Application of Alpha-Particle Spectroscopy in the Material Testing

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Abstract:- This report is based on an experimental study on the energy of alpha particles that were emitted by a triple-alpha sources named ^{239}Pu, ^{241}Am and ^{244}Cmin a chamber that has a diffused p-i-n detector and measure the alpha particle properties at different pressures. Properties of these alpha-particles and the radioactive particles of some minor corrections to the biggest transaction of certain productivity and the radioisotopes, which include; absolute activities, peak energies, ranges, intensity and energy straggling, were all investigated. The results of the absolute activities for the radioisotopes in the alpha sources were;555 ± 57 Bq, 375 ± 40 Bq, 101 ± 14 Bq for ^{239}Pu, ^{241}Am and ^{244}Cm respectively. Different peak energies of the triple alpha-particles were observed to have similar shape as predicted by the radioisotopes decay scheme. Experimentally, the ranges of the triple alpha particles were found to be as follows; 6.06 ± 0.46 mg cm^{-2}, 6.82 ± 0.53 mg cm^{-2} and 7.46 ± 0.53 mg cm^{-2} for ^{239}Pu, ^{241}Am and ^{244}Cm respectively at standard temperature and pressure.As the air pressure increases, the intensity of the triple alpha peaks decreases and the energy straggling which caused the increase in widths of the alpha peaks were observed.

I. INTRODUCTION

Alpha particles are used in smoke detectors, radioisotope thermoelectric generators, unsealed source radiotherapy and etc. The aim of the experiment is to understand the properties of the alpha particles and the radioisotopes releasing these particles, which includes; absolute activities, peak energies, ranges, intensity and energy straggling. Moreover, as they were dealing with silicon p-i-n detectors, alpha particle spectroscopy is widely used as detectors.

The presence of direct and indirect radiation can be found by detecting the energy charged particles generated by radiation detection. Subsequently alpha particles produced have distinctive energy depending on the nucleus emitted and the daughter cells product, it is likely to know the source of primary radiation by the peak energy of the alpha particles produced.

Semiconductor detectors have a very good energy resolution, high intrinsic detection efficiency and fast response in detecting alpha particles. The amount of energy require to produce ion pair in a semiconductor material 10 times smaller than that in a molecule of gases, so the greater number of ion pairs produced in semiconductor. Good energy resolution follows as the greater ion pair causes the motion/random fluctuation minimal. This large number of ion-pairs makes the random fluctuations relatively smaller, and a good energy resolution follows. Semiconductor detectors have higher compactness than of gas-filled detectors;and recombination happens easily after the ion-pairs. This produces a high intrinsic detection efficiency and fast response.

The research works have two sections. The first is alpha spectroscopy and the second is the range of alpha particles in air and the effect of energy straggling.

II. THEORY

Most of the alpha particles have energy between 3 and 7 MeV, depending on the emission of the half-life is long or short, respectively. Alpha particles are doubly helium nuclei, (consists of two protons and two neutrons) bound together into one particle. They have high ionizing power and lose energy readily when they travel in air. Therefore, the range of alpha particles in air is typically just a few centimeters. The alpha particles detected in this experiment were produced from alpha decay of three radioisotopes ^{239}Pu, ^{241}Am and ^{244}Cm. Alpha decay results from the Coulomb repulsion between the alpha particle and the rest of the nucleus. Originally, the alpha particle and the rest of the nucleus are bound together by the short range nuclear force. The coulomb repulsive between themprovides a certain amount of energy for the alpha particles to escape from the nucleus. In fact, this energy is not enough for alphaparticles to escape the potential well from the strong nuclear force inside the nucleus. However, the quantum tunnelingeffect allows these particles to escape even though they have insufficient energy to overcome the nuclear force.

The main alphadecay process of ^{239}Pu, ^{241}Am and ^{244}Cm can be described by the following equations:

$$^{239}_{94}Pu \rightarrow \ ^{235}_{92}U + \ ^{4}_{2}\alpha \qquad (1)$$

$$^{241}_{95}Am \rightarrow \ ^{237}_{93}Np + \ ^{4}_{2}\alpha \qquad (2)$$

$$^{244}_{96}Cm \rightarrow \ ^{240}_{94}Pu + \ ^{4}_{2}\alpha \qquad (3)$$

Moreover, alpha transitions to excited quantum states of the daughter nuclei are possible, and thus there are more than one possible peak energies possible for ^{239}Pu, ^{241}Am and ^{244}Cm. X-ray is given out when the excited daughter nuclei drops to a lower energy state. Typically, in the case for these three radioisotopes, x-ray of a few tens eV may be observed as weak “fine structure” peaks. Below in the Table 1, are the three Isotopes of alpha particles used in this research work as shown Detailed decay scheme can be found in [1].

Radioisotope	Energy (keV)	Branching ratio (%)
^{239}Pu	5105	11.5
	5143	15.1
	5155	73.3
^{241}Am	5389	1.3
	5443	12.7
	5486	86.0
	5513	0.12
	5546	0.25
^{244}Cm	5763	23.6
	5805	73.3

Table 1 Alpha particles emissions from ^{239}Pu, ^{241}Am and ^{244}Cm [5]

Alpha particles are normally detected using semiconductor detectors because of their good energy resolution, high intrinsic efficiency and fast responses. The detector used in this experiment is a thermally diffused p-i-n detector which is similar to a silicon surface barrier detector. Basically, silