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Warhead Monitoring by Environmental Activation Products Detection

P. Marleau*[1] and M. Shinner[2]

[1]*Sandia National Laboratories, Livermore, CA 94551*

*pmarlea@sandia.gov

[2]*University of California Berkeley, Department of Physics*

Abstract

Through a series of measurements with a high purity germanium detector, we have established that the past presence of neutron emitting material can be detected by the decay of activation products in aluminum containers, tungsten shielding, and concrete floors even several days after last exposure. The time since last exposure can be estimated by the gamma-ray detection rate. These findings may lead to interesting new concept of operations (CONOPS) in the detection of illicit special nuclear material (SNM) or the verification of the absence (or presence) of SNM containing objects in facilities and/or transit even after the material has been removed.

In the work presented here, we measure the products of neutron activation in materials that may be commonly found in environments near SNM, whether in storage or transport. Neutrons emitted by an SNM containing item interact with and activate certain specific isotopes that may be present nearby. Through a combination of inelastic reactions, these isotopes are transformed into metastable products (having half-lives on the order of 10s of minutes to 10s of hours) imprinting a record of the presence of SNM in the recent past.

As the metastable isotopes decay, they emit gamma-rays with energies characteristic of the material type. The decay gamma rays can then be detected hours or days after the SNM has been removed, verifying its past presence even though the SNM may now be quite some distance away. Further, the relative detection rates of these characteristic decay gamma-rays can be used to estimate the time since last neutron exposure (i.e. when the SNM was moved).

This detection method may find application in which the presence or absence of SNM containing items must be verified. For example, containers and other objects within a facility, such as the floor itself may be used to confirm declarations that treaty accountable items have not been stored in empty containers or storage areas within some period of time. Additionally, the detection of activated shielding material such as tungsten may be an indication that some effort is being taken to hide the presence of SNM containing items.

1. NUCLEAR MATERIAL DETECTION CONCEPT OF OPERATIONS

In this work, we explore the potential of a different concept of operations (CONOPS) for confirmation or detection of the presence of special nuclear material (SNM). In this CONOPS, the surrounding environment acts as a detector as neutrons emitted by the SNM interact with and "activate" certain specific isotopes that may be present nearby. Through a combination of (n,g), (n,p), or (n,α) reactions, these isotopes are transformed into metastable products (having half-lives on the order of 10s of minutes to 10s of hours) imprinting a record of the presence of SNM in the recent past.

As these metastable isotopes decay, they emit gamma-rays with energies characteristic of the material type. The decay gamma rays can then be detected hours or days after the SNM has been removed, verifying its past presence even though the SNM may now be quite some distance away. Further, if more than one activation product (having a different decay constant) is detected, then the relative detection rates of their characteristic decay gamma-rays can be used to estimate the time since last neutron exposure (i.e. when the SNM was moved).

Here we seek to measure the products of neutron activation in materials that may be commonly found in environments near SNM, whether in storage or transport. A survey of candidate isotopes was made with the following criteria:

1. They must have a significant natural abundance.
2. They must have a reasonable neutron cross section (order of millibarns or higher).
3. The reaction process must create a metastable product with a half-life between 10s of minutes and days.
4. The decay of the metastable isotope must result in a detectable gamma ray.

A list of candidate isotopes meeting these criteria can be found in Table 1. Many of these isotopes are commonly found in construction materials such as concrete and rebar, containers, and radiation shielding.

The proposed detection method may also find application in which the presence or absence of SNM containing items must be verified. For example, containers and other objects within a facility, such as the floor itself may be used to confirm declarations that treaty accountable items have not been stored in empty containers or storage areas within some period of time. Additionally, the detection of activated shielding material such as tungsten may be an indication that some effort is being taken to hide the presence of SNM.

Table 1 – List of candidate isotopes with metastable neutron activation products.

Isotope	Natural Abundance (%)	Activation Process	Cross-Section (average over fission or thermal spectrum from ENDF (1)) (barns)	Product	Product Half-life (hours)	Gamma Energies (keV)
Na-23	100	(n,g)	0.53	Na-24	15.0	1368 2754
Al-27	100	(n,α)	0.9 (mb)	Na-24	15.0	1368 2754
Al-27	100	(n,g)	0.23	Al-28	0.05	1779
Al-27	100	(n,p)	0.9 (mb)	Mg-27	0.16	844 1014
K-41	6.7	(n,g)	1.48	K-42	12.3	1525
Mn-55	100	(n,g)	13.3	Mn-56	2.6	847 1811 2113
Fe-56	91.8	(n,p)	1.6 (mb)	Mn-56	2.6	847 1811 2113
Ni-64	0.93	(n,g)	1.5	Ni-65	2.52	1116 1482
W-186	28.4	(n,g)	37.5	W-187	23.7	480 618 686 773

We will explore these scenarios in a series of laboratory measurements described in Section 2. Research questions probed include the following:

1. Can it be determined whether a container has been exposed to neutrons in the recent past as indicated by the presence of activation products?
2. Can it be determined whether a floor has been exposed to neutrons in the recent past as indicated by the presence of activation products?
3. Can it be determined whether shielding material is present as indicated by the presence of activation products?
4. If so, then what is the maximum time since last exposure that is detectable?
5. If so, can the length of time since last exposure be determined?

