Using a Germanium detector to measure contaminant substances in mud

Jade Salisbury

10867885

Department of Physics and Astronomy

University of Manchester

Second year laboratory report

April 2023

This experiment was performed in collaboration with Samuel Novak

Abstract

Using a germanium semiconductor diode gamma-ray detector, the proportion of a contaminant (identified to be ^{137}Cs was discovered and measured in two separate dirt samples from Sellafield, Cumbria. After determining its activity, the proportion and subsequent mass of the contaminant in the samples was identified to be $(1.63 \pm 0.15) \times 10^{-14}$kg in the first and $(8.08 \pm 1.87) \times 10^{-15}$kg in the second. This is in the correct order of magnitude if the 22TBq ($\approx$ 6.9g) [1] of ^{137}Cs released from the Windscale fire is assumed to be mostly distributed within a few kilometers of the nuclear plant.

[1] J. R. Cooper, K. Randle, and R. S. Sokhi, *Radioactive Releases in the Environment : Impact and Assessment*. John Wiley and Sons, Inc, 2003.

1 Introduction

Germanium detectors are a type of semiconductor detector which became popular in the middle of the 20th century after their creation and the improved ability to grow sufficiently sized germanium crystals. They must be cooled with liquid nitrogen (or other means) to effectively detect gamma-rays at a high resolution, as the experiment requires. Most rocks and soil contain trace amounts of heavy radioactive elements with long half-lives such as ^{238}U and ^{232}Th, which slowly decay into more stable isotopes. Identifying the contaminant can be done by examining the spectrum from samples and finding energies which don't belong to any daughter nuclei at their measured intensities.

In October 1957, the two-pile Windscale (now Sellafield) site in northwest England caught fire, causing a nuclear accident. In the aftermath of the fire over 22TBq of ^{137}Cs [1] (among many other radioactive substances) was released into the atmosphere and ground around Sellafield. Some mud from around Sellafield was retrieved for this experiment and the main goal is to identify the mass of contaminant ^{137}Cs in the mud from its gamma ray emissions.

2 Theory

There are four important ways in which gamma rays (photons) interact with matter, but only three of them are relevant this experiment: through the photo-electric effect, Compton scattering and pair production. All three of these interactions cause the production of a high energy electron which can be detected by the Germanium detector. The fourth interaction, called photodisintegration is not detectable detectable by semiconductor diode detectors as it does result in these high speed electrons.

The photoelectric effect is the emission of electrons when electromagnetic radiation hits a material. It was proposed by Einstein to be the result of light waves actually being made up from light quanta called photons [2]. One incoming photon can only "knock out" one electron from the material, and so the energy of the photo-electron is entirely decided by the energy of the photon which caused it to be emitted. This is governed by the equation

$$K_{max} = h\nu - W, \tag{1}$$

where K_{max} is the maximum kinetic energy of the photo-electron, h is Planck's constant, ν is the frequency of the incoming light and W is the work function. The work function is the minimum required energy to remove an electron from the surface of the material. K_{max} is defined as the maximum kinetic energy as it is possible for the incident photon to entirely miss a surface electron and go some distance into the material before it causes an emission, in such cases, the electron would lose some kinetic energy on its journey to the surface of the material, where it would finally be emitted. It should be noted that if an inner electron does not have sufficient

energy to escape the material after an interaction with a photon, it will stop before it reaches the surface and will not be emitted. The photoelectric effect occurs in the range of energies of a few eV to a few keV and has a probability of occurring proportional to Z^n where Z is the atomic number and n varies between 4 and 5 [3].

