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Abstract

Gamma rays are ideal probes for the determination of information about the special nuclear material that is in the transparency regime. Gamma rays are good probes because they interact relatively weakly with the containers that surround the SNM under investigation. In addition, gamma rays carry a great deal of information about the material under investigation. We have leveraged these two characteristics to develop three technologies that have proven useful for the measurements of various attributes of plutonium. These technologies are Pu-300, Pu-600 and Pu-900. These technologies obtain the age, isotopics and presence/absence of oxide of a plutonium sample, respectively. Pu-300 obtains the time since the last ^{241}Am separation for a sample of plutonium. This is accomplished by looking at the ^{241}Am/^{241}Pu ratio in the energy region from 330-350 keV, hence the name Pu-300. Pu-600 determines the isotopics of the plutonium sample under consideration. More specifically, it determines the ^{240}Pu/^{239}Pu ratio to determine if the plutonium sample is of weapons quality or not. This analysis is carried out in the energy region from 630-670 keV. Pu-900 determines the absence of PuO_2 by searching for a peak at 870.7 keV. If this peak is absent then there is no oxide in the sample. This peak arises from the de-excitation of the first excited state of ^{17}O. The assumption being made is that this state is populated by means of the $^{17}O(\alpha,\alpha')$ reaction. The first excited state of ^{17}O could also be populated by means of the $^{14}N(\alpha,p)$ reaction, which might indicate that this is not a good signature for the absence of PuO_2 , however in the samples we have measured this peak is visible in oxide samples and is absent in other samples. In this paper we will discuss the physics details of these technologies and also present results of various measurements.

Introduction

The Fissile Material Transparency Technology Demonstration (FMTTD) had specific requirements for attributes that had to be determined for plutonium sources. Mainly, these are the

- presence of plutonium
- time since the last americium separation
- plutonium isotopics
- absence of plutonium oxide
- mass of the plutonium sample
- symmetry of the source.

The first four of these attributes were ideally suited for determination through gamma ray spectrometry. We developed several algorithms and leveraged other algorithms that had already been developed to determine the attributes under consideration. There are three codes that have been developed and implemented for the FMTTD. These codes are Pu-300, Pu-600 and Pu-900. Pu-

300 obtains the time since the last ^{241}Am separation for a sample of plutonium. This is accomplished by looking at the $^{241}Am/^{241}Pu$ ratio in the energy region from 330-350 keV, hence the name Pu-300. Pu-600 determines the isotopics of the plutonium sample under consideration. More specifically, it determines the $^{240}Pu/^{239}Pu$ ratio to determine if the plutonium sample is of weapons quality or not. This analysis is carried out in the energy region from 630-670 keV. Pu-900 determines the absence of $Pu0_2$ by searching for a peak at 870.7 keV. If this peak is absent then there is no oxide in the sample. This peak arises from the de-excitation of the first excited state of ^{17}O.

These codes are part of a system that includes the software and the hardware. The software – in terms of how the physics algorithms are implemented - will be described below. The hardware - for the spectrometry system - was mostly commercial off the shelf (COTS) components. The gamma ray radiation is detected in HPGe detector. The Pu-300 and Pu-600 share a detector. This detector is a 50 percent N-type detector. We chose an N-type detector to limit the amount of neutron damage in the crystal. The signals from the preamp of the HPGe detector were sent to two sources. The first output goes to a shutter system and the second output goes to a Canberra InSpector™. The shutter is a mechanical device that keeps the dead time in the detector at a relatively constant rate. The shutter was implemented so that no information can be discerned about the strength of the source. The issue here is that the gamma flux from a classified source is sensitive. By keeping the counting time and detector to the source distance constant very little information can be ascertained. A picture of the hardware is shown in figure 1.

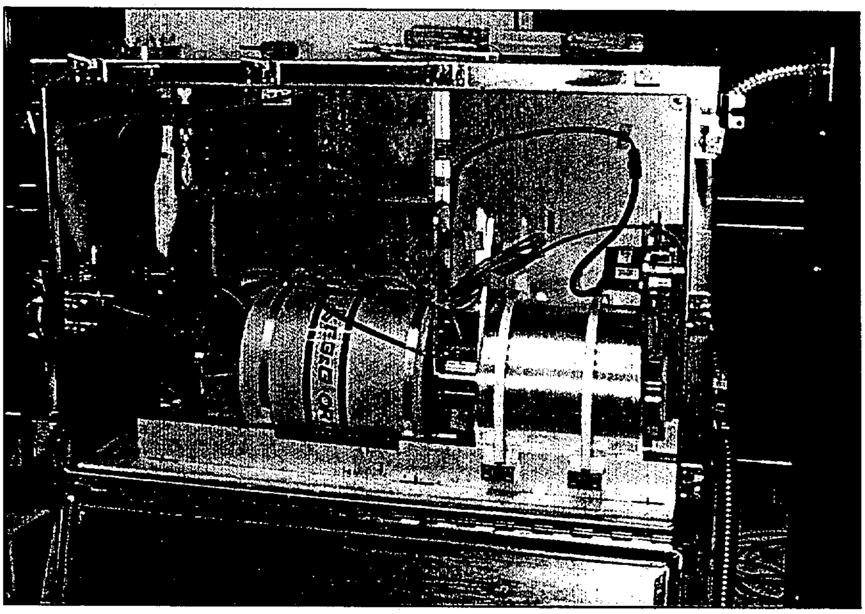

Figure 1. Pu300/Pu600 Hardware

The second analog output from the HPGe detector is input into a Canberra InSpector™ . This instrument is a combination high voltage supply, spectroscopy amplifier and Multi-channel analyzer. The InSpector™ is controlled by a simple C program which runs under DOS. This control program takes advantage of the Canberra DOS libraries. The control program was "home-brewed" so that all over-head from the commercial code could be avoided. In addition, the code had to be made to run under DOS with very little memory. The control program was written in C and took advantage of the Canberra DOS Libraries. The computer used for the control and analysis is a single board computer based on a PC104 bus. The single board computer is booted off a pROM and contains only volatile memory. In addition, to the DOS operating system, the pROM contains the control and analysis software for the system. When the system is in its operational mode, it is operated using a single button. This eliminates any ambiguity in the operation of the system.

Pu300 – Plutonium Presence and time since Americium separation

This code was developed to determine the time since the last americium separation. The method is based on determining the ^{241}Am/^{241}Pu ratio – in the 300 keV region of the spectrum – in order to obtain the time since the last americium separation. The method takes advantage of the fact that ^{241}Pu decays to ^{241}Am. By knowing the half-life for the decay of ^{241}Pu to ^{241}Am and the ^{241}Am to ^{241}Pu it is possible to determine to age since separation. However, this is slightly complicated by the fact that ^{241}Pu has a small branch that decays through ^{237}U. The branching ratio through ^{241}Am is 99.998 % and the branching ratio through ^{237}U is 0.002 % . Since the decay rates are known, as are the half-lives, the time since separation can be determined.

The heart of the method relies on the fact that it is possible de-convolute several closely spaced peaks – in the pulse height distribution – to determine the amount of ^{241}Am and ^{241}Pu in the sample. An example of the pulse height distribution that is used for the analysis under this method is shown in figure 2.

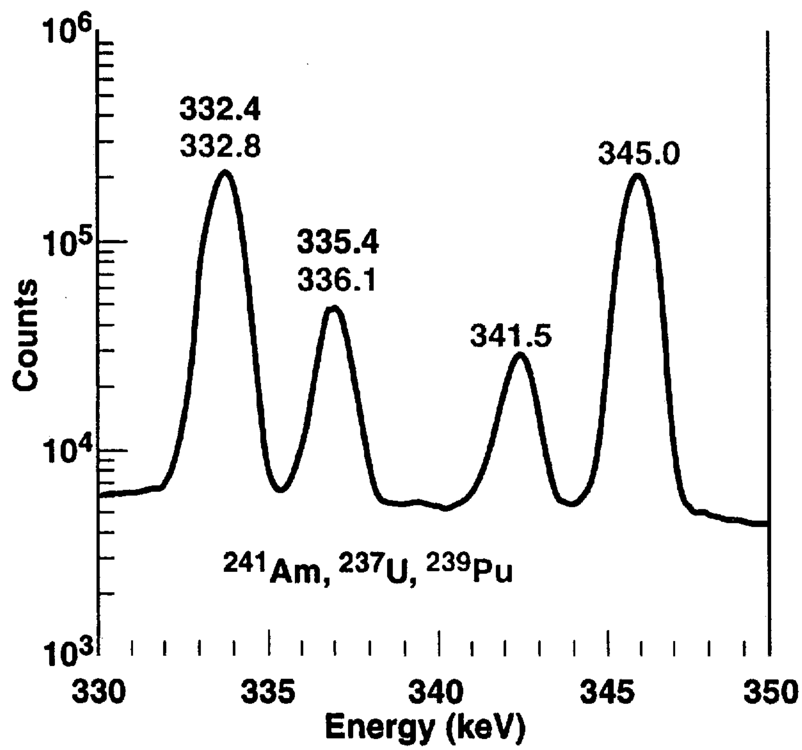

Figure 2. Pulse Distribution for the γ rays from a plutonium sample between 330 and 350 keV.