2. EXPERIMENTAL SET-UP

Three sets of measurements were made to investigate the research questions posed in Section 1:

1. Measurements made at the surface of an aluminum shipping and storage container (H-Gear) after exposure to a neutron source.

2. Measurements made at the surface of shielding material (tungsten) after exposure to a neutron source.
3. Measurements made on the floor after exposure to a neutron source.

In all cases, a Cf-252 fission neutron source with an emission rate of ~2e5 n/s moderated by a 2cm thick shell of high density polyethylene (HDPE) was placed at the center of an aluminum H1333 Shipping and Storage Container (H-Gear) described in (2) (~60 cm off the floor) along with two 1" x 4" x 8" tungsten shields as illustrated in Figure 1. After a specified length of exposure time, the neutron source was removed from the area before measurements were made with a Canberra High Purity Germanium (HPGe) Cryo-pulse 5-plus spectrometer (3).

Measurements at the surface of the H-Gear container were made near an iron bolt as shown in Figure 1 and Figure 2 (left) in an attempt to gain sensitivity to the detection of both aluminum and iron activation. Measurements of the tungsten shield were made up against its outer surface as shown in Figure 2 (center). To facilitate multiple simultaneous measurements, the tungsten bricks were removed from the H-Gear and placed on a patch of floor several feet away after exposure as shown in Figure 9. Finally, measurements of the floor were made after the H-Gear was moved. The germanium detector was positioned as close to the center of the patch of floor under the H-Gear as possible as shown in Figure 2 (right).

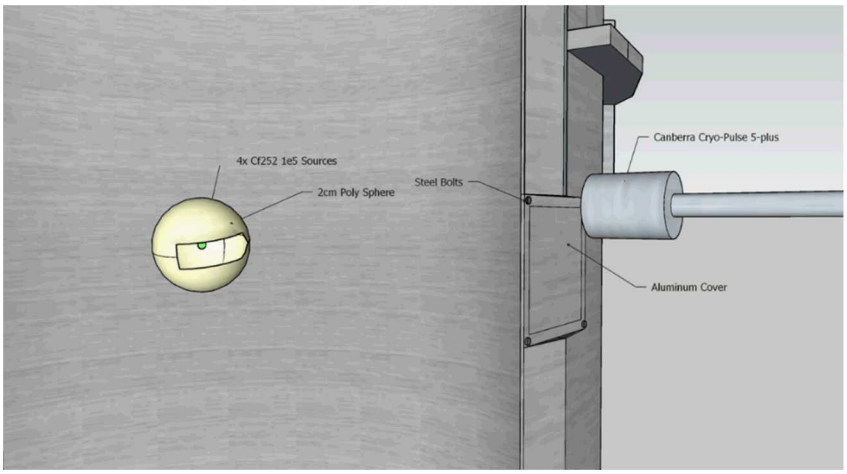

Figure 1 – Illustration of the HDPE moderated Cf-252 source positioned inside the H-Gear

Figure 2 – Photographs of the H-Gear and the HPGe detector set up with (center) and without (left) a tungsten shield and measurements on the floor underneath the H-Gear (right). The yellow tape marks indicate the position of the H-Gear during neutron exposure.

To get an idea for the detector count rates (which drive the minimum measurement times), that we might expect, an MCNP (4) simulation was performed. A 1e5 n/s neutron point source with a Cf-252 energy spectrum was simulated at the center of a 2cm thick shell of HDPE suspended 60 cm above a 100cm x 100cm x 10 cm thick concrete slab. We used the "regular concrete" composition from the Compendium of Material Composition Data for Radiation Transport Modeling (5). After the equivalent of 20 hours of irradiation, we tallied the gamma-ray interactions within modeled HPGe detector at various energies from various activation products. The results shown in Figure 3 indicate that we should expect on the order of hundreds of counts per hour. Primarily, these will originate from the decay of Na-24.

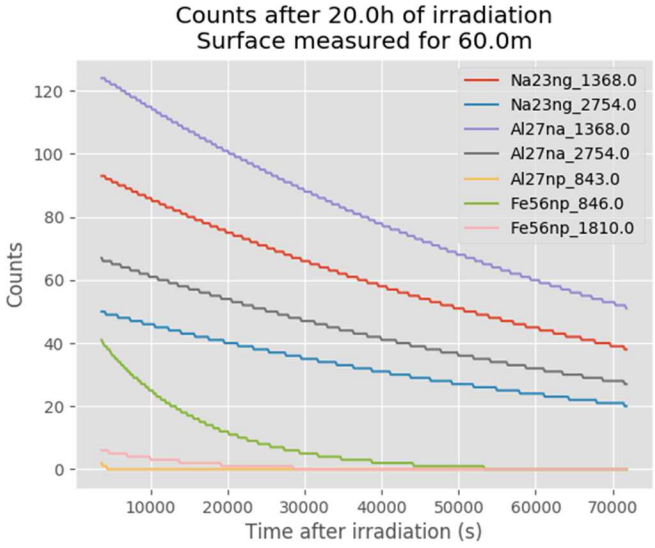

Figure 3 – Results of an MCNP simulation of the decay gamma-ray rates from activation products in a 100x100x10 cm3 slab of concrete after a 20 hour exposure to a moderated 1e5 Cf-252 neutron source 60 cm away.

3. RESULTS

3.1. Measurements at the surface of H-Gear

In this first set of measurements, we exposed the H-Gear container to the moderated Cf-252 source for approximately 20 hours. Once the source was removed, a series of data sets were collected with the face of the HPGe detector close against the side of the H-Gear container: six 10-minute runs followed by three 60-minute runs. The detector was positioned near a steel bolt to gain some sensitivity to iron activation processes as shown in Figure 2 (left). A 1-hour background data set was also collected the day before neutron exposure. The data analysis program PeakEasy (6) was then used to identify and fit excess peaks that were not present in the background. Figure 4 identifies 1778 keV decay gamma-rays at a rate of 1.9 ± 0.6 g/minute. These are most likely from the decay Al-28 produced by neutron capture on Al-27. Having a half-life of only 2.8 minutes, this peak quickly loses strength. However, it was still detectable in the second hour of measurement.

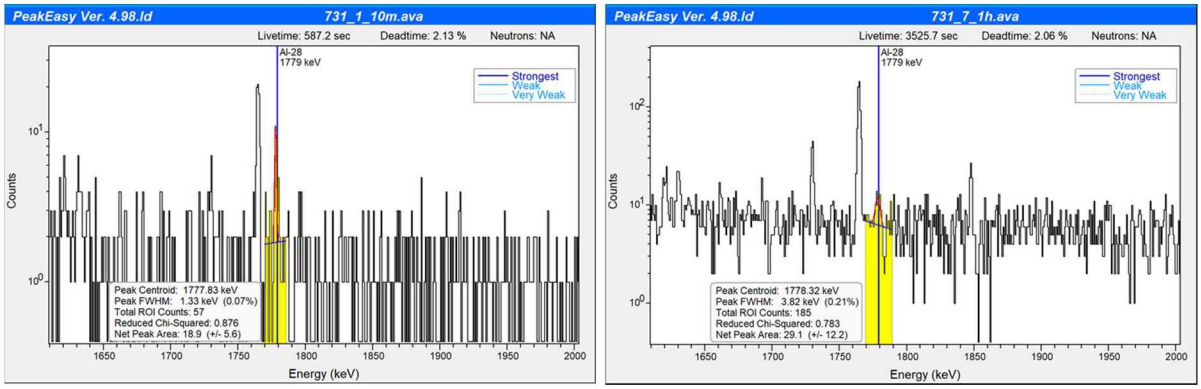

Figure 4 – PeakEasy display showing regions of ^{28}Al decay gamma energies integrated 0-10 minutes (left) and 60-120 minutes (right) after last neutron exposure.

We also investigated energy regions characteristic of the decay of Mg-27 and Mn-56 from (n,p) reactions on Al-27 and Fe-56 respectively. The results shown in Figure 5 are marginal for Mg-27 with a net peak area average of 1.7 ± 0.8 gammas/minute at 1014 keV in the first 10 minutes after neutron exposure and Mn-56 with a net peak area average of 1.3 ± 0.4 gammas/minute at 847 keV in the first hour after neutron exposure. The 847 keV region could not be reliably fit in the first 10 minutes.

However, the decay of Na-24 from (n,α) activation of Al-27 is much more easily detected with a net peak area of 66.5 ± 19.1 gamma rays at 1369 keV and 33.0 ± 6.2 gamma-rays at 2754 keV in the first hour after last neutron exposure as seen in Figure 6.