The second way gamma rays interact with matter is through Compton scattering. Gamma-rays are again treated like photons as they are incident on a particle. Both the photon and the particle experience a change in angle, as an elastic collision, which assumes the photon has momentum, takes place. The scattered gamma-ray has an energy equal to

$$E' = \frac{h\nu}{1 + \frac{h\nu}{m_e c^2}(1 - \cos\theta)} \tag{2}$$

where E' is the new energy, m_e is the particle mass (typically an electron), c is the speed of light, θ is the scattering angle and h and ν are as before [4]. The electron recoils at an increased velocity which depends on the momentum (energy) of the photon from the scattering interaction. This phenomena occurs at photon energies around 511keV, if the electron is to gain any decent speed. The probability of Compton scattering per atom is proportional to Z, the atomic mass [3]. Compton scattering at lower energies is called Thomson scattering.

The final light-matter interaction is pair-production. At high gamma-ray energies, pair production becomes possible (first at ≈ 1 MeV). This is when a neutral boson (photon) creates a particle and anti-particle pair in its place. As the electron has the lowest mass of the elementary particles, it requires the lowest energy photons and is more likely to happen randomly. This experiment only concerns the production of electrons (and by extent positrons) through this process. Pair-production must also occur near a nucleus in order to conserve both energy and momentum. If a photon has an energy above the combined mass energy of the produced particles, it has an increased probability to occur. However, since the photon is fully destroyed, this excess of energy cannot be removed from the universe, and so is distributed to the kinetic energy of the produced particles, giving us even more high velocity electrons that can be detected by the electrodes in the Germanium detector. The probability of pair-production per nucleus varies approximately as the square of the absorber atomic number (Z^2) [3]. Additionally, the positrons created in the same process quickly annihilate creating an influx of new gamma-rays at 511keV, which are also picked up by the detector.

3 Experimental approach

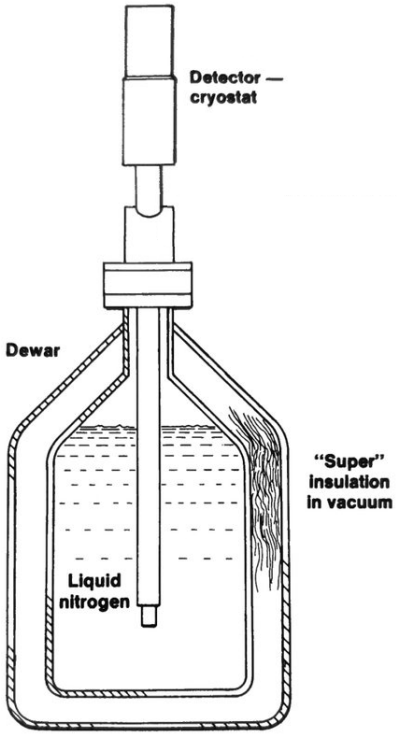

Figure 1: The Germanium semiconductor detector used in this experiment. This is a modification of an image found in [3] with the cryostat pointing upwards.

The Germanium detector in this experiment is a type of semiconductor diode detector. Semiconductor detectors work via the detection of high speed electrons in the depletion layer of a semiconducter material which each produce a number of electron-hole pairs proportional to the energy of the incident gamma-ray. [3] A number of electrons are transferred from the valence band to the conduction band of the semiconductor, allowing them and their holes to flow freely towards electrodes in an electric field resulting in detectable pulse. As the energy required to make an electron-hole pair is known, the energy of the incident gamma ray can be calculated.

Semiconductors make such good solid-state detectors since their band gap is a small 0.7eV, meaning they have an exceptionally high resolution as a small difference in radiation energy will create a unique number of electron-hole pairs. This small energy gap, however, means that the detector cannot be operated at room temperature as too many thermally excited electrons would jump into the valence band. This is why the detector is cooled to 77K with liquid nitrogen as seen in figure 1.

