International Journal of Research Publication and Reviews

Journal homepage: www.ijrpr.com ISSN 2582-7421

Study of Materials for Application in Alpha Counting Spectroscopy

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ABSTRACT

This paper describes the study of materials for application of alpha-particle counting for triple-alpha sources emitted by Plutonium, Americium and Curium (^{239}Pu, ^{241}Am and ^{244}Cm) in a chamber with a thermally diffused p-i-n detector and measure the alpha particle properties at different pressures. Properties of these alpha particles and the radioisotopes, includes the absolute activities, peak energies, ranges, intensity and energy straggling were investigated using earliest method. The alpha particles from the said source had estimated ranges of 4.78, 5.37 and 5.99 for ^{239}Pu, ^{241}Am and ^{244}Cm, respectively. The absolute activities for the radioisotopes in the triple-alpha source were found to be 555 ± 57 Bq, 375 ± 40 Bq, 101 ± 14 Bq for ^{239}Pu, ^{241}Am and ^{244}Cm respectively. Peak energies of the alpha particles were observed to have similar pattern as expected by the decay scheme of the radioisotopes. The intensity of alpha peaks decreases when air pressure increases and energy straggling which caused the increase in the widths of the alpha peaks were also observed.

Keywords: Plutonium-239, Americium-241, Currium-244, Alpha-spectroscopy machine.

Introduction

Alpha particle spectroscopy plays a growing importance in environmental protection as the focus of the nuclear industry shifts from nuclear weapon and fuel production to issues with waste management, decontamination and decommissioning of sites. Although alpha particles are not very penetrating they may cause a lot of damage to a material at the depth they reach. If an α-emitter is ingested internally they do not have to travel very far to reach living tissue or cell to cause serious damage; as it was the case for the poisoning of Russian ex-spy Alexander Litvinenko with Polonium-210 which is a pure alpha-emitter [1]. In the case of an alpha-emitter being inhaled it can cause serious damage to the lungs and respiratory system (tract). An important alpha-emitter occurring in nature is the long-life uranium-238 (U-238) and U-235 used in nuclear industry. Alpha spectroscopy is widely used to analyze environmental samples for contamination with uranium, plutonium, and isotopes found only in waste products of nuclear fuel.

Alpha Decay

Atoms tend to be unstable if they have a large number of nucleons. The nucleus of an atom is stopped from falling apart by the short-range, strong force that holds the protons and neutrons together. As the atomic number increases so does the electrostatic repulsion between the protons which is balanced by the short-range (~10^{-15} m) strong nuclear force between the nucleons. The presence of neutrons compensates for the electrostatic repulsion between the protons via the strong attractive force. More neutrons are needed to balance the long range electrostatic force as the proton number in a nucleus increases [6]. However, the more protons there are in a nucleus the greater the distance between them becomes. Eventually, the electrostatic forces dominate over the strong force and the nucleus decays in order to become more stable [7]. Many unstable heavy nuclei with an excess number of neutrons decay primarily by emitting alpha particles, usually in a succession of alpha decays to more stable lighter nuclei. It is well-known today that alpha particles are helium ($^{4}_{2}He^{+2}$) nuclei with two neutrons and two protons, first demonstrated in 1907 by Ernest Rutherford and Royds [2, 3] Classically, alpha-particles do not have enough energy to escape the strong potential of the nucleus. However, the tightly bound alpha particle formed in the nucleus tunnels through the coulomb potential barrier in the decay process, known as the quantum tunnelling effect [4]. The alpha decay is schematically illustrated shown below.

