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Justification of Shielding Requirements for Plutonium Sources

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Abstract

This report provides a justification for shielding plutonium sources when testing radiation detection instruments for standard performance requirements and test methods. The use of copper as a shielding material is proposed to provide comparable test results when testing radiation detection instruments in different locations using different plutonium sources with varying ^{241}Am content.

Introduction

In 2008 the National Institute of Standards and Technology (NIST) participated in the Domestic Nuclear Detection Office (DNDO) test of advanced spectroscopic portal (ASP) monitors to characterize the performance of these systems. Several plutonium sources were used for testing the alarm and radionuclide identification response of the ASPs. During these tests it was found that the americium-241 (^{241}Am) content in the various plutonium sources had a significant impact in the identification of plutonium-239 (^{239}Pu). This report summarizes how this problem was addressed.

Differences in the ^{241}Am content are due to the isotopic composition (^{241}Pu content) and age of the Pu source. It became clear that it was necessary to limit the emission of the 59.54 keV gamma-ray line in order to have comparable instrument response results when testing with different sources in different test locations. At that point in time the DNDO and NIST scientists decided that for weapons grade plutonium (WGPu) sources the net count rate of the 59.54 keV gamma-ray line from ^{241}Am shall be no more than 10 times greater than that of the net count rate of the 414 keV line for ^{239}Pu (e.g., if the count rate for ^{239}Pu is 100 cps then the count rate for the 59.54 keV line for ^{241}Am shall not exceed 1,000 counts per second (cps)). This amount allows the detection of ^{241}Am without distorting the high energy spectrum that could potentially prevent the identification of ^{239}Pu. Based on NIST and DNDO calculations it was determined that a medium-z material, such as copper, was the best option to effectively reduce the ^{241}Am contribution with minimum distortion of the energy spectrum in the energy range between 300 keV and 500 keV.

This approach was later used for other DNDO test campaigns including those named Eland and PaxBag. When Pu sources were used in test campaigns, it was clear that the identification response (radionuclide identification answer) provided by the radionuclide identifier detectors (RIIDs), ASP, mobile systems with spectroscopic capabilities, spectrometric personal radiation detectors, stations with spectroscopic capabilities, and spectrometric pedestrian portal monitors, strongly depended on the amount of ^{241}Am present in the source.

As performance standards were being developed or revised, the importance of using this technique became clearer. Testing of radiation detection instruments against the standards can occur at different testing facilities and having access to the same Pu source is almost impossible due to availability and shipping restrictions.

During the development of the RIIDs technical capability standard (TCS) document several measurements using different Pu sources were made at Los Alamos National Laboratory (LANL) by NIST staff. The measurements were performed with bare and shielded Pu sources. Results showed that the ^{241}Am content varied over approximately three orders of magnitude indicating that it was also important to specify the shielding material. During the TCS working group meetings the use of cadmium (Cd) or copper (Cu) for shielding the Pu sources was discussed.

The ASTM C 993 – 97 standard [1] also discusses the ^{241}Am content in Pu sources. This standard defines the following:

> *standard plutonium test source*—a metallic sphere or cube of low-burnup plutonium containing at least 93 % ^{239}Pu, less than 6.5 % ^{240}Pu, and less than 0.5 % impurities.
>
> *Discussion*—A cadmium filter can reduce the impact of ^{241}Am, a plutonium decay product that will slowly build up in time and emit increasing amounts of 60-keV radiation. Begin use of a 0.04-cm thick cadmium filter when three or more years have elapsed since separation of plutonium decay products. If ten or more years have elapsed since separation, use a cadmium filter 0.08 cm thick. The protective encapsulation should be in as many layers as local rules require. A nonradioactive encapsulation material, such as, aluminum (0.32 cm-thick) or thin (0.16 cm-thick) stainless steel or nickel, should be used to reduce unnecessary radiation absorption.

