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In-situ Alpha Detector for Wet Soil

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Abstract

In some areas of historical nuclear site, the soil was contaminated with alpha sources. Before any field work, the soil has to be surveyed using alpha detector. However, if the soil is wet, the alpha radiation is almost completely attenuated by the water film. The current detectors used in field survey measure the alpha particles from a short distance, so it cannot detect the alpha particles when the soil is wet. As a consequence, the field work has to be hold on, and wait the soil to dry out. The objective of this work is to develop an instrument which could provide in-situ alpha radiation measurement for wet soil. Some experiments had been carried out in our lab, and are discussed in this paper.

1. Introduction

In some areas in the historical nuclear site, the soil has been found to be contaminated with Alpha sources. In the field, surveyors use a square-head alpha counter to scan the soil to determine the contamination. However, this technique only works for dry soil. If the soil is wet, the alpha radiation is almost completely attenuated by the water film. If it has rained, or the soil is wet from dew, the current field detection technique does not work since the alpha counter cannot detect the radiation. Hence, the field work has to wait the soil to be dried out. The objective of this task was to develop an instrument which can provide in-situ alpha radiation measurement for wet soil, so that the field work is not affected by rain or dew.

Historically, silver-activated zinc sulphide (ZnS[Ag]) scintillator has played a key role in early experiments for alpha particle interactions because of its very high scintillation efficiency. As powder is easier to be spread on top of wet soil, ZnS[Ag] powder was our major target scintillator. Other scintillators were also used in our experiment, such as yttrium aluminum perovskite crystal doped with cerium (YAP[Ce]) [1] and linear alkylbenzene (LAB) gel scintillator.

This paper summarizes the experiments that have been carried out in our lab and discusses the path forward in developing a prototype alpha detector for wet soil.

2. Experiments Using Pu-239 Alpha Source

It was convenient to use a known source as the starting point to test the proposed method. A Plutonium-239 (Pu-239) alpha source was used for the experiments. Pu-239's principal mode of

decay is alpha decay with a half-life of 24110 years. This alpha source comprised a Pu-239 film deposited on a metal disk. The activity of the source was 200 Bq.

2.1 Experiment Setup

Both the ZnS[Ag] and the YAP[Ce] scintillator are able to detect alpha particles and were examined in this experiment. The block diagram of the experimental configuration is shown in Figure 1. The light emitted from the scintillator is collected with a Hamamatsu PMT assembly, and is converted to electrical pulses through this PMT and fed to a pre-amplifier. The pulses were integrated in the pre-amplifier, from which the output signal was fed into the linear amplifier, where the pulses were shaped and amplified. A multiple channel analyzer (MCA) was used to read the output pulses from the linear amplifier and generate the spectrum. A computer was used to log the data from MCA.

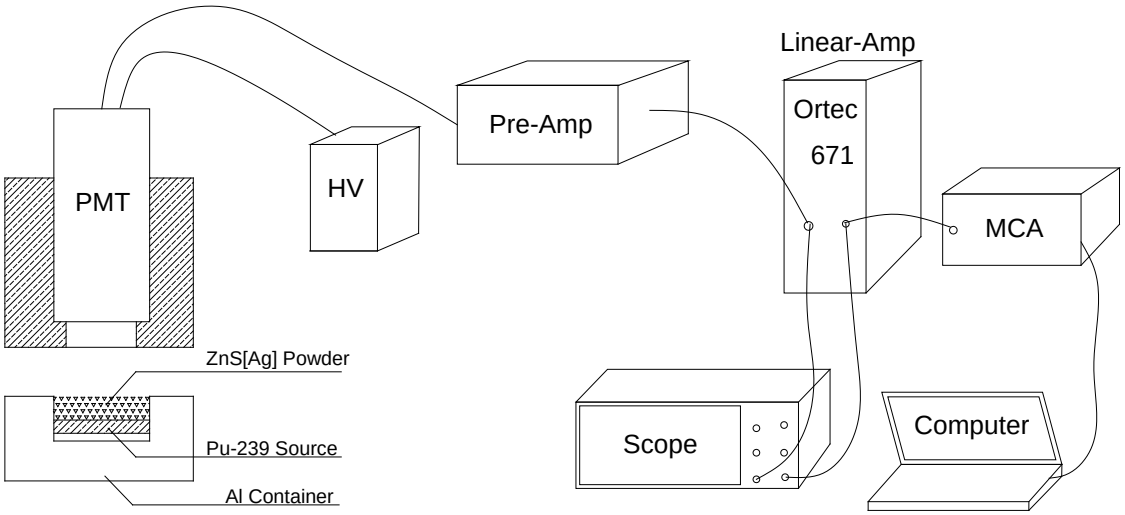

Figure 1 Block diagram of the experimental configuration for alpha detection

The assembly of the PMT and the bottom container is shown in Figure 2. The PMT was enclosed in an aluminum casing, and the PMT window can be seen in the figure. As seen in Figure 2 (a), the Pu-239 alpha source was located at the bottom of the container, and the YAP[Ce] crystal was placed on top of the Pu-239 source. As shown in Figure 2 (b), some ZnS[Ag] powder was distributed on top of the Pu-239 source disk. The bottom container was then filled with water, and the ZnS[Ag] scintillator powder gets distributed on the Pu-239 source. The experiments were carried out in the soil lab at Chalk River Laboratories.

2.2 Experiment Results

First, various background spectra were checked with or without the YAP[Ce] scintillator and ZnS[Ag] scintillator presented. Four major background-spectrums are illustrated in Figure 3. In overall, the background pulses are located at fairly low energy channels, does not affect the measurement in the high energy range.

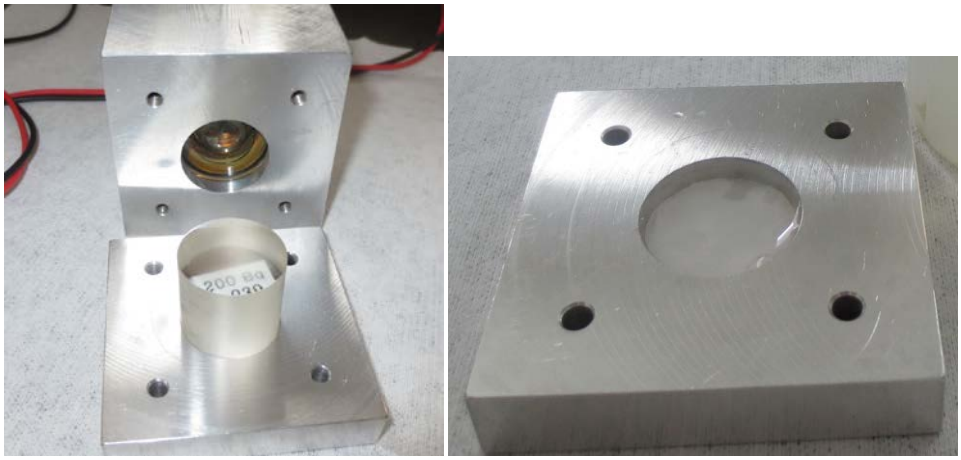

Figure 2 PMT and the Bottom Container with Pu-239 Source in the Container
a) YAP Scintillator on top of the source; b) ZnS[Ag] on top of the source

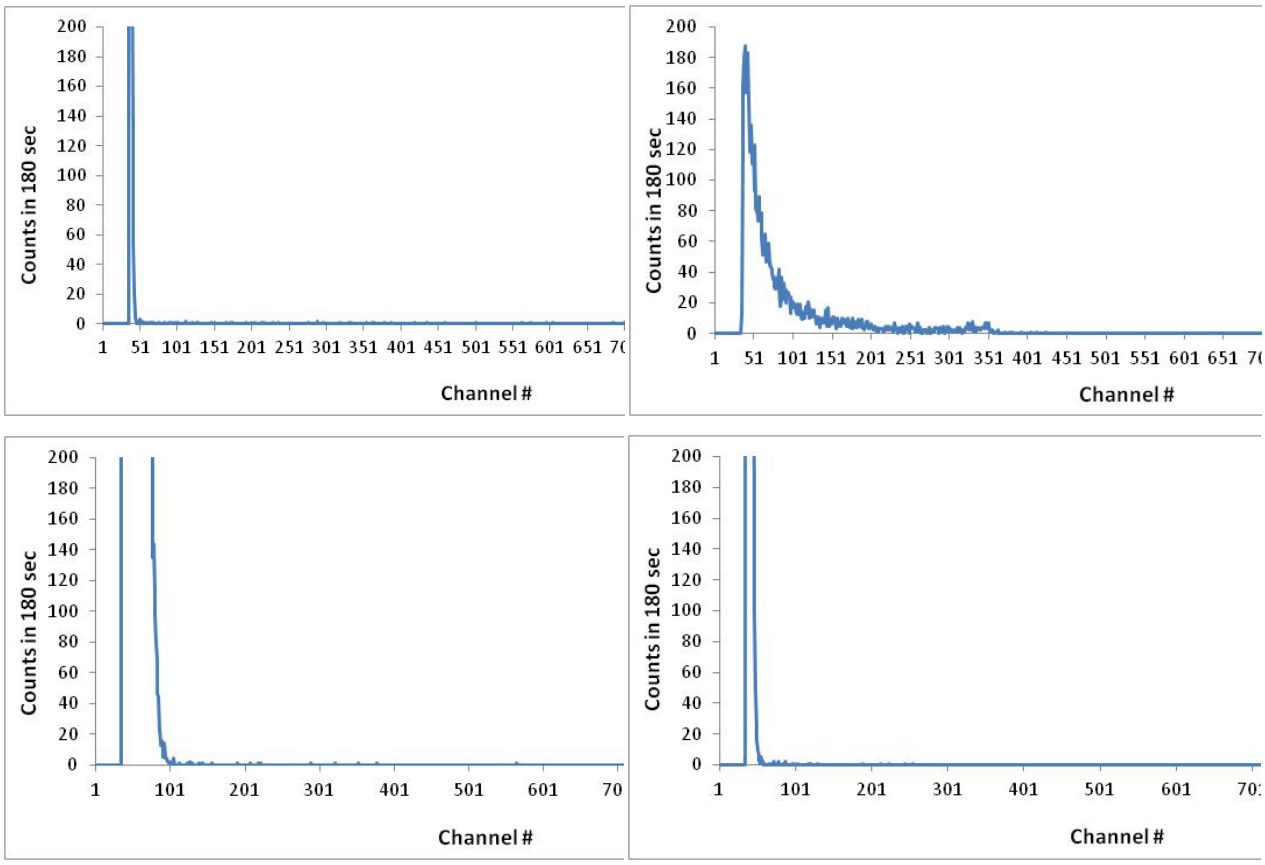

Figure 3 Background spectrum checks. a) Pu-239 source without scintillator; b) YAP[Ce] without Pu-239 source; c) ZnS[Ag] dry powder without source; d) ZnS[Ag] wet powder without source;

It worth to note that the background from ZnS[Ag] powder has huge counts at low energy band in Figure 3 c) and d). That is because ZnS[Ag] is sensitive to alpha, beta, and X-rays. The ambient air contains some radioactive particles, such as radon. These particles interact with the ZnS[Ag] and generate background photons.

After the background check, both scintillators were used to count the activity of the Pu-239 source. Since the ZnS[Ag] was in the form of powder, various thicknesses were tested. YAP[Ce] scintillator was a cylindrical crystal, so only one spectrum was taken for this scintillator. However, due to the form-factor of the YAP[Ce] crystal, it is not easy to make sufficient contact between the crystal surface and the soil particles. Also, the YAP[Ce] crystal was larger in volume, so it is also sensitive to gamma, which made the separation of alpha particles and gamma photons difficult. Hence, the focus of this experiment was to test ZnS[Ag] powder.

All the spectra collected in this report were from the MCA with 180 seconds counting (3 minutes).

2.2.1 Results from YAP Crystal

With the YAP[Ce] scintillator sitting on the top of the Pu-239 source, as shown in Figure 2 (a), the spectrum is shown in Figure 4. There is a clear peak between channel 350 to channel 600, which was caused by the alpha source. The total number of counts in this region was 15419 over a period of 180 seconds, which equals 86 counts/s. The PU-239 source had an activity of 200 Bq, which counts emissions from both sides of the source. However, only emissions from one side can reach the crystal. With 100% efficiency, the count rate should be 100 counts per second. Hence the counting efficiency of the YAP[Ce] is about 86/100 = 86%.

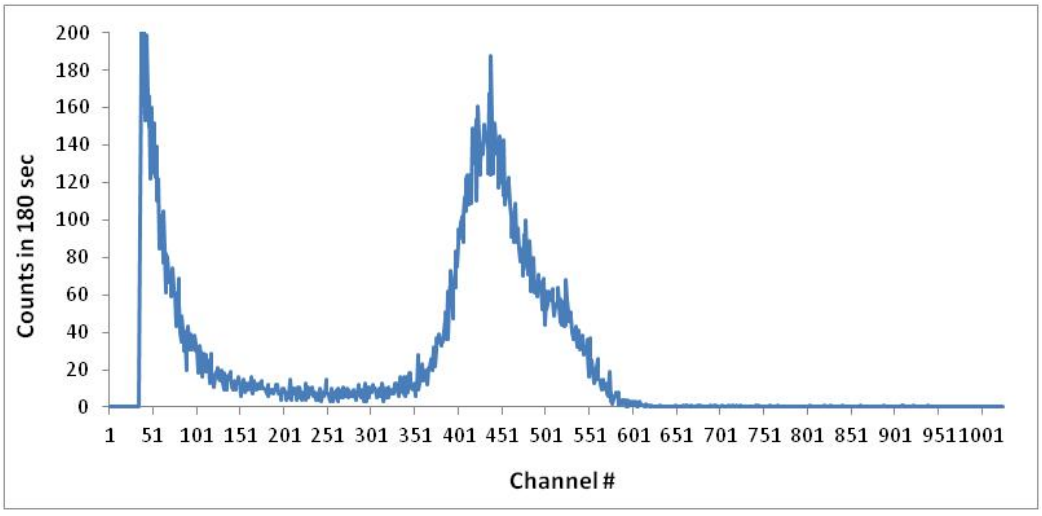

Figure 4 Pulse height spectrum from the YAP[Ce] with Pu-239 alpha source

2.2.2 Results from ZnS[Ag] Powder

The ZnS[Ag] powder is mixed with water before it is applied on the top of the Pu-239 source. Therefore, a layer of water always existed on top of the Pu-239 source. This simulates the case of wet soil. ZnS[Ag] emits light when interacting with alpha particles, but it also absorbs some of the light it emits. If the ZnS[Ag] powder is too thin, there is not enough powder to cover the whole surface of the Pu-239 source. However, if the ZnS[Ag] powder is too thick, some of the light will be self-absorbed. So various thicknesses of ZnS[Ag] powder were tested and their spectra are shown in Figure 5 a) to d).

With the first spray of ZnS[Ag] powder (mixed with water), only a very thin layer of powder was deposited on the Pu-239 source. The very thin layer of powder was somewhat transparent and the color of the Pu-239 source disk was still discernible. After the second spray, the powder layer fully covered the surface of the Pu-239 disk. This case is referred to as "thin" in this report. The case after third spray is referred to as thick and the case after fourth spray is referred to as very thick.

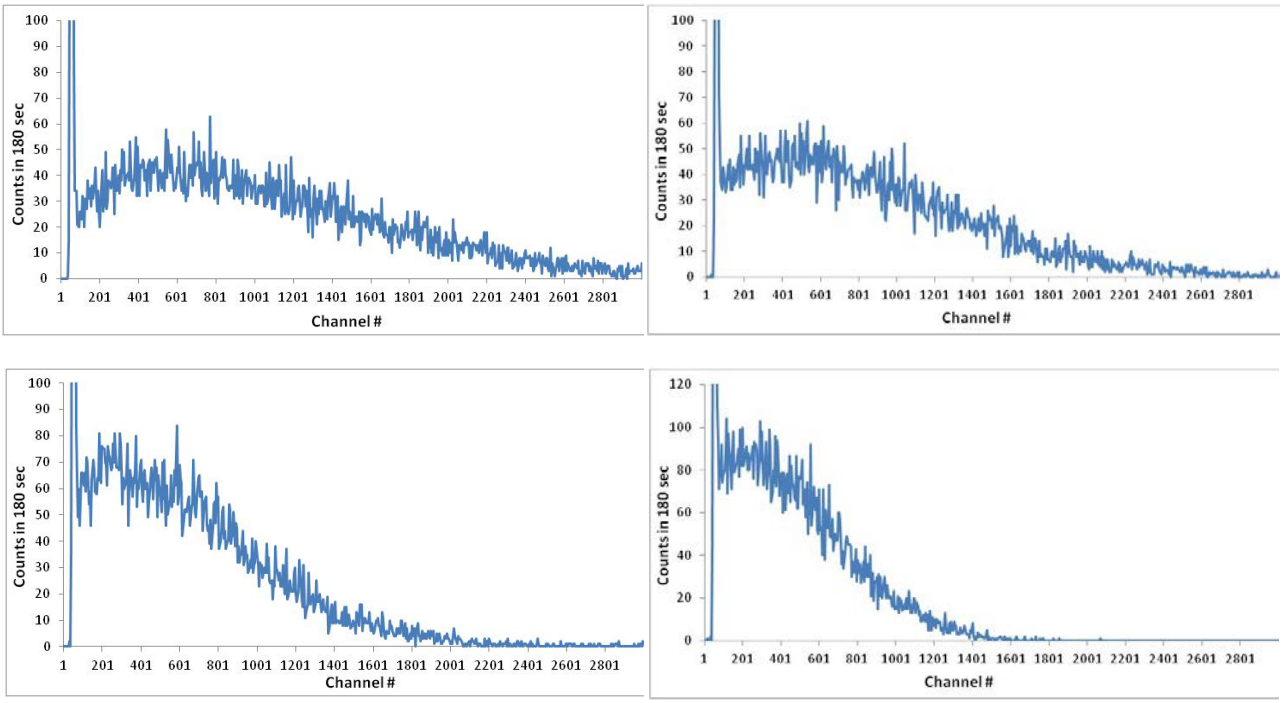

Figure 5 Pulse height spectrum of the ZnS[Ag] wet powder with Pu-239 source. a) very thin ZnS[Ag] powder; b) thin ZnS[Ag] powder; c) thick ZnS[Ag] powder; d) very thick ZnS[Ag] powder;

The spectrum did not change significantly when the powder thickness was increased from very thin to thin. However, as the power layer got thicker, the spectrum's shape did change. The main peak shifted to lower energy. This is because the thicker ZnS[Ag] powder absorbs more light, so less photons reach the PMT. Hence, the individual pulse height tends to lower.

The background spectrum stopped at channel #106. The region of interest (ROI) was defined from 106 to the maximum channel #. The count numbers in ROI are plotted in Figure 6. The counts increased slightly from very thin to thin, and then started to decrease. However, the change was within 15% of the maximum counts.

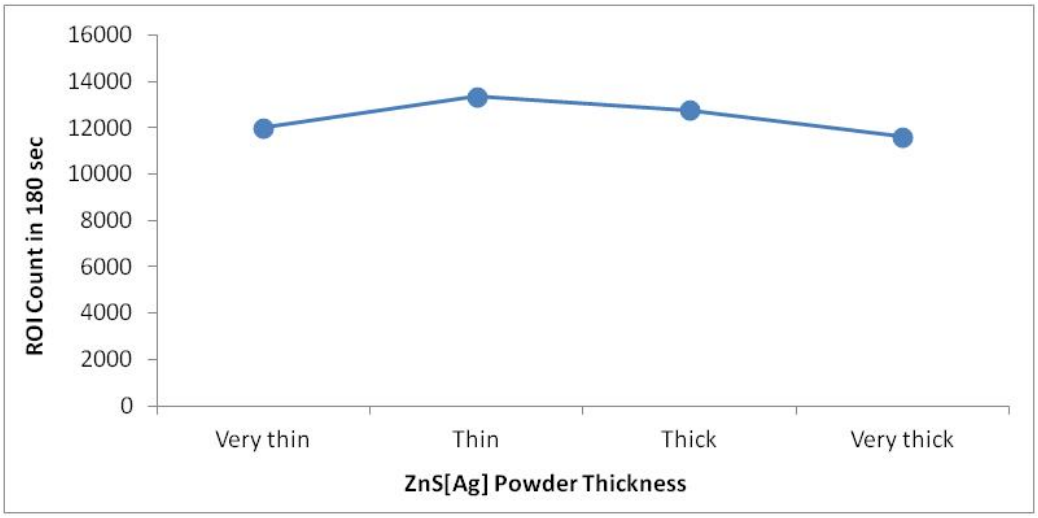

Figure 6 ROI counts vs. ZnS[Ag] powder thickness with Pu-239 alpha source

The maximum count rate of the ZnS[Ag] was 13346 counts in 180 seconds, or 74 counts per second. Thus the counting efficiency of the powder was 74/100 = 74%.

3. Experiments with Contaminated Soil

The soil sample was taken from the east swamp in waste management area, and this sample has been analyzed by direct gamma spectrum and the spectrum from the chemical extraction of the soil. The analysis results are documented in Appendix A and Appendix B.

The analysis shows that 1 gram of the dry soil has 100 Bq of gross alpha, 1100 Bq of gross beta, 105 Bq of Co-60 gamma, and 102 Bq of Cs-137 gamma. Therefore, the experiment results should consider how to distinguish between these types of radiation.

The soil sample is shown in Figure 7 (a) on the left and the small container shown in Figure 7 (b) contains the soil sprayed with the water and ZnS[Ag] powder mixture.

Figure 7 Soil sample. a) in bottle (left); b) mixed with ZnS[Ag] powder (right)

3.1 Experiment Setup

The experiment configuration was basically the same as the configuration with Pu-239. The one difference is that a larger size container was used, which is shown in Figure 7. Since the soil was black, which caused it to absorb a significant amount of the emitted light, the PMT high voltage was adjusted to -1000 volts from -700 volts to obtain higher gain.

3.2 Experiment Results

It was observed that the ZnS[Ag] generates low energy light pulses from the ambient radioactive particles. The background spectrum of ZnS[Ag] wet powder was measured first. Then the soil sample was sprayed with the mixture of water and ZnS[Ag] powder. It took about 2 minutes to have the powder deposit to the bottom of the water. Six spectra were taken after that. It was observed that the spectrum changed with time. More and more pulses of higher energy were observed, so the gain of the linear amplifier was lowered from 10x1.0 (coarse gain 10, and fine gain 1.0) to 5x1.0, and lowered down to 5x0.2 at the end.

To facilitate comparison of the spectra, the channel numbers were extended at lower linear-amp gain to ensure the same channel # represented the same energy in each spectrum.

3.2.1 Background Checking

The background stops at channel # 600 according to the background spectrum in Figure 8 a). Hence, the ROI in the spectra is defined as Channel #600 to maximum channel #.