$^{A}_{Z}X \rightarrow {}^{A-4}_{Z-2}Y + {}^{4}_{2}\alpha$

(A = mass number, Z = atomic number, X = parent nucleus, Y = daughter nucleus)

The energy released in alpha decay, Q, is determined by the difference in mass of the parent nucleus and the decay products, which include the daughter nucleus and the alpha particle. The decay energy, Q for a parent and daughter nucleus at ground state is given by conservation of mass energy [3].

$$Q = \Delta E = \Delta mc^2 = (m_x - m_y - m_\alpha)c^2$$

The kinetic energy of the alpha particle is slightly less than Q because of the small recoil energy of the daughter nucleus. If the parent nucleus is at rest when it decays, the alpha particle and the daughter nucleus must have equal and opposite momentum. If p is the magnitude of the momentum of either particle, the decay energy is

$$Q = \frac{p^2}{2m_y} + \frac{p^2}{2m_\alpha} = \frac{p^2}{2m_\alpha}\left(1 + \frac{m_\alpha}{m_y}\right)$$

therefore Energy of alpha particle E_α ($p^2/2m_\alpha$) can be given in terms of the mass number of the parent nucleus, A as

$$E_\alpha = \frac{A-4}{A}Q$$

Alpha particles from a given parent isotope are emitted with a specific characteristic energy, always the same for a given parent isotope. Hence, by measuring the energy of the emitted α-radiation, it is possible to identify and measure the origin of the primary radiation. The general decay energy range for alpha particles is 2-8 MeV.

Interactions of Alpha Particles

The alpha particle loses energy primarily through the ionization and excitation of atoms on its path as it traverses through matter. The fact an alpha particle has a double charge and a large mass compared to an electron, its electric field pulls the electrons out of an atom it passes. The energy transfer from an alpha particle to an electron may be sufficient to knock it out of an atom and thus ionize it, or it may leave the atom in an excited state [8]. Because alpha particle has higher mass, collisions with electrons have little effect on their direction of travel and each collision takes a little of the alpha particles kinetic energy. Their travel paths remain in a fairly straight line. When an alpha particle is moving rapidly it is more likely to excite rather than ionize an atom. As an alpha particle loses energy through collisions along it path, it is continually slowed down. At slower speeds the alpha particle spends more time in the vicinity of each atom it passes giving it a greater opportunity to attract an electron, increasing its ionizing ability.

Energy loss in a material is a statistical stochastic process. Due to variations in the random collisions experienced along the path cause a slight 'straggling' around the mean penetration range with a Gaussian distribution.

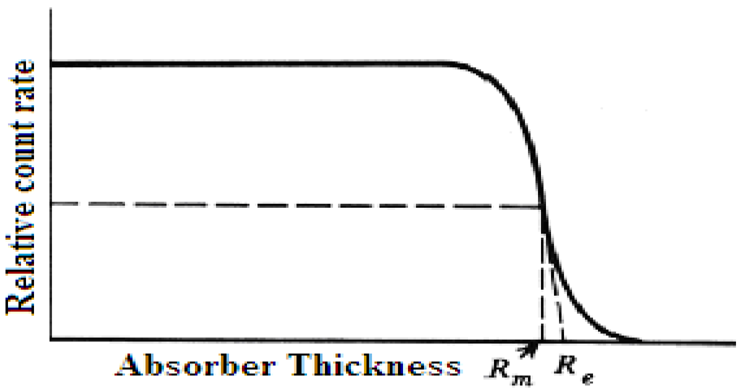

Diagram 1. Alpha particle absorption curve where R_m is the mean range and R_e is the extrapolated range. [3]

The rate of energy loss per unit path length for alpha particles is defined as its stopping power, S, and may be calculated using the low energy (E<10MeV) form of the Bethe-Bloch equation:

$$S = \frac{kz^2\rho_e}{v^2} ln\left[\frac{2m_e v^2}{I}\right]$$

where **k** = a constant, **z** = charge on the alpha particle, $\boldsymbol{\rho_e}$ = electron density of the absorber, **v** = velocity of the alpha particle, $\mathbf{m_e}$ = electron mass and **I** = average ionization potential of the absorber atoms.