The guidance provided in the American Standard Test Method (ASTM) standard is a rule of thumb approach (i.e., providing shielding thicknesses based on the age of the Pu sources) but it does not provide the necessary accuracy for the comparisons of test results when testing against a performance standard. For the purpose of the ASTM standards this rule of thumb is enough as it does not compare the performance requirements between different instruments, especially spectrometric type instruments. Spectrometric type instruments may not identify the presence of ^{239}Pu when a large amount of ^{241}Am is present in the spectrum. A more accurate determination of the emission of the 59.54 keV gamma-ray line from ^{241}Am is obtained by measuring the energy spectrum of the Pu source using a high purity germanium (HPGe) detector and limiting the net count rate of the 59.54 keV gamma-ray line from ^{241}Am to be no more than 10 times greater than that of the net count rate of the 414 keV line for ^{239}Pu, as adopted by DNDO for testing of radiation detection instruments.

The use of Cd to shield the Pu sources was further discussed in the TCS working group meetings. Machining of Cd is hazardous if inhaled due to the toxicity of the dust particles. Therefore, the use of copper was suggested. LANL proposed the use of a specific copper alloy to minimize differences when testing at different facilities. The use of copper with more than 99.9 % Cu content listed in the ASTM B152 standard [2] was proposed for the TCS.

Supporting data

During the development of the RIIDs TCS several measurements using different Pu sources were made in LANL. Figure 1 and Figure 2 show the energy spectra of different WGPu sources acquired using a NaI(Tl) and HPGe detector respectively. The differences in the structure in the 60 keV energy region due to the presence of ^{241}Am is clearly seen. Figure 3 shows the energy spectra of a bare and shielded WGPu source acquired at Oak Ridge National Laboratory (ORNL). The shielding used for this measurement was 5 mm steel.

Calculations were performed to look at the difference in the attenuation coefficients and factors when using different materials for shielding the Pu sources in order to reduce the ^{241}Am contribution while minimizing the attenuation of the lines in the 300 to 500 keV energy region. Table 1 summarized the calculations for the different proposed materials. The Photon Cross Sections Data (XCOM) Database [16] was used to obtain the numbers listed in Tables 1 and 3. This database does not provide uncertainties for the calculated attenuation coefficients. Therefore, it is not possible to estimate the uncertainties for the values listed in Tables 1 and 3.

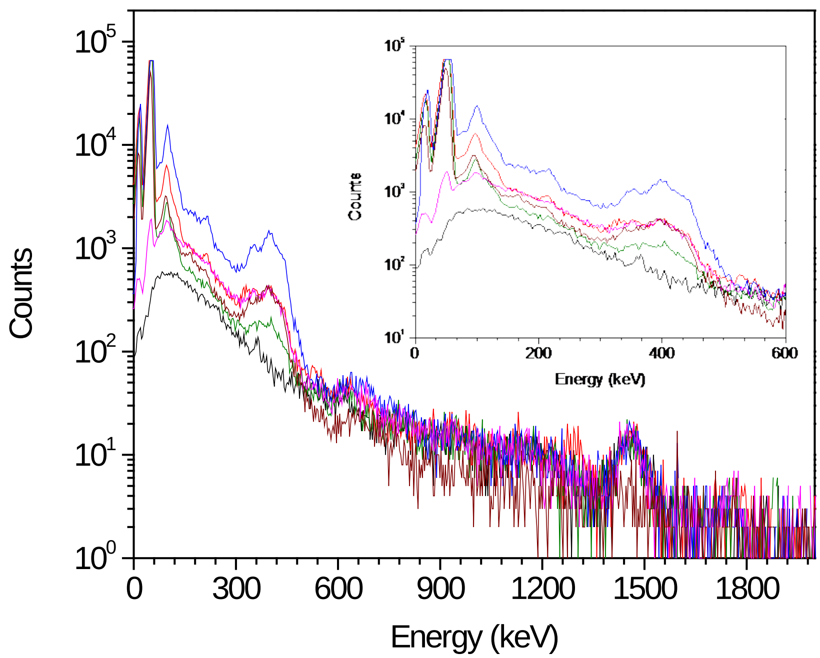

Figure 1: Energy spectra of different bare Pu sources from LANL using a NaI detector. The black line represents the background.