The experiment consisted of three main segments. The first two of the segments involve calibration and measurement of relative efficiencies of the Germanium detector, and will be fully detailed in this section

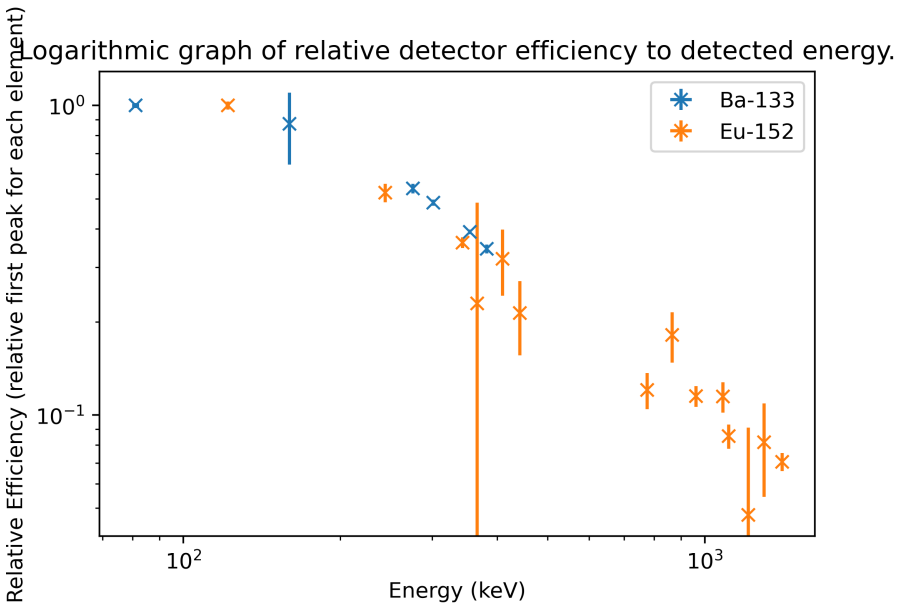

Figure 2: Efficiencies relative to the lowest energy peak (i.e. the lowest energy has a value of 1, as it is the most efficient), with associated errors.

To calibrate the detector, several samples which have known gammma-ray emissions (known as peaks, as there is an increased number of detections at these energies compared to the low-level background noise) at given energies were used. A measurement of over 300 seconds can be taken with the sample above the detector pictured in figure 1 to allow it to receive energy readings for both the background and the decays from the sample. Starting with the highest intensity peaks (peaks with more detections than any other) the energies of each peak can be given assigned a value in keV. This method relies on none of the peaks associated with background emissions being higher than any of the peaks from the sample. Therefore, a background reading for the same amount of time should be performed and removed from the MAESTRO (or any other multichannel analysing software) viewing window before examining peaks. The samples initially used for calibration were ^{137}Cs, ^{60}Co, ^{22}Na and ^{241}Am, which could then be used to measure the peaks on a sample of ^{152}Eu. The spectrum of this was saved and the identified peak energies also noted so that they could be used on any other day to reconstruct the calibration.

Before taking measurements of the mud samples some measurements of ^{152}Eu and ^{133}Ba were taken, and using the relative expected intensities information provided by binders in the lab rooms, the relative efficiency of the detector could be calculated, as it varies with the incident energy, i.e. the effectiveness of the detector at detecting certain energy gamma rays. Plotting relative efficiency against energy gave figure 2.[1]

[1]This follows closely the same graph for a germanium detector found in [3].

The sources of error in this experiment are as a result of several factors, all of which are exhibited during and combined after measurement, before being quantified by the software in the form of error on the net area, which is to say the error on the number of counts at a particular energy. Some of these sources include: leakage current, which is a result of themally excited electrons and reduced at lower operation temperatures; background noise, as the experiment was performed in a lab alongside many other nuclear lab experiments which involve the handling of numerous other radioactive isotopes which our detector could pick up gamma-rays from; cosmic ray noise, which can be included in the background but may fluctuate more. Additionally, due to the statistical nature of the generation of photons through the phenomena described in section 2 (i.e. not always guaranteed), there will always be some low-level noise associated with all experiments similar to this one. These errors on net area for the final part of the experiment are the same sources as those used to calculate error on efficiency shown in figure 2.