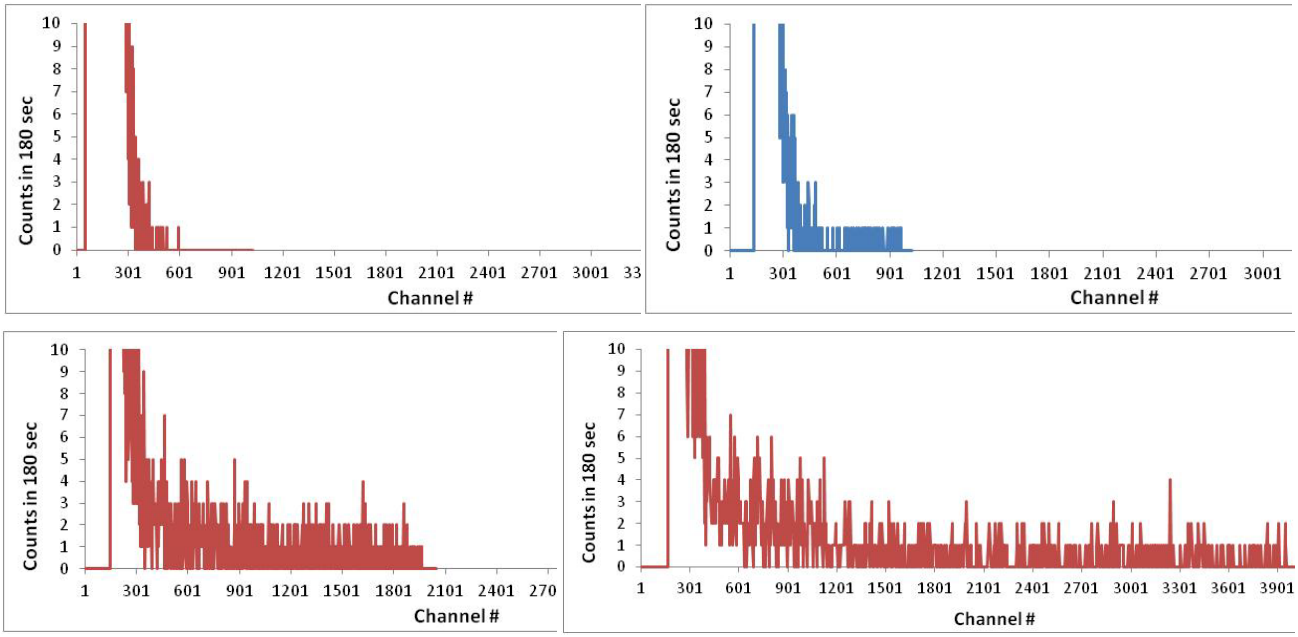

Figure 8 Pulse height spectrum from soil sample mixed with ZnS[Ag] powder.
a) background (without soil); b) 2 min after mixing; c) 14 min after mixing; d) 25 min after mixing;

3.2.2 Results from the Soil Sprayed with ZnS[Ag] Powder

Six spectra were taken after the ZnS[Ag] powder was deposited on the surface of the soil. Only three of them are shown in Figure 8 b) to d).

Clearly, a lot more pulses were present compared to the background spectrum, and the distribution of these pulses extends to higher energies with time.

The soil sample used in the experiment contained alpha, beta and gamma radiation. All of them could possibly generate the scintillation pulses. Compared to alpha and beta particles, the gamma photons can penetrate the water easily. Considering the small amount of the ZnS[Ag] powder, the sensitivity of ZnS[Ag] to gamma photons should be extremely low. Also if the signal pulses were generated from gamma photons, the time-dependent effect should not happen. Therefore, the signal pulses were not caused by the gamma photons.

Both alpha and beta particles only travel limited distance in water. Alpha particles can travel about 37 μm in water with 5 MeV energy [2], which is about the alpha decay energy of Am-241. While beta particles could travel longer distance in the range of 174 μm for 0.5 MeV beta particles [3], which is the beta decay energy of Strontium-90.

The counts in the ROIs of the six spectra are plotted in Figure 9. The explanation for the time-dependent effect is that the ZnS[Ag] particles take time to get close enough to the soil particle surface for the alpha or beta particles to reach them. In time, more and more ZnS[Ag] particles reach the effective range of the alpha or beta particles, so the energy received from the alpha or beta particle increases with time.