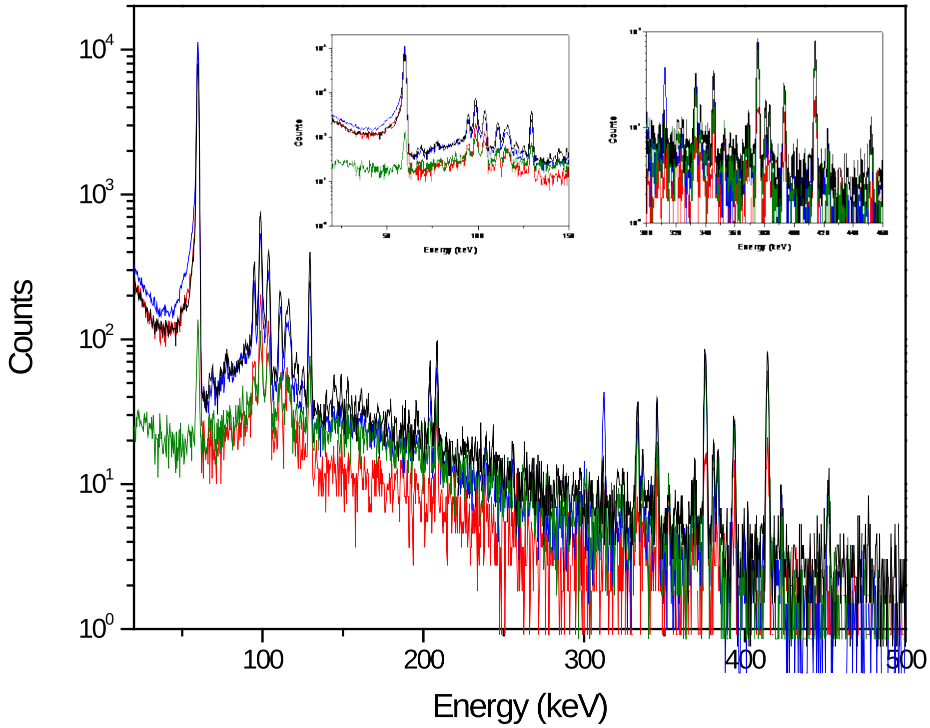

Figure 2: Energy spectra of different bare Pu sources from LANL using a HPGe detector

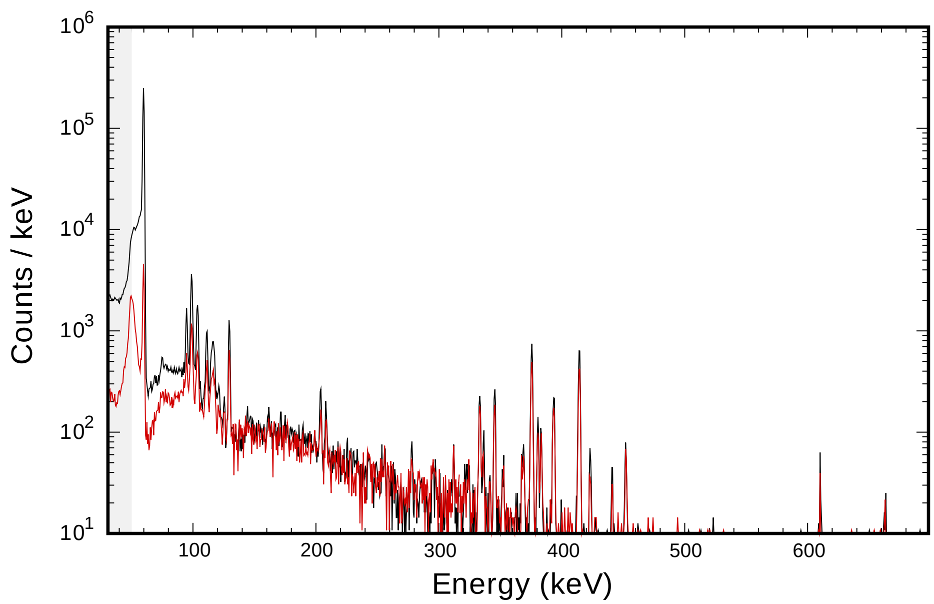

Figure 3: Energy spectra of bare and shielded Pu source from ORNL using a HPGe detector. The black line represents the bare Pu spectrum and the red line represents the spectrum of the shielded Pu source using a 5 mm steel plate.