In theory, it is possible to integrate the Bethe-Bloch equation over the entire path of the alpha particle to deduce its range. However, at low velocities this expression breaks down and empirical range-energy relations are often used instead. For alpha particles, it is common to use the following relationship:

$$R_\alpha = 0.318 E_\alpha^{3/2} cm of air$$

where E_α is in MeV and air is at standard temperature and pressure (STP)

Experimental Methods

Among the earliest methods used for measuring the energy of charged particles were the deflection of charged particles in electric and magnetic fields. While these techniques are still very important for absolute measurements the equipment required is impractical for general use due to its large size, complexity and costs. Semiconductor detectors are now routinely used for alpha spectroscopy.

Silicon surface barrier (SSB) detectors are traditionally used for alpha particle spectroscopy. These detectors typically consist of wafer of n-type silicon with a very thin gold contact evaporated on the front face and an aluminium contact evaporated on the back surface. A thin oxide layer forms beneath the gold layer upon exposure to oxygen [10]. This oxide layer acts as a narrow p-type region. When the junction between the n-type and p-type regions is reverse biased, the flow of two different charges creates a depletion zone, an electrically neutral area, where an electric potential difference is established, and hence an electric field [9]. Radiation is measured by means of the number of charge carriers set free in the detector, which is arranged between the two electrodes. When an alpha particle passes through the field region, it creates electron and hole pairs. The number of electron-hole pairs is proportional to the energy transmitted by the radiation to the semiconductor. Consequently, a number of electrons are transferred from the valence band to the conduction band, and an equal number of holes are created in the valence band. The electrons and holes swept apart by the strong electric field travelling to their counterpart electrodes. The motion of these charge carriers induces a sudden change in voltage between the two electrodes which can be measured via an external circuit. [3]

The thermally diffused p-i-n detector used in this experiment is similar to the SSB detector and operates with the same principles. However, the contacts are generated by bombarding the surfaces of the silicon wafer with dopant ions rather than by thermal evaporation of metal contacts. The result is a slightly thicker contact, with a modified internal filed distribution within the silicon wafer itself. [3]

A detailed description of the experimental procedure is given in the laboratory script *REP 7* and will be briefly outlined here. To avoid alpha particles losing energy before they reach the detector, alpha spectroscopy is carried out with the source and the detector inside a vacuum with the source mount just a few centimetres below the detector. The experimental set-up is shown in Diagram below.

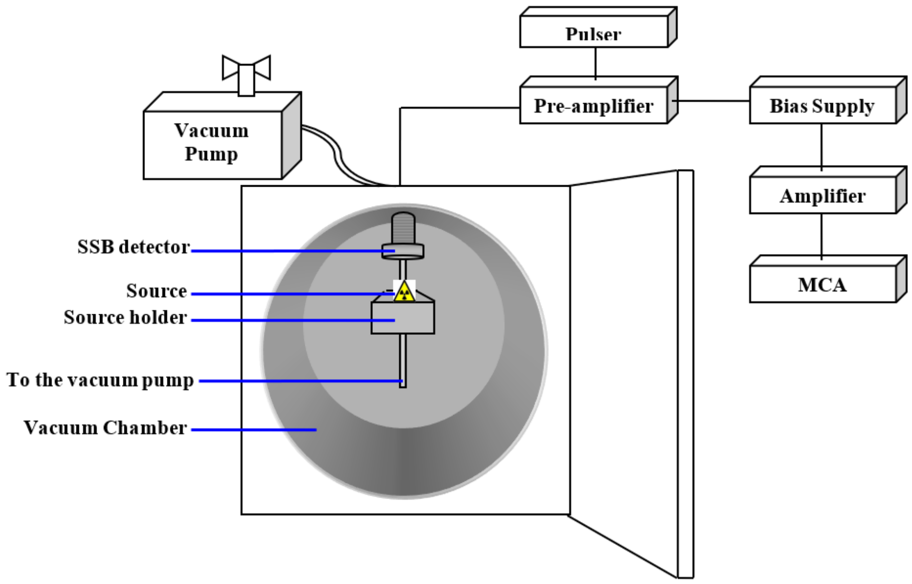

Diagram 2. Experimental arrangement for alpha spectroscopy and range measurement by varying the air pressure inside the vacuum chamber.