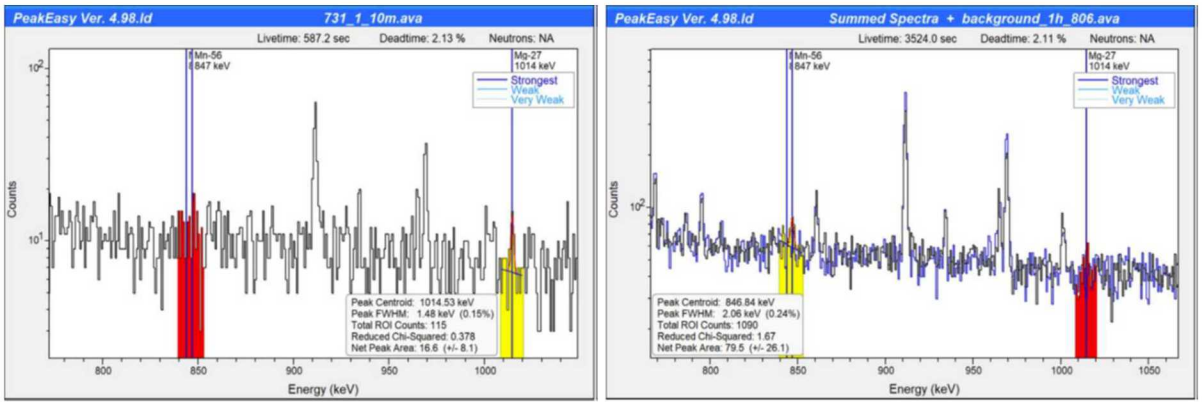

Figure 5 – PeakEasy display showing region of ^{27}Mg and ^{56}Mn decay integrated 0-10 minutes (left) and 0-60 minutes (right) after last neutron exposure.

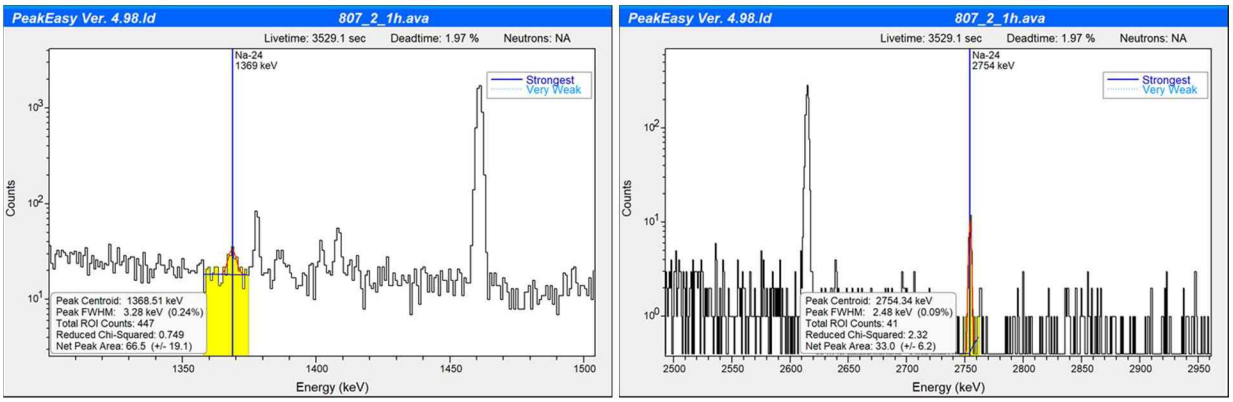

Figure 6 – PeakEasy display showing the 1368 keV (left) and 2754 keV regions of ^{24}Na decay integrated from 0-60 minutes after last neutron exposure.

The moderated Cf-252 source was then placed back into the H-Gear container with tungsten shield and left for a 1-week exposure. A second set of measurements were taken over a period of two days after last exposure. The net peak areas in the 1369 keV and 2754 keV regions were estimated as a function of time since last neutron exposure. The data is presented in Table 2 and the sum of the two energy regions is plotted in Figure 7 as a function of time since last neutron exposure. The x-axis error bars represent the duration of the measurements and the red line is an exponential with the expected half-life from Table 1 and a scaling factor fit to the measured data. It can be seen that the strength of this signal decays as expected.

Table 2 – Peak areas of estimates from measurements at the surface of the H-Gear in the regions of 1369 keV and 2754 keV as a function of time since last neutron exposure.

Start (mins)	Stop (mins)	Peak Area (1369 keV)	Uncertainty (1369 keV)	Peak Area (2754 keV)	Uncertainty (2754 keV)
5	125	52.5	19.5	24	7.5
1332	1452	7.5	18.3	8	5.7
2776	2986	0	0	7	7