Table 1: Summary of attenuation calculations

Shielding Material	Density (g/cm^3)	Attenuation coefficient (cm^2/g)	Attenuation Factor for 1 cm material	Attenuation Factor for 5 mm material
Aluminum	2.699	60 keV – 0.2778 400 keV – 0.09276	60 keV – 0.472 400 keV – 0.779	60 keV – 0.687 400 keV – 0.882
Iron	7.874	60 keV – 1.205 400 keV – 0.09400	60 keV – 7.57×10^{-5} 400 keV – 0.477	60 keV – 8.70×10^{-3} 400 keV – 0.691
Copper	8.960	60 keV – 1.593 400 keV – 0.09413	60 keV – 6.33×10^{-7} 400 keV – 0.430	60 keV – 7.95×10^{-4} 400 keV – 0.656
Cadmium	8.650	60 keV – 5.975 400 keV – 0.1129	60 keV – 3.58×10^{-23} 400 keV – 0.377	60 keV – 5.98×10^{-12} 400 keV – 0.614

There are many different copper and copper-alloy compositions that are grouped into the categories: copper, high copper alloy, brasses, bronzes, copper nickels, copper–nickel–zinc (nickel silver), leaded copper, and special alloys. The following table lists the principal alloying element for four of the more common types, along with the name for each type.

Table 2: Classification of copper and its alloys and associated Unified Numbering System (UNS) number

Family	Principal alloying element	UNS number
Copper alloys, brass	Zinc (Zn)	C1xxxx–C4xxxx,C66400–C69800
Phosphor bronze	Tin (Sn)	C5xxxx
Aluminum bronzes	Aluminum (Al)	C60600–C64200
Silicon bronzes	Silicon (Si)	C64700–C66100
Copper nickel, nickel silvers	Nickel (Ni)	C7xxxx

The composition of these different alloys meets different standards requirements as shown in Table 3. Due to the difference in elemental composition, the x-ray and gamma-ray attenuation through different materials can vary from alloy to alloy. It is therefore recommended that in order to gain consistency in testing, the Pu source be shielded using copper with a purity of 99.90% or better (i.e., C101 or UNS No. C11000) to reduce the low-energy emissions from the 59.54 keV ^{241}Am line. The copper thickness will depend on the ^{241}Am content of the source, in order to ensure that the net count rate of the 59.54 keV gamma-ray line from ^{241}Am is not more than 10 times greater than the net count rate of the 414 keV line for ^{239}Pu, while the 59.54 keV gamma-ray line from ^{241}Am is still visible in the energy spectrum.

Table 3: Nominal composition of copper alloys

Alloy Name	Nominal composition	Attenuation Coefficient (cm^2/g)	Calculated Density (g/cm^3)
Copper (ASTM B152, B124)	More than 99.9 % Cu	60 keV – 1.593 400 keV – 0.09413	8.960
Gilding metal (ASTM B36)	95.0 % Cu, 5.0 % Zn	60 keV – 1.601 400 keV – 0.09419	8.847
Cartridge brass (ASTM B19,	70.0 % Cu, 30.0 % Zn	60 keV – 1.644	8.321