The source used in this experiment is a triple-alpha source containing Plutonium-239 (^{239}Pu), Americium-241 (^{241}Am) and Curium (^{244}Cm). *Table 1* shows a table of energies corresponding to alpha particle emissions from these isotopes.

Isotope	Energy (MeV)	Branching Ratio (%)
^{239}Pu	5.155 5.143 5.105	73.3 15.1 11.5
^{241}Am	5.546 5.513 5.486 5.443 5.389	0.25 0.12 86.0 12.7 1.3
^{244}Cm	5.805 5.763	73.3 23.6

Table 1. Alpha energies from triple alpha source containing ^{239}Pu, ^{241}Am, ^{244}Cm.

In the first part of the, the triple alpha source deposited on one side of a metal disk is mounted on the source holder and the vacuum chamber is fully evacuated. A spectrum is acquired using the experimental arrangement shown in *Diagram 2.* The triple-alpha source should have three groups of alphas from the three different components of the source with energies indicated in *Table 1*. The multi-channel analyzer (MCA) is calibrated from channel to energy scale using the main energy peaks (i.e. the peaks with highest branching ratios) corresponding to the three components of the source. The values of full-width-at-half-maximum (FWHM) are determined for the main energy peaks.

The MCA is calibrated again, however, this time a more accurate method which includes lower energies than before. A new spectrum is collected as before. The pulser is used to inject a small charge into the preamplifier of known magnitude. The output of the pulser is adjusted until a peak appears close to the ^{241}Am peak. The FWHM of the pulser peak is determined.

The pulser output is then adjusted so that it overlies the ^{241}Am peak at 5.47 MeV exactly. Several pulser peaks are recorded on the same spectrum with pulser outputs corresponding to 1, 2, 3, 4, 5 and 6 MeV and the peaks are used to calibrate the MCA.

Following calibration, a set of seven alpha spectra at gas pressures of 0, 100, 200, 300, 400, 500, 600 and 760mBar in air are recorded using the same source. In each case, the peak channel position and integral counts under each of the 3 main peaks are determined.

Finally, the intensity of alpha peaks is measured as the pressure increases by recording the count rate in that peak.

Results and Discussion

The alpha spectrum for the triple source was accumulated for 20 minutes and the MCA calibrated using the main energy peaks indicated in *Figure 1*.

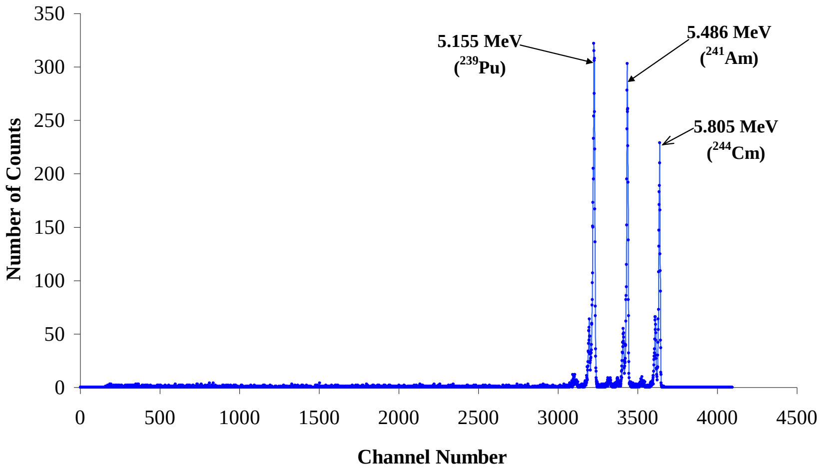

Figure 1. The ^{239}Pu, ^{241}Am and ^{244}Cm spectra collected for 20 minutes showing the main peaks.