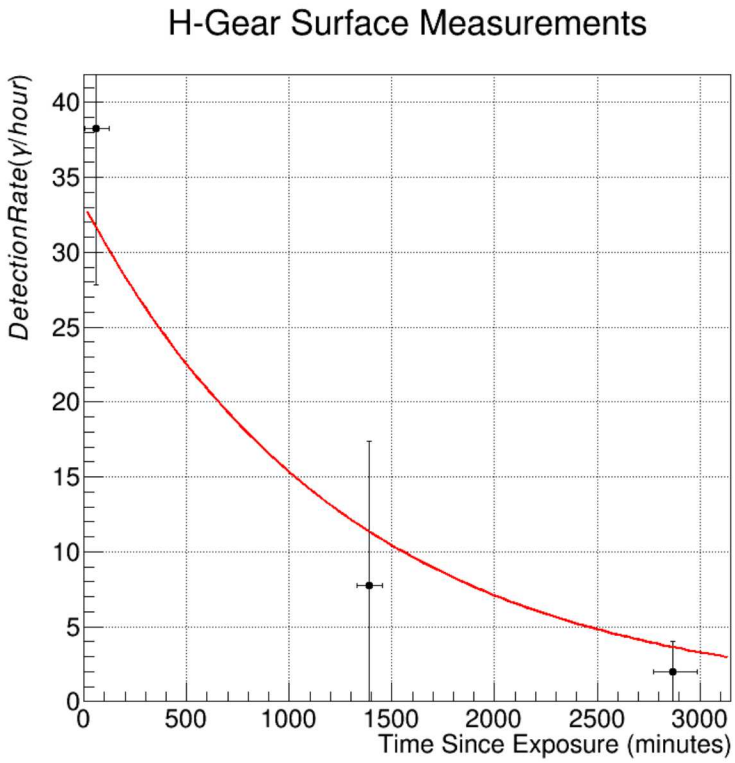

Figure 7 – Sum of net gamma-ray counts per hour in the 1368 keV and 2754 keV regions as a function of time since last neutron exposure.

Finally, to explore the possibility of estimating the time since last neutron exposure from a single measurement of the H-Gear surface (assuming one knows the starting detection rate, i.e. the functional scaling factor), we use the detection rate uncertainty of the second two hour measurement to estimate the time as 1888.0 minutes (the true time is 1388 minutes). Unfortunately, the statistical uncertainty is too high for this to be a significant detection. However, a limit can be placed that the time since last exposure has been at least 841 minutes. Statistically significant estimates are not feasible with 2-hour measurements with the detector that was used in this study for times since last exposure greater than approximately 16 hours. We estimate that a ~6 hour measurement would be required to achieve significant detection 24 hours after last neutron exposure.

3.2. Tungsten Shielding Measurements

The second set of measurements investigate whether the presence of tungsten shielding can be detected after exposure to neutrons. Following the same 20-hour exposure as described in Section 3.1, preliminary data was collected with the HPGe detector face in close proximity to the outer surface of two 1" x 4" x 8" tungsten plates. Figure 8 shows the energy spectrum in regions expected from the decay of W-187 integrated for the first 70 minutes after last neutron exposure. Peaks at the expected energies are quite strong with an average detection rate greater than 25 gamma rays per minute. Even characteristic x-rays from tungsten were easily detectable as shown in Figure 8 (right). Given our experimental arrangement, the presence of tungsten would be quite easy to detect.

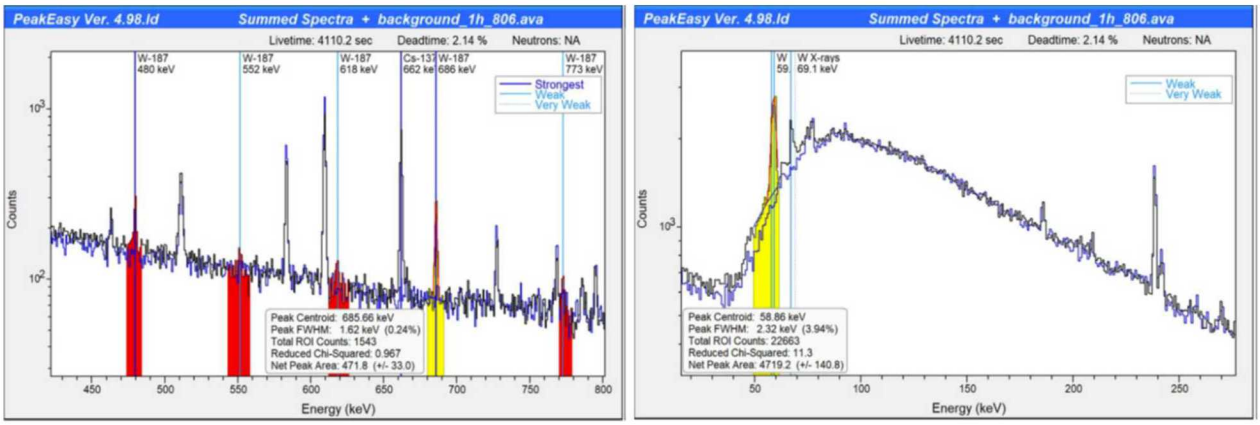

Figure 8 – PeakEasy display of the energy regions expected from the decay of W-187 in the 400-800 keV region (left) and low energy x-ray region (right).

