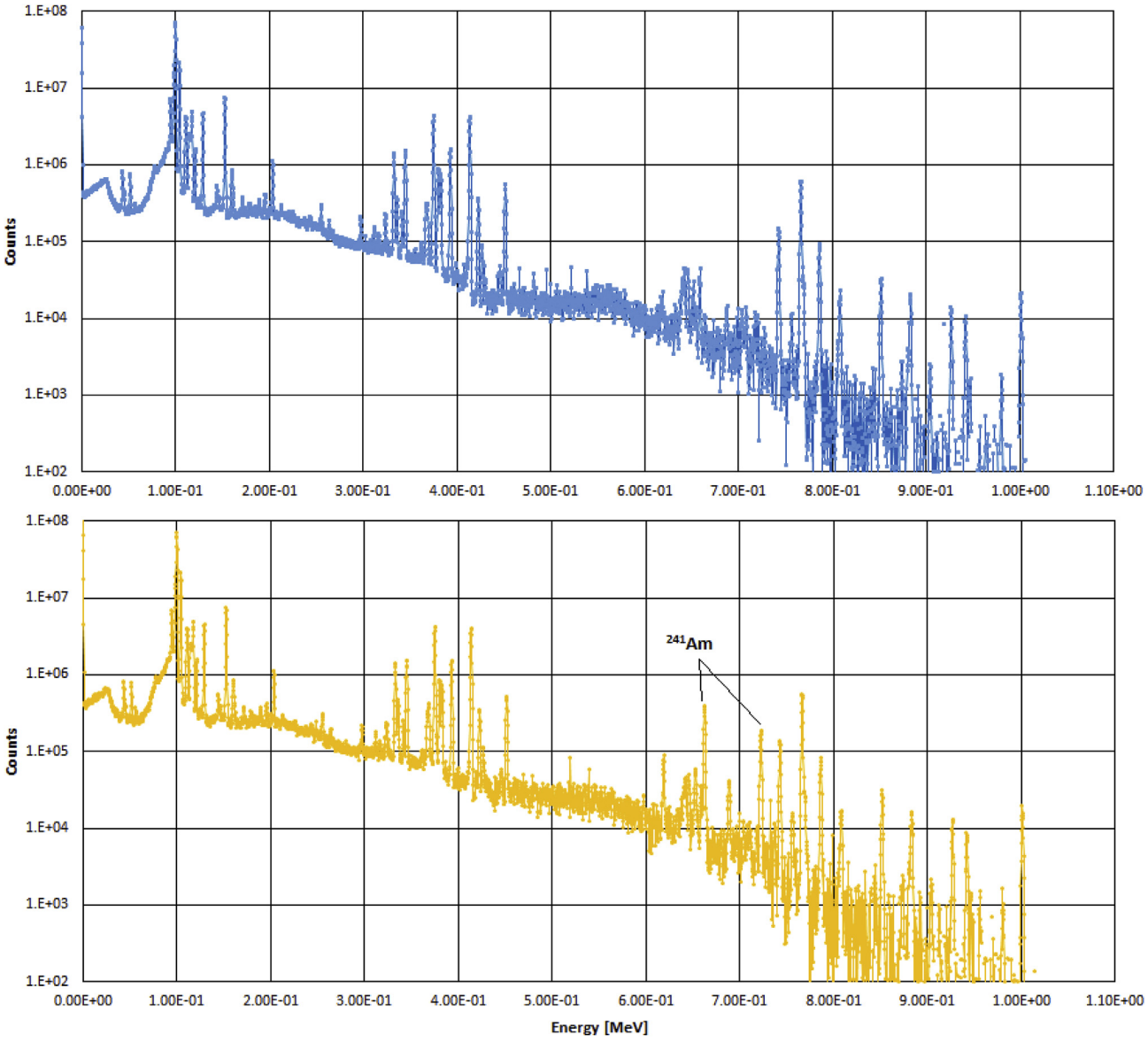

Fig. 2. Gamma ray spectra, top: pre diversion, bottom: post diversion.

Table 9
Doubles counts pre-diversion and post-diversion after decay.

	Singles	Doubles	Triple
Pre-diversion	58,135,203	9,156,926	1,716,938
Post-diversion with reduced ^{252}Cf mass	54,846,301	9,008,149	1,676,515
Percent Difference	5.66%	1.62%	2.35%

^{241}Am is not safeguarded, it may be possible for the state diverting the PuO_2 to create and separate their own ^{241}Am, if they have reprocessing capabilities.

4. Conclusion

This study shows that although difficult, currently used safeguards techniques can effectively detect the diversion. The detection of the diversion via mass and calorimetry measurements can be prevented with the addition of aluminum and ^{241}Am. Only considering the doubles counts or calculating the mass using singles, doubles, and triples counts from AWCC measurements in fast or thermal mode would not detect the diversion. However the difference in the singles counts before and after diversion is

Table 10
Multiplicity Counts in thermal mode with 252Cf [counts].

	Singles	Doubles	Triples
Pre-Diversion	69,473,509 ± 10,312	11,511,497 ± 61,885	2,294,626 ± 180,615
Post-Diversion	66,366,355 ± 10,152	11,455,697 ± 59,696	2,278,987 ± 170,712
Difference	3,107,154 ± 14,471	55,800 ± 85,984	15,639 ± 248,524
Percent Difference	4.47 ± 0.02%	0.48 ± 0.75%	0.68 ± 10.83%

significant enough for the diversion to be detected in both fast and thermal mode. However, neutron scattering due to sample geometry and (alpha, neutron) rate changes due to PuO_2 density can have considerable effects on the singles counts. It is unknown by the authors if an approximate 5% difference in the singles counts is sufficient for an IAEA inspector to investigate further. Follow up measurements of this can would indicate a material diversion due to the decrease in doubles counts. The relatively short half-life of ^{252}Cf will cause this difference to increase to a statistically significant value in a relatively short time span. This time limit represents a significant challenge in the adversary's perspective for this diversion technique. In addition, gamma ray measurements of the can pre diversion and post diversion show that the addition of ^{241}Am is detectable. The addition of the ^{241}Am will also impact the isotopics calculated via gamma ray spectroscopy. This would then lead to additional error in the multiplicity and calorimetry measurements that would increase the possibility of detection. The difficulty in obtaining the materials for this diversion and the precision required in adding the specified mass of replacement materials makes this scenario a low probability event. Overall, this diversion scenario can be detected relatively quickly and would not be a viable manner for a state to covertly obtain enough plutonium for a weapon.

Declaration of competing interest

There is no conflict of interest with this work.

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