The peaks from ^{239}Pu at 341.5 and 345 keV are used to constrain the amount of ^{239}Pu in the 332.4/332.8 and the 335.4/336.1 keV doublets. This essentially reduces the problem to the solving a system of simultaneous equations with two equations and two unknowns to determine the $^{241}Am/^{241}Pu$ ratio to determine the age since the last Americium separation. Figure 3 below shows the results of the measurements during the prove-out process for the method.

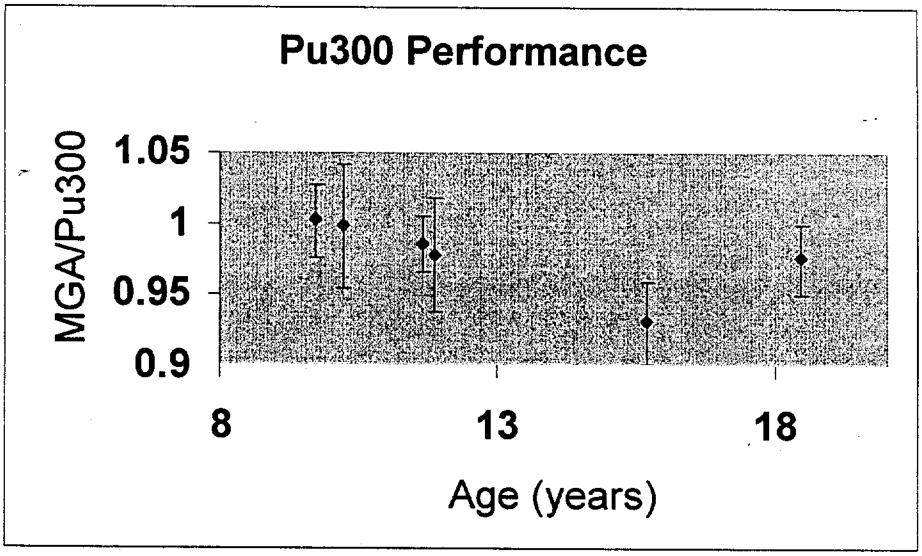

Figure 3. Example of Pu300 performance as a function of age of sample.

Figure 3 shows the age obtained from the Pu300 compared to the certified age from MGA. If the method were perfect the values for all the data points in the graph in figure 3 would all be one. The graph shows the method works reasonably well over a large spread of plutonium "ages". We have found that this method has been fairly stable in all of our tests. However, we still regard this technique as still under development.

Pu600 – Plutonium Presence and Plutonium Isotopics

In addition to the age measurements the same detector performs isotopic measurements on the sample. The measurements are normally done in series with the isotopics performed first, followed by the age measurements. The measurements are done in series, the two methods do not share any information between them.

We call the isotopics method PU600 because it looks at the gamma ray pulse height distribution in the 600 keV region. Specifically, the region of interest is between about 630 – 670 keV. An example of the pulse height distribution is shown below.