While the main peaks of ^{239}Pu**,** ^{241}Am and ^{244}Cm can be identified note that the peak width is wide and some of the fine details, i.e. other alpha emissions similar in energy, cannot be seen. *Figure 2* displays the graph of FWHM versus peak energy for the 3 main peaks which shows a near straight line relationship. A linear increase in FWHM is to be expected with increasing peak energy. This is because the beam of alpha particles with higher energy will pass through more amounts of silicon which means that there would be more straggling and therefore lower resolution. However, the results show an almost straight line relationship. This will be discussed later.

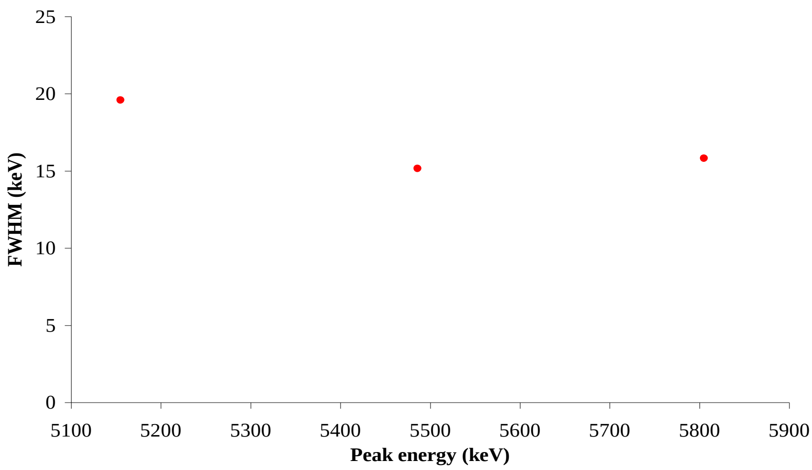

Figure 2. The graph of FWHM versus peak energy.

The MCA was calibrated again using the pulser outputs corresponding to 1, 2, 3, 4, 5 and 6 MeV. The spectra are displayed in *Figure 3*. The FWHM due to counting statistics alone was estimated to be 14.41 keV.

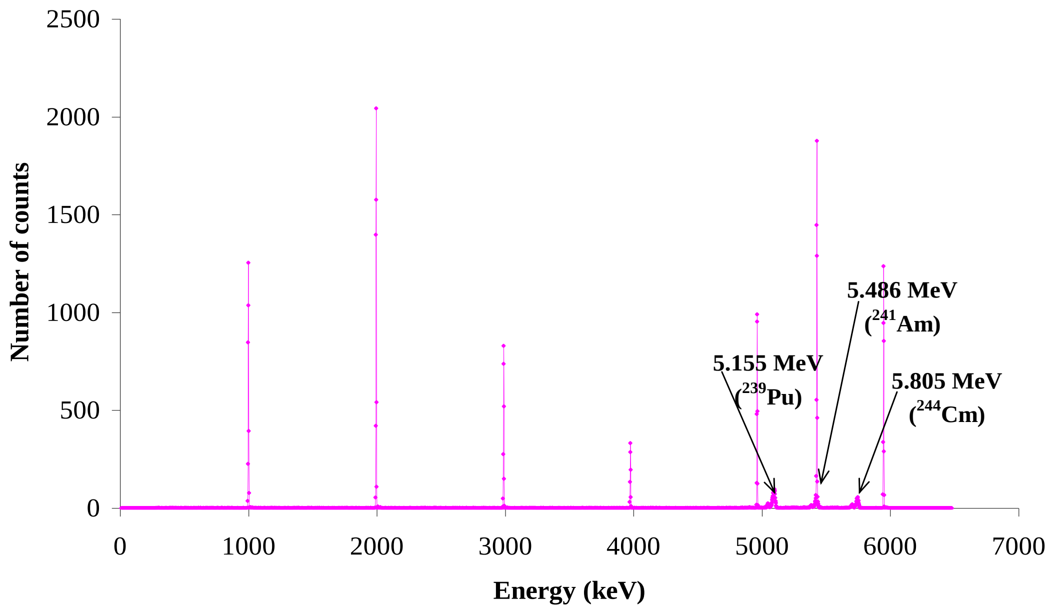

Figure 3. Pulser Calibration Spectrum.