After these preliminary measurements, the moderated Cf-252 source was then placed back into the H-Gear container with tungsten shield and left for a 1-week exposure. After exposure, the neutron source was removed, and the tungsten shield was placed on the floor about 10 feet from the H-Gear as shown in Figure 9. A second set of measurements were made over a period of 3.5 days after last exposure. The net peak areas at 480 keV, 552 keV, 618 keV, and 686 keV were estimated as a function of time since last neutron exposure. The data is presented in Table 3 and the sum of the all energy regions is plotted in Figure 10 as a function of time since last neutron exposure. The x-axis error bars represent the duration of the measurements and the red line is an exponential with the expected half-life from Table 1 and a scaling factor fit to the measured data. The expected half-life matches the data quite well.

Finally, to explore the possibility of estimating the time since last neutron exposure from a single measurement (assuming the starting detection rate, i.e. the functional scaling factor, is known), we use the detection rate uncertainty of the second two hour measurement to estimate the time as 1672 minutes with an uncertainty of + 184 minutes and – 169 minutes. This is accurate to the true time since last exposure of 1574 minutes within uncertainties. Significant detection of the presence of tungsten was achieved as long as 3.5 days after last neutron exposure.

Figure 9 – Photograph of measurements made of the tungsten shielding. After neutron exposure, the shielding was placed on the floor approximately 6 feet from the H-Gear and measurements were made with the face of the detector in close proximity to the outer surface of the shielding.

Table 3 - Peak areas of estimates from measurements at the surface of the tungsten shield in the regions of 480 keV, 618 keV, 686 keV, and 773 keV as a function of time since last neutron exposure.

Start (mins)	Stop (mins)	Peak Area (480 keV)	Uncertainty (480 keV)	Peak Area (618 keV)	Uncertainty (618 keV)	Peak Area (686 keV)	Uncertainty (686 keV)	Peak Area (773 keV)	Uncertainty (773 keV)
136	196	271.1	36.1	114.2	26.4	430.8	29.8	118.8	23.7
1516	1636	295.3	48.8	82.3	36.5	350.4	36.8	167.1	28.3
3014	3134	187	75.6	88.5	57.7	248.9	36.3	0	0
4255	4375	78.2	33.3	18.03	49.3	134.1	41.4	0	0
4379	4739	230.3	63	0	0	199.9	51.5	290.8	48.4

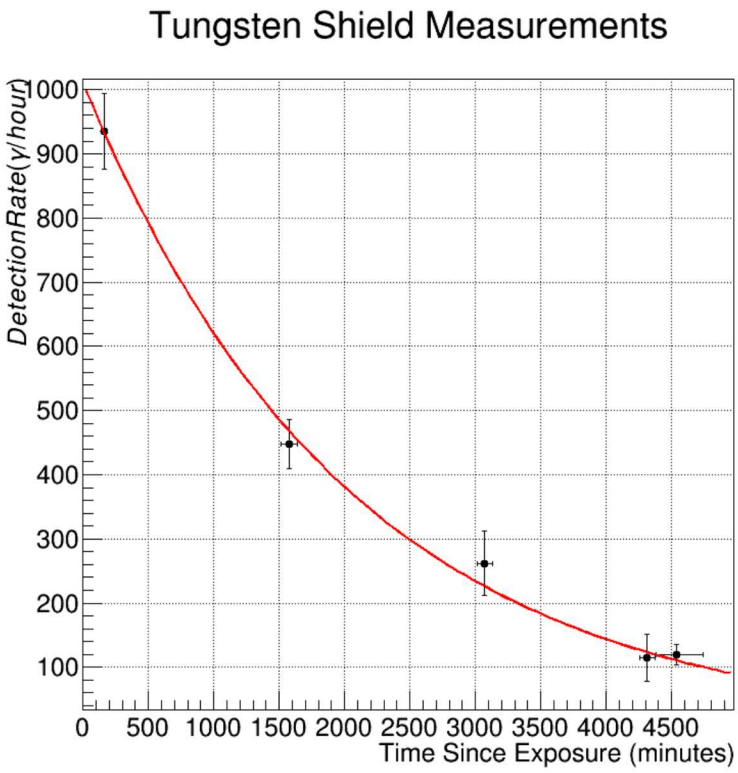

Figure 10 – Sum of net gamma-ray counts per hour in the 480 keV, 618 keV, 686 keV, and 773 keV regions as a function of time since last neutron exposure. The exponential curve has the half-life expected for the decay of W-187 with a fit to the scaling factor.

3.3. Floor Measurements

In the third set of measurements, after the H-Gear was exposed to the moderated Cf-252 source for 12 hours, the source was removed, the H-Gear was moved, and a series of data sets were collected with the the HPGe detector on the floor centered on the patch of floor that was directly below the source as shown in Figure 2 (right). The data analysis program PeakEasy was then used to identify and fit excess peaks that were not present in the background. Figure 11 (left) identifies 1369 keV decay gamma rays at a rate of 0.6 ± 0.3 g/minute averaged over the first hour primarily caused by neutron capture on Na-23 and (n,α) on Al-27. Figure 11 (right) identifies 2754 keV decay gamma rays at a rate of 0.6 ± 0.1 g/minute averaged over the first hour.

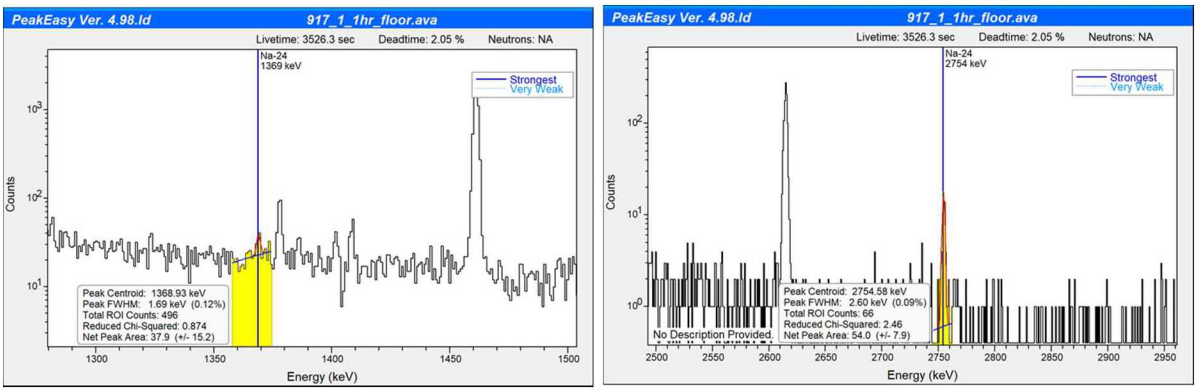

Figure 11 – PeakEasy display of the energy regions expected from the decay of Na-24 at 1369 keV (left) and 2754 keV (right).

The moderated Cf-252 source was then placed back into the H-Gear container with tungsten shield and left for a 1-week exposure. After these preliminary measurements, a second set of measurements were made over a period of 3.5 days after last exposure. The net peak areas at 1369 keV and 2754 keV were estimated as a function of time since last neutron exposure. The data is presented in Table 4 and the sum of the all energy regions is plotted in Figure 12 as a function of time since last neutron exposure. The x-axis error bars represent the duration of the measurements and the red line is an exponential with the expected half-life from Table 1 and a scaling factor fit to the measured data. Again, the expected half-life matches the data quite well.

To explore the possibility of estimating the time since last neutron exposure from a single measurement (assuming the starting detection rate, i.e. the functional scaling factor, is known), we use the detection rate uncertainty of the second three-hour measurement to estimate the time as 1909 minutes with an uncertainty of + 436 minutes and – 326 minutes. This is consistent with the true time since last exposure of 1758 minutes within uncertainties.

Two weeks after the last exposure, the moderated Cf-252 source was then placed back into the H-Gear container with tungsten shield and left for another 1-week exposure. A third set of measurements were made to fill in shorter times since last neutron exposure. The data is presented in Table 5 and the sum of the all energy regions is also plotted in Figure 12 as a function of time since last neutron exposure. These results are consistent with the previous measurement as well as the half-life of Na-24.

Table 4 - Peak areas of estimates from the first set of measurements on the floor in the regions of 1369 keV and 2754 keV as a function of time since last neutron exposure.

Start (mins)	Stop (mins)	Peak Area (1369 keV)	Uncertainty (1369 keV)	Peak Area (2754 keV)	Uncertainty (2754 keV)
198	318	135	27.2	106	11.2
1654	1894	70	35.7	60	10
3079	3439	12	42.7	40	10

Table 5 - Peak areas of estimates from the second set of measurements on the floor in the regions of 1369 keV and 2754 keV as a function of time since last neutron exposure

Start (mins)	Stop (mins)	Peak Area (1369 keV)	Uncertainty (1369 keV)	Peak Area (2754 keV)	Uncertainty (2754 keV)
0	120	122	26.0	123	12.1
120	360	257	36.8	199	16.6
360	600	166	37.1	209	15.3

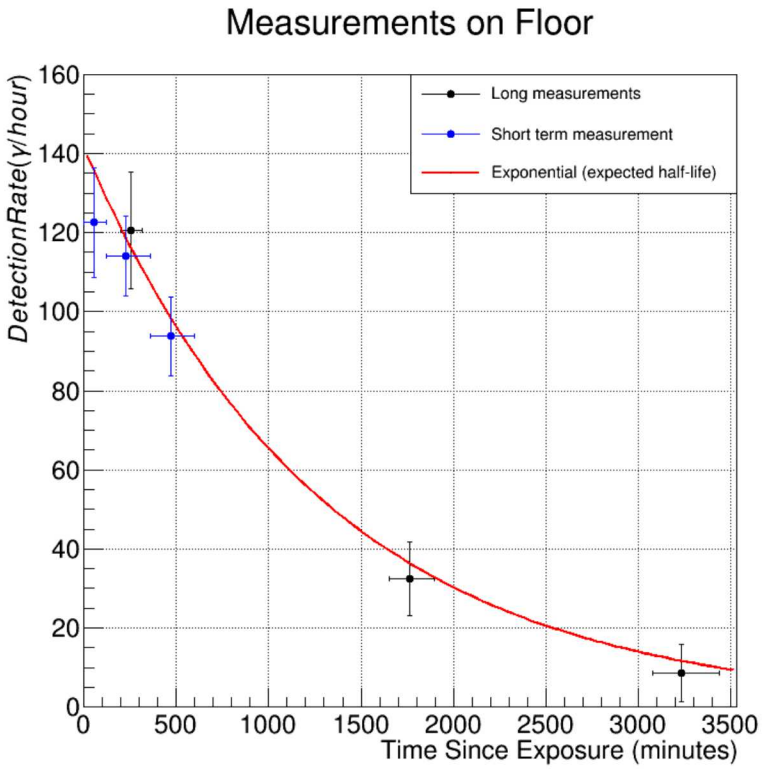

Figure 12 – Sum of net gamma-ray counts per hour in the regions of 1368 keV and 2754 keV as a function of time since last neutron exposure. The black points were taken as course, long term measurements while the blue were short measurements within the first 12 hours after last neutron exposure. The exponential curve has the half-life expected for the decay of Na-24 with a fit to the scaling factor.

4. CONCLUSIONS

Through a series of measurements, we have established that the past presence of neutron emitting material can be detected and confirmed by the decay of activation products in aluminum containers, tungsten shielding, and concrete floors. We have shown that both the floor under a storage container and tungsten shielding have detectable levels of metastable product decay gamma rays at least 3 days after exposure to a 2e5 n/s fission source.

Further, with an assumption about the source strength and quantity of activated material present (e.g. thickness of shielding or concrete), the time since last exposure can be estimated to within 3 hours from a 2-hour measurement on tungsten and 5 hours with 6 hour measurement on the floor even after more than 24 hours have passed.

Unfortunately, despite the fact that the H-Gear container weighs approximately 400 lbs, the decay gamma-ray detection rate was roughly a factor of three lower than that found on the floor. This is most likely because of the contribution of Na-23(n,g)Na-24 activation in the concrete which has an activation cross section approximately 500 times higher than that of Al-27(n,a)Na-24 activation in the container walls as shown in Table 1. Nevertheless, we have determined that a 2-hour measurement against the container wall is enough to detect the past presence of our 2e5 n/s fission source as long as 16 hours after exposure.

These findings may lead to interesting new CONOPS in the detection of illicit SNM or the verification of the absence (or presence) of SNM containing objects in facilities and/or transit even after the material has been removed. Detection of activated containers might be used to verify that they have recently stored SNM. Detection of activated shielding might be used to detect attempts to hide SNM. And detection of activated patches of floor might be used to verify that SNM was present and also possibly how many SNM containing objects were there (with the detection of multiple “hot spots”).

5. ACKNOWLEDGEMENTS

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6. REFERENCES

1. **Group, Cross Section Evaluation Working.** *ENDF/B-VI Summary Documentation.* National Nuclear Data Center, Brookhaven National Laboratory. Upton, NY, USA : s.n., 1991. IAEA-NDS-61, Rev.3.
2. **L.A. Brown, M.C. Higuera.** *Weapon Container Catalog Volumes 1 & 2.* Sandia National Laboratories. 1998. SAND97-8017.
3. **Canberra.** Cryo-Pulse 5-plus Data Sheet. [Online] http://www.canberra.com/products/detectors/pdf/Cryo-Pulse_5_plus_C0514.pdf.
4. **Laboratory, Los Alamos Scientific.** *MCNP: a General Monte Carlo Code for Neutron and Photon Transport.* Group X-6, Los Alamos National Laboratory. 1979.

5. **McConn, R.J., Gesh, C.J., Pagh, R.T., Rucker, R.A., Williams, R.G.** *Compendium of Material Composition Data for Radiation Transport Modeling.* Pacific Northwest National Laboratory. 2011. PNNL-15870 Rev.1.
6. **Rooney, B., Felsher, P.** PeakEasy Home Page. [Online] https://peakeasy.lanl.gov/.