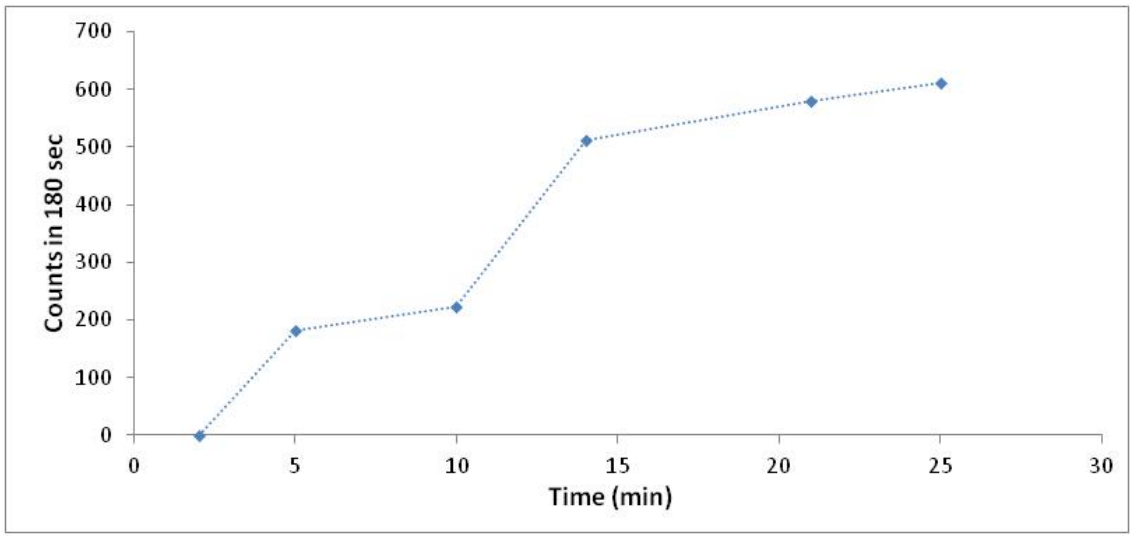

Figure 9 ROI counts vs. time

There is no conclusive evidence thus far as to whether these signal pulses are from alpha or beta particles. Further experiments with pure alpha and beta contaminated soil will provide better data. The pulse height and shape will be monitored to distinguish the alpha and beta particles.

3.2.3 Observation from gel scintillator

Some tests using gel scintillator were also carried out in the lab. However, no significant pulses were observed so far. The soil sample was mixed with the gel scintillator. The major reason could be that the light yield in the gel scintillator is low. Therefore, the signal pulses are masked by the noise. Further testing is expected for using gel scintillator in the wet-soil detector.

4. Summary

The experiments using Pu-239 alpha source demonstrated that both the ZnS[Ag] powder scintillator and the YAP[Ce] scintillator crystal are capable of detecting the alpha particles efficiently without water present. The light yields from both scintillators are strong. With Pu-239 source covered by the mixture of ZnS[Ag] powder and water, and the alpha particles were detected with an efficiency of 74%. However, the YAP[Ce] was not tested in this case.

A soil sample was taken from the east swamp in the waste management area. This sample contains alpha, beta and gamma sources. With the ZnS[Ag] scintillator applied on top of the soil sample, the radioactive particles could be detected. It is clear that the gamma photons did not generate significant signal from the experiment results, which showed time dependant spectrum change. The number of pulses at higher energies increased with time. The explanation for this phenomenon is that the ZnS[Ag] powder takes time to get close to the soil particles. The alpha and beta particles

can only travel limited distance in water. If the ZnS[Ag] particles are not sufficiently close to the soil, the alpha and beta emissions from the soil could not be detected.

The work for 2013/2014 fiscal year is to perform further experiments to distinguish the alpha and the beta particles in soil sample using pulse shape or energy discrimination, and develop a prototype instrument for field operation.

5. Acknowledgements

The experiments were carried out in Doug Kelly's soil lab, which is located in B513 at Chalk River Laboratories. Doug provided great help in setting up the experiment and taking the gamma spectrum of the soil. The soil sample was supplied by Jenny Zhou, who works in Analytical Chemistry Branch of AECL. Jenny also provided the analysis report of the soil sample as our reference.

Ghaouti Bentoumi designed the experiment apparatus and had the apparatus manufactured in machine shop. Ghaouti Bentoumi, Shuwei Yue and Liqian Li worked as a team, carried out the experiments.

6. References

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