The air pressure inside the vacuum chamber was varied from 0 to 760mBar and the corresponding peak energies were measured. The air thickness corresponding to these pressures were calculated using the knowledge that the density of air is 1.293mgcm^{-2} and detector-source distance is 1.5cm. The plots of peak energy versus air pressure for ^{239}Pu, ^{241}Am and ^{244}Cm are shown in *Figure 3*. By extrapolating the curves, the air thickness at which the alpha particles are finally stopped can be estimated.

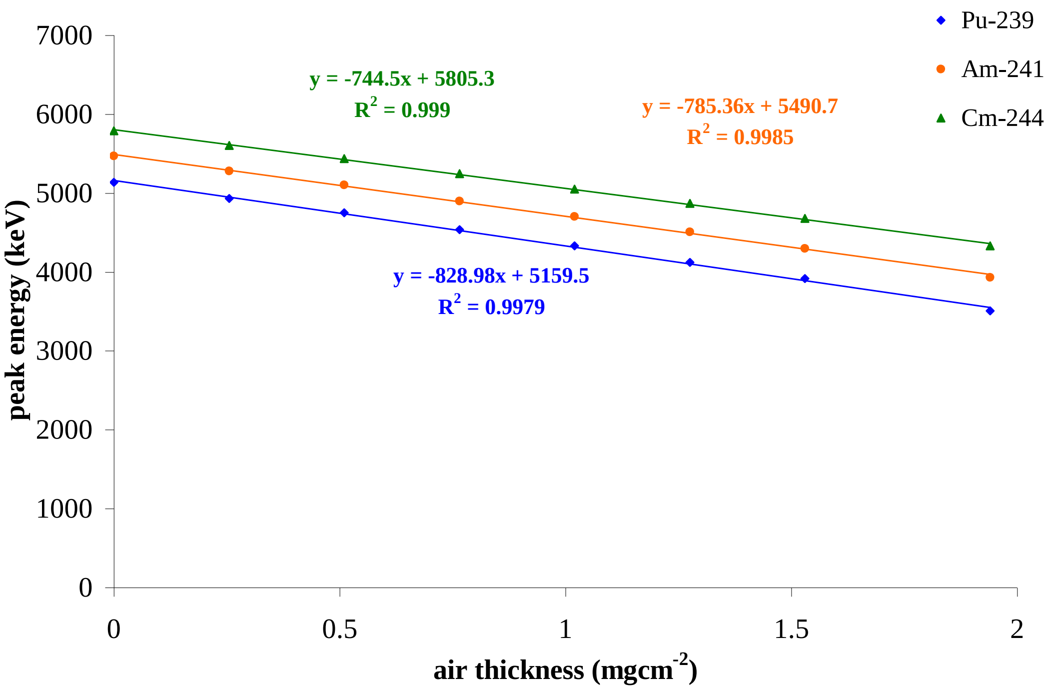

Figure 3. Peak energy versus Air Thickness for the triple alpha source.

In order to confirm the validity of the empirical expression $R_\alpha = 0.318E_\alpha^{3/2}cm$of air, the expected and observed range of alpha particles for each of the 3 components of the source are as follows:

Isotope	Energy (MeV)	Expected Range (cm)	Observed Range (cm)
^{239}Pu	5.155	3.72	4.78
^{241}Am	5.486	4.07	5.37
^{244}Cm	5.805	4.45	5.99

Figure 8 shows how the FWHM varies as the air pressure inside the vacuum chamber is increased. The FWHM increases as the air pressure is increased.