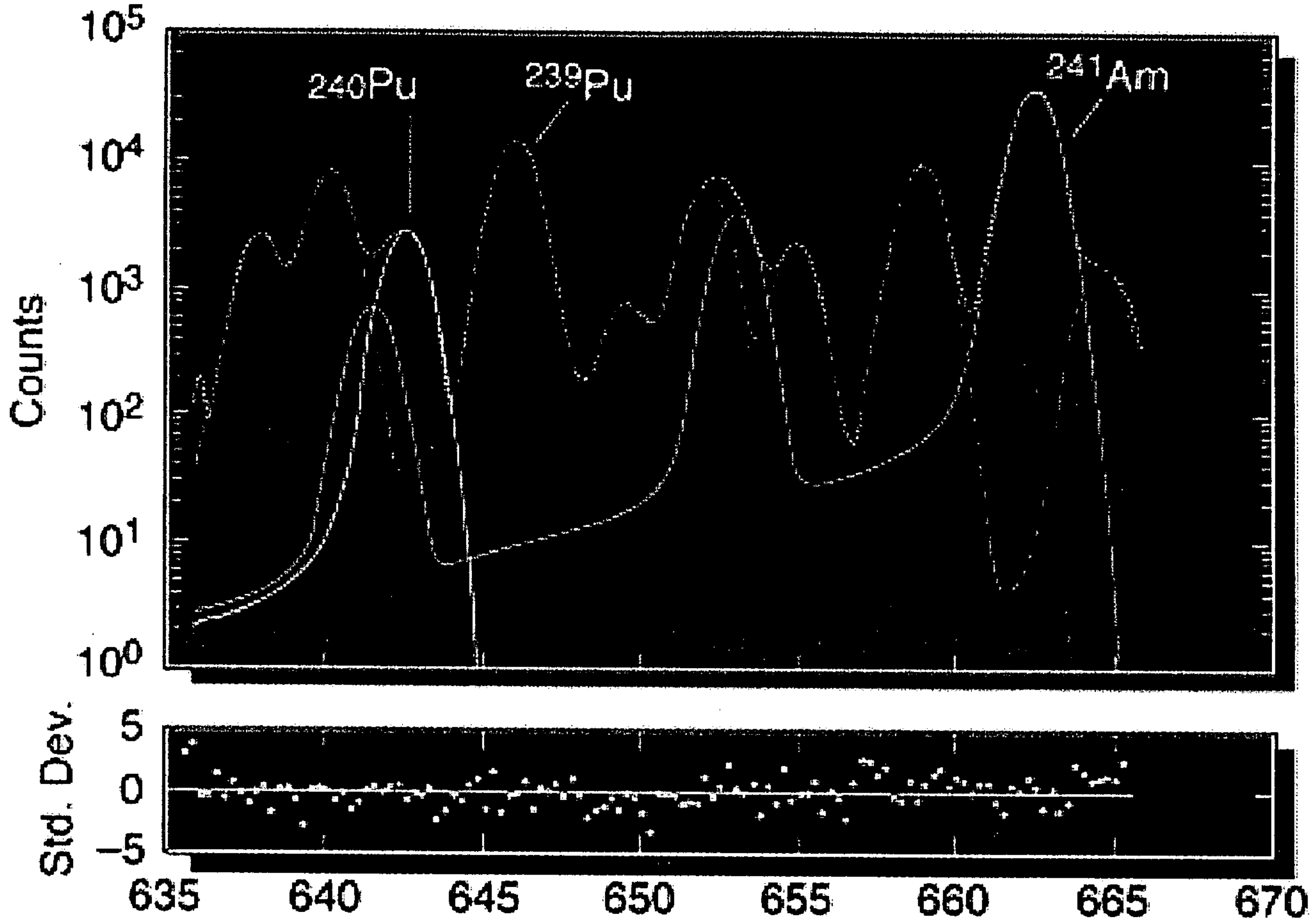

Figure 4. An example of the pulse height distribution for the 600 keV region from plutonium.

As is clear from figure 4 that there is a great deal of information contained in this pulse height distribution. The procedure to determine the ^{240}Pu/^{239}Pu ratio is analogous to the determination of the age in Pu300. The code fits the ^{239}Pu at 646 keV and uses the area in this peak (and the branching ratios) to constrain the peaks in the triplet in the region around 635-642 keV. Some of the results of this method are shown in figure 5. The data in this figure are a ratio of the Pu600 determined isotopics compared to the certified values of the isotopics. If the measurements were perfect the result would be a one for all the values. The graph in figure 5 clearly shows that the method works very well.

In addition to the isotopics we determine the presence of plutonium by the ^{239}Pu at 646 keV. We demand that there be a 5-sigma peak at this energy to indicate that plutonium is present.

We have been using this technology for several years and regard it as a fairly mature technology.

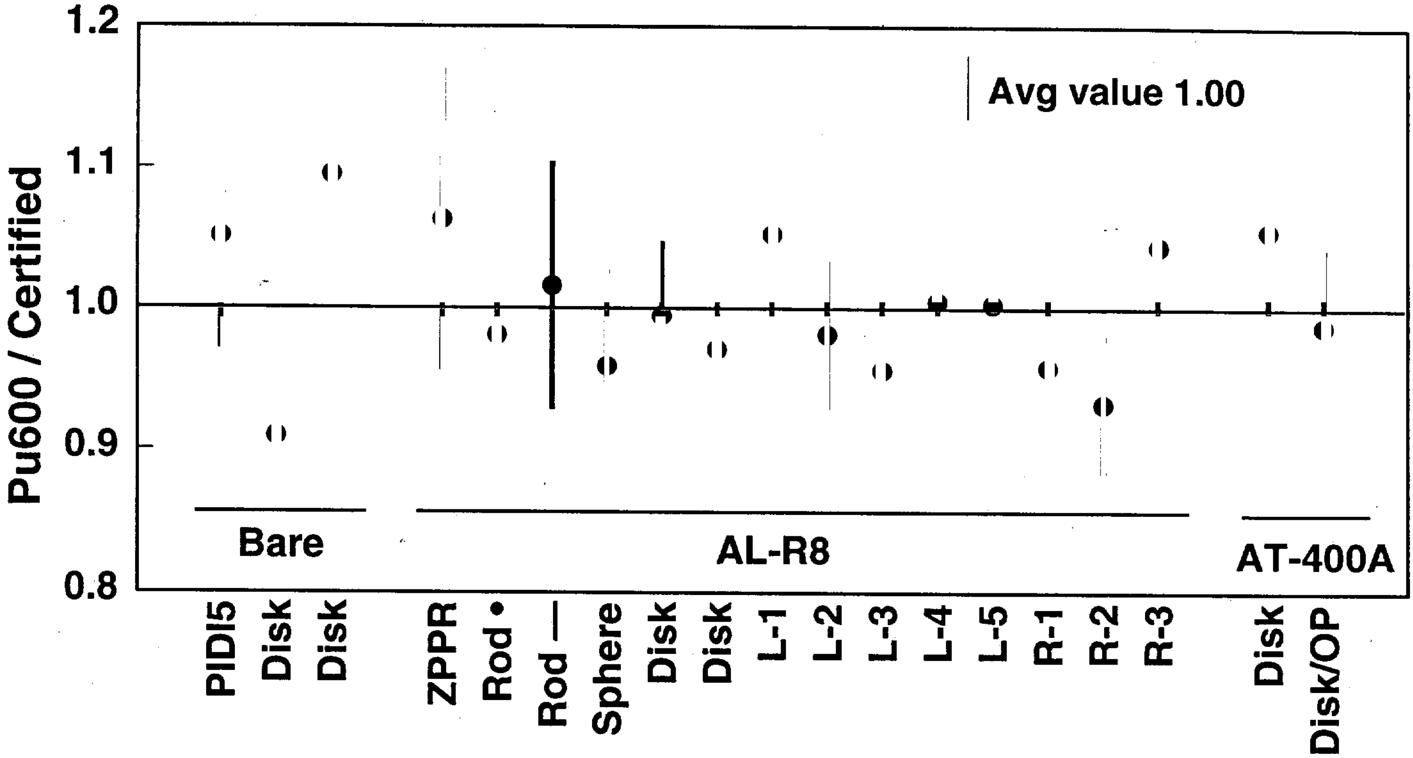

Figure 5. Results of Pu600 for isotopic measurements compared to the certified values.

Pu900 – Absence of Plutonium Oxide

The newest attribute that we have been concerned with is the absence of plutonium oxide in a sample. It is desirable in a weapons dismantlement regime it is necessary the sample under consideration is a metal not an oxide. We made measurements on candidate oxide samples and found that an 870.7 keV line was present in all of the oxide samples and not present in the metal samples. This gamma ray arises from the decay of the first excited state of ^{17}O. We had initially thought that this line arose exclusively from $^{17}O(\alpha,\alpha')$. However, it has become increasingly evident that this line may arise from $^{14}N(\alpha,p)\,^{17}O$. The interplay between these two mechanisms is unknown at this time and is the subject for a research program. However, for the time being it is save to say that the presence of the 870.7 keV indicates that there is plutonium oxide present.

Conclusion

In this paper we have described three methods that determine the presence of plutonium, the age since the last americium separation, the isotopics of the sample and the absence of plutonium oxide. The methods have worked very well under a great deal of measurements. We acknowledge that more work needs to be done in the area of the absence of oxide attribute and that is being pursued. All in all, we believe that we have three relatively stable and very useful techniques for arms dismantlement transparency.

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