Simulated PVT Detector Response with Unshielded/Shielded SNM and Mixed Radionuclides

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DHS HS-STEM Summer Internship Project

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08/01/2013
Introduction

The U.S. Department of Homeland Security (DHS) sponsors a 10 week internship for students majoring in homeland security related science, technology, engineering, and mathematics (HS-STEM) disciplines. As an awardee of the DHS-STEM program, I was given the opportunity to work at Lawrence Livermore National Laboratory (LLNL) under my mentor, Harry Martz. My work at LLNL dealt with official use only or proprietary content. As consequence, I am unable to disclose many project details. Furthermore, the details that I can disclose about the work I performed are limited and I will be using arbitrary units for my results. Throughout the internship period, I met with my mentors and presented my results.

Project

Description:

An anthracene doped polyvinyltoluene (PVT) detector is a plastic scintillator detector, which is an organic polymer that becomes luminescent when irradiated. PVT detectors are cheap, easy to produce, and have a short photon decay time; but, also have lower efficiency and poor resolution. Due to poor energy resolution, PVT detectors have very little photo-peak information and usually rely on the Compton edge feature from Compton scattering for isotope identification. It is of interest to know if it is possible to find a combination of radioisotopes that can match the count rate and energy response of a special nuclear material (SNM) with a PVT detector. SNMs of particular interest include highly-enriched uranium (HEU) and weapons-grade plutonium (WGPu). Aged HEU and WGPu sources were assumed. The initial isotopic composition used for HEU and WGPu is shown in Table 1.
Table 1. Isotopic composition of HEU and WGPu.

<table>
<thead>
<tr>
<th>Isotope</th>
<th>HEU Wt. Percent</th>
<th>Isotope</th>
<th>WGPu Wt. Percent</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-232</td>
<td>$1 \times 10^{-7}%$</td>
<td>Pu-236</td>
<td>$5 \times 10^{-9}%$</td>
</tr>
<tr>
<td>U-234</td>
<td>1%</td>
<td>Pu-238</td>
<td>0.015%</td>
</tr>
<tr>
<td>U-235</td>
<td>93%</td>
<td>Pu-239</td>
<td>93.63%</td>
</tr>
<tr>
<td>U-236</td>
<td>0.445%</td>
<td>Pu-240</td>
<td>6%</td>
</tr>
<tr>
<td>U-238</td>
<td>5.55%</td>
<td>Pu-241</td>
<td>0.355%</td>
</tr>
</tbody>
</table>

Methods:

Multiple radioisotopes were considered for HEU and WGPu. The sources included low energy, high energy, and mixed energy gamma emission sources and neutron emission source Cf-252. Each source was simulated with various levels of shielding consisting of low atomic number (Z), high Z, mixture of low and high Z, and moderator. The shielding levels are outlined below in Table 2.

Table 2. Shielding Specifications for HEU and WGPu.

<table>
<thead>
<tr>
<th>Shielding</th>
<th>Material for HEU</th>
<th>Material for WGPu</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bare (B)</td>
<td>None</td>
<td>None</td>
</tr>
<tr>
<td>White (W)</td>
<td>Thin Medium Z</td>
<td>High Z</td>
</tr>
<tr>
<td>Light Gray (LG)</td>
<td>Low Z + Medium Z</td>
<td>Low Z + Medium Z + Moderator</td>
</tr>
<tr>
<td>Gray (G)</td>
<td>Thick Medium Z</td>
<td>Think Low Z + Medium Z + Moderator</td>
</tr>
<tr>
<td>Dark Gray (DG)</td>
<td>Thick High Z</td>
<td>Thick High Z + Moderator</td>
</tr>
</tbody>
</table>

LLNL software RadSrc was used to compute the photo-peak emission from the continuous decay of HEU or WGPu. The transport code Monte Carlo N-Particle version 6 (MCNP6) was used to simulate and compare HEU or WGPu with the selected sources. Multiple techniques were utilized to find an optimal mixture of selected radionuclides. These techniques included the method of least squares and optimization codes.
Results:

The gamma intensity for the bare sources was found to be $9.480 \times 10^4 \, \gamma/s/g$ for HEU and $1.542 \times 10^8 \, \gamma/s/g$ for WGPu. WGPu also had a neutron emission of $86.45 \, n/s/g$. The effect of the different levels of shielding on the photon emission of HEU is shown if Figure 1.

As seen above in Figure 1, the most prominent photo-peak is seen at 186 keV from the alpha decay of U-235 to Th-231. Another important photo-peak is seen at 2.61 MeV from the beta decay of Tl-208 to Pb-208. Attenuation of this higher energy photo-peak fills in most of the higher energy spectrum seen in Figure 1. The photon emission of WGPu, which includes the contribution from continuous decay and spontaneous fission, with different levels of shielding is shown below in Figure 2.
The prominent photo-peak seen in the WGPu occurs at 59.5 keV from the beta decay of U-237 or alpha decay Am-241 to Np-237. Due to its lower energy, this main photo-peak is significantly attenuated even by a small amount of shielding. The contribution from spontaneous fission products is seen in the smoother sections, which dominate after 1.1 MeV. The neutron emission of WGPu at various shielding levels is shown in Figure 3.
As seen in Figure 3, both the Bare and White shielding spectrum, which lie on top of each other, increase rapidly from low energy up to about 500 keV. On the other hand, the Light Gray, Gray, and Dark Gray shielding include a moderator which moderates the higher energy neutrons and results in a larger amount of low energy neutrons. After about 500 keV, the energy spectra for all shielding levels decrease roughly linearly on the logarithmic scale.

PVT detector response for HEU is shown in Figure 4 for Bare shielding and in Figure 5 for Gray shielding.

![PVT Detector Response for HEU in Bare Shielding](image)

**Figure 4.** Comparison of HEU and selected sources in Bare shielding.
It is apparent from Figures 4 and 5 that there are significant differences in the energy spectrum between HEU and the selected radionuclides. This causes the shielding to have a different effect on HEU and the selected sources, which is seen when comparing Figures 4 and 5. For example, going from Bare to Gray shielding, the low energy source is attenuated much greater than HEU. On the other hand, the optimized combination source for HEU provided a reasonably close comparison with HEU for all tested levels of shielding as seen in Figures 4 and 5. The difference between HEU and the selected sources for all shielding cases is shown in Figure 6.
A radioisotope can be calibrated to a specified activity to yield an equivalent gross rate as HEU for a particular level of shielding as is clearly shown in Figure 6. However, due to the difference in the energy spectra, this level of activity will not provide a good match when looking at other levels of shielding. For example, the low, mid, and mixed energy sources yielded equivalent gross counts to HEU in the Bare shielding; however, once shielding is introduced, the low energy source quickly becomes much less than HEU and the mid and mixed energy sources become much greater than HEU. In contrast, the HEU optimal combination source yielded similar gross counts over all shielding cases.

PVT detector response for WGPu in Bare and Gray shielding is shown in Figures 7 and 8 respectively.
Just like the results for HEU, there are significant differences between the energy spectrum of WGPu and the radionuclide sources as shown in Figures 7 and 8. The optimized combination source for WGPu provides a similar spectrum compared to WGPu. The difference between WGPu and the selected sources is outlined in Figure 9.
It is shown in Figure 9 that no radionuclide source alone could provide a good match across all shielding levels. Conversely, the optimal combination source provides comparable gross counts to WGPu across all shielding levels.

**Experience**

My time at LLNL has been a rewarding experience and has benefited me greatly. Throughout the internship, I have gained invaluable professional work experience. I was given the opportunity to apply my hard earned knowledge and skills that I have gained throughout my college education to help solve an important real-world problem of national concern. My work has furthered my applied knowledge, growth, and value as a nuclear engineer. I was also able to attend multiple lectures where I have developed a more well-rounded understanding of the various threats to the United States and the defenses against these threats. This internship has also given me the opportunity to make valuable networking connections with not only my mentors, but also with fellow students.
My experience at LLNL has strongly impacted my future academic and career plans. Before the internship, I was undecided on whether I wanted to go into industry or research. I enjoy contributing to important real-world problems such as global security. I find it a rewarding and meaningful experience when my hard work can make a difference. I now believe that I could have a successful and rewarding career at the national laboratories undertaking problems of national concern. This has impacted my academic plans as well. I have been committed to pursuing a Master’s degree in Nuclear Engineering, but was undecided about continuing on to a Doctorate degree. After consulting with my mentors, I am now leaning towards pursuing a Doctorate degree.

**Conclusion**

By the end of the internship, I had accomplished all the main goals that my mentors desired from me. I was able to successfully investigate the effect that various shielding had on HEU, WGPu, and multiple radionuclides; and derive optimal radionuclide combinations that can provide matching count rate and energy response for HEU or WGPu with a PVT detector. I have found contributing to important national problems to be a rewarding experience and can see myself pursuing a successful and fulfilling career at the national laboratories.