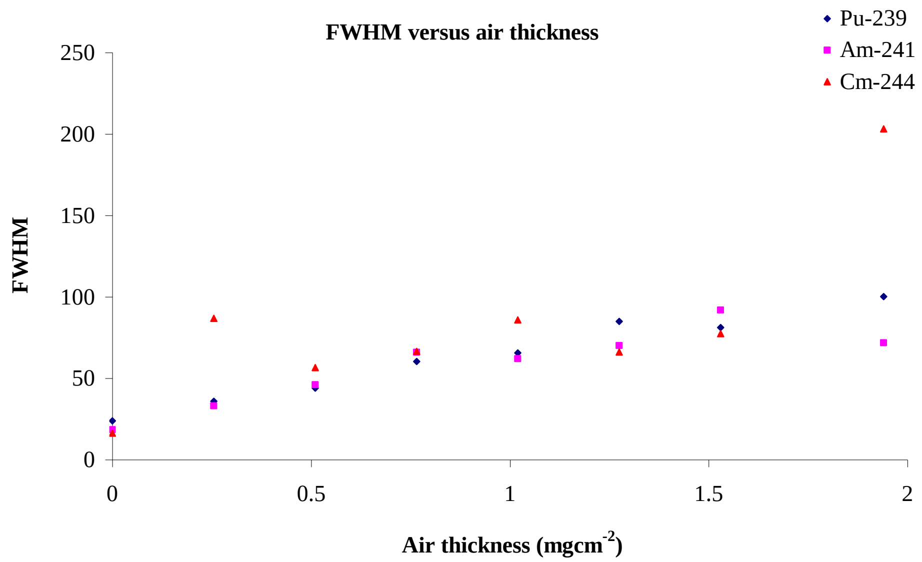

Figure 4. FWHM versus air thickness for the triple alpha source.

The increase in the widths of alpha peaks as the pressure increases is due to energy straggling. The increase in the energy distribution of the alpha particles reaching the detector is due to fluctuations in the number of atomic collisions along its track through air. A theoretical prediction to describe energy straggling was developed by Bohr that may be written as:

$FWHM = 41.6(\Delta x)^{1/2}$ keV

where Δx is the air-path thickness ($mgcm^{-2}$)

To validate this prediction a graph of FWHM of the ^{241}Am versus square root of the air thickness was plotted in *Figure 5*. The observed FWHM line is slightly higher than the expected.

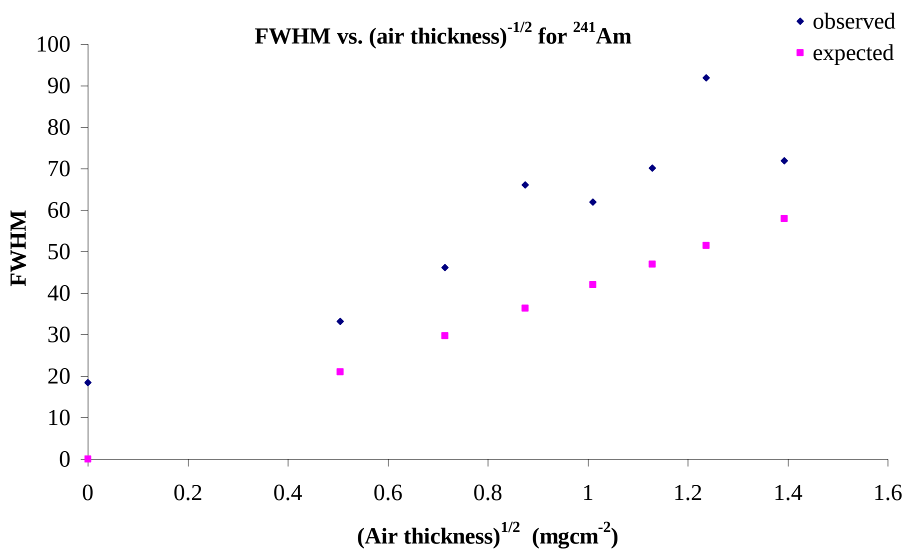

Figure 5. The expected and observed FWHM versus air thickness for ^{241}Am.