B36, B135)		400 keV – 0.09451	
Phosphor bronze (ASTM B103, B139)	89.75% Cu, 10.0 % Sn, 0.25 % P	60 keV – 2.448 400 keV – 0.09458	8.697
Yellow or High brass (ASTM B36, B135)	65.0% Cu, 35.0 % Zn	60 keV – 1.652 400 keV – 0.09782	8.223
Naval brass (ASTM B21)	60.0 % Cu, 39.25 % Zn, 0.75 % Sn	60 keV – 1.728 400 keV – 0.09492	8.129
Muntz metal (ASTM B111)	60.0 % Cu, 40.0 % Zn	60 keV – 1.661 400 keV – 0.09464	8.127
Aluminium bronze (ASTM B169, B124, B150)	92.0 % Cu, 8.0 % Al	60 keV – 1.546 400 keV – 0.09408	8.780
Beryllium copper (ASTM B194, B196)	97.75 % Cu, 2.0 % Be, 0.25 % Co or Ni	60 keV – 1.588 400 keV – 0.09411	8.320
Cupronickel	70.0 % Cu, 30.0 % Ni	60 keV – 1.569 400 keV – 0.09513	8.943
Ounce metal Copper Alloy C83600 (also known as "Red brass" or "composition metal") (ASTM B62)	85.0 % Cu, 5.0 % Zn, 5.0 % Pb, 5.0 % Sn	60 keV – 2.484 400 keV – 0.1154	9.198

Summary

The use of any copper with more than 99.9 % Cu content listed in the ASTM B152 standard [2] is an adequate choice for reducing the amount of the ^{241}Am gamma-ray emission from Pu sources. Commonly used copper types for this type of application are the UNS No. C11000 or the C10100. The C11000 (equivalent to the C101 British designation) copper has a purity of 99.90 %. The C10100 (equivalent to the C110 British designation) copper is the oxygen free type and has the highest purity of 99.99 %.

The thickness of the copper shielding shall be based on the ^{241}Am content of the source. The copper thickness needs to be chosen to ensure that the net count rate of the 59.54 keV gamma-ray line from ^{241}Am to be no more than 10 times greater than that of the net count rate of the 414 keV line for ^{239}Pu while the 59.54 keV gamma-ray line from ^{241}Am is still visible in the energy spectrum.

References

1. ASTM C 993 – 97, “Standard Guide for In-Plant Performance Evaluation of Automatic Pedestrian SNM Monitors.”
2. ASTM B152/B152M-09, “Standard Specification for Copper Sheet, Strip, Plate and Rolled Bar.”
3. ASTM B124/B124M-12, “Standard Specification for Copper and Copper Alloy Forging Rod, Bar, and Shapes.”
4. ASTM B36/B36M-08a, “Standard Specification for Brass Plate, Sheet, Strip, and Rolled Bar.”
5. ASTM B19-10, “Standard Specification for Cartridge Brass Sheet, Strip, Plate, Bar, and Disks.”
6. ASTM B135-10, “Standard Specification for Seamless Brass Tube.”

7. ASTM B103/B103M-10, “Standard Specification for Phosphor Bronze Plate, Sheet, Strip, and Rolled Bar”
8. ASTM B139/B139M-07, “Standard Specification for Phosphor Bronze Rod, Bar, and Shapes.”
9. ASTM B21/B21M-12, “Standard Specification for Naval Brass Rod, Bar, and Shapes.”
10. ASTM B111/B111M-11, “Standard Specification for Copper and Copper-Alloy Seamless Condenser Tubes and Ferrule Stock.”
11. ASTM B169/B169M-10, “Standard Specification for Aluminum Bronze Sheet, Strip, and Rolled Bar.”
12. ASTM B150/B150M-08, “Standard Specification for Aluminum Bronze Rod, Bar, and Shapes.”
13. ASTM B194-08, “Standard Specification for Copper-Beryllium Alloy Plate, Sheet, Strip, and Rolled Bar.”
14. ASTM B196/B196M-07, “Standard Specification for Copper-Beryllium Alloy Rod and Bar.”
15. ASTM B62-09, “Standard Specification for Composition Bronze or Ounce Metal Castings.”
16. Xcom website, http://physics.nist.gov/PhysRefData/Xcom/html/xcom1.html