4 Results

Sample 1 (keV)	Sample 2 (keV)	Present in Background?
238.50	238.50	Yes
294.79	293.31	Yes
351.08	351.08	Yes
511.07	509.59	Yes
582.18	580.69	Yes
608.84	607.36	Yes
660.69	660.69	No
909.56	not present	No
1042.88	not present	No
1118.43	1116.94	No
1272.49	not present	No
1457.66	1459.14	Yes
not present	1589.50	Yes
not present	1682.27	Yes
1761.34	1761.34	No
2201.30	not present	No
2610.16	2611.64	No

Table 1: The gamma ray emissions in both mud samples, and if a background of the same energy is present.

The initial results from examining two different mud samples are as displayed in figure 1, where overlapping energies are paired together. Using the lab binders, each energy detected in the samples could be matched up with an appropriately close gamma-ray emission for an isotope in

the decay chain of one of the two of the most commonly occurring radioactive elements in the environment, ^{238}U and ^{232}Th [5]. The exception to this is the 660.69keV ray. While 660.69keV is close to a gamma-ray energy found in ^{214}Bi, a daughter element to ^{238}U, the intensities of this energy shown in both of the samples is much increased compared to what would be expected from the activity of the Uranium, suggesting that the contaminant must contain some other isotope emitting at this energy.

The contaminant is ^{137}Cs. As no noticeable emission is usually expected at the found energy, it can be assumed that all of the detected counts at this energy are from decaying caesium nuclei, and so, a relationship between counts and overall activity can be derived by using the calibration sample of caesium with a known activity. Due to the fact that mass of a substance is directly proportional to its activity, the errors on the mass can be propagated simply by multiplying them by the same constants as the activity. Following these steps yielded a total mass of caesium in the samples to be $(1.63 \pm 0.15) \times 10^{-14}$kg in the first and $(8.08 \pm 1.87) \times 10^{-15}$kg in the second.

If the 22TBq [1] of released ^{137}Cs is adjusted for the 66 years of decay since the accident and converted to mass, it can be written as ≈4.3g. Crudely estimating a spread distance from the accident cite to be a 10km radius, and that the atoms spread no further than around 1m into the ground, the caesium density expected is around 10^{-11}kgm^{-3}. The samples had volumes of around 2×10^{-4}m^3, so their caesium densities can be estimated as also $\approx 10^{-11}$kgm^{-3}.

5 Conclusions

In conclusion, it was extremely evident that there was some contaminant present in the mud samples provided. The contaminant was easily identified as we had been working with caesium plenty throughout the weeks of experimentation and recognised the gamma-ray energy. Testing the overall activity of the samples led to finding the masses of caesium equal to $(1.63 \pm 0.15) \times 10^{-14}$kg in the first and $(8.08 \pm 1.87) \times 10^{-15}$kg in the second, both of which are not unrealistic masses in the circumstances.

References

[1] J. R. Cooper, K. Randle, and R. S. Sokhi, *Radioactive Releases in the Environment : Impact and Assessment.* John Wiley and Sons, Inc, 2003.

[2] A. Einstein, “”Über einen die Erzeugung und Verwandlung des Lichtes betreffenden heuristischen Gesichtspunkt” [On a Heuristic Point of View about the Creation and Conversion of Light],” *Annalen der Physik (in German)*, vol. 322, pp. 132–148, Jan. 1905.

[3] G. F. Knoll, "Radiation Interactions, Semiconductor Diode Detectors, Germanium Gamma-Ray Detectors," in *Radiation detection and measurement*, ch. 2, 11, 12, New York ;: Wiley, 3rd ed. ed., 2000.

[4] A. H. Compton, "A quantum theory of the scattering of x-rays by light elements," *Phys. Rev.*, vol. 21, pp. 483–502, May 1923.

[5] https://www.ncbi.nlm.nih.gov/pmc/articles/PMC6572226/, *Natural Radiation in the Rocks, Soils, and Groundwater of Southern Florida with a Discussion on Potential Health Impacts.* International journal of environmental research and public health vol. 16,10 1793. 21 May. 2019, doi:10.3390/ijerph16101793., 03/05/2023.