The count rate versus air thickness was plotted for of ^{239}Pu, ^{241}Am, and ^{244}Cm; and the results are displayed in *Figure 6*. The ^{239}Pu emits highest alpha particles with highest energy, and ^{244}Cm the lowest. Therefore ^{239}Pu in the source is more alpha particles than both ^{241}Am and ^{244}Cm. Alpha particles from a single type of source are mono-energetic particles which means they all have the same energy. Therefore there is a fairly well defined depth to which the particles penetrate into a material and this property is exhibited in *Figure 6.* The count rate remains constant with varying thickness until alpha particles are finally stopped (more measurements needed to see this) and as shown *Diagram 1*.

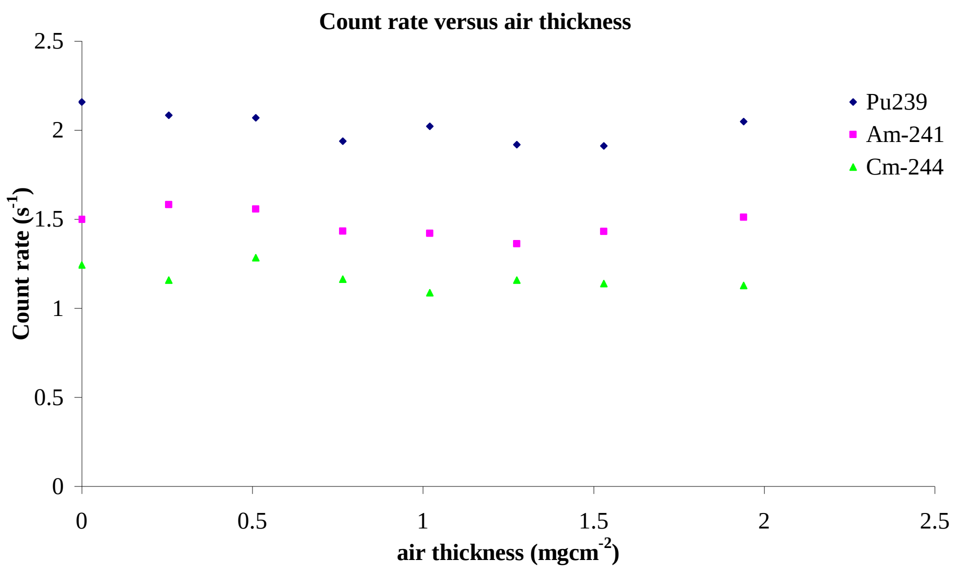

Figure 6. The count rate versus air thickness for the triple alpha source.

While the major peaks can be distinguished clearly due to their energy separation, finer details of the source spectrum are not resolved due to straggling even in the vacuum. The FWHM values were increased more with increasing air thickness adding to the broadening of energy spectrum when alpha particles reach the detector.

In *Figure 2* the FWHM is expected to increase with increasing energy, however an almost straight line is observed. This could be due to experimental technique while choosing a region of interest to determine the FWHM. The fact that peaks are not resolved very well means the FWHM of another peak might be included in the measurements; and it is also possible that the source is damaged and this might also add to the straggling of energy.

The expected range and observed range have difference of approximately 30% which dependent on the calibration of MCA and resolution of the peaks.

Conclusions

The alpha particles from the triple source had estimated ranges of 4.78, 5.37 and 5.99 for ^{239}Pu, ^{241}Am and ^{244}Cm, respectively, approximately 30% of the expected values.

The resolution of the spectrum does not enable us to see the finer details in the spectrum. However, in conclusion it can be seen that there is a fairly well defined depth to which the particles penetrate into a material as demonstrated.

Acknowledgement

I would like to use this opportunity to express my appreciation to the member of staff and PhD students who assisted me in working towards achieving this experiment.

Special note of thanks to all the staff in the Department of Physics, Sokoto State University for their support for the successful completion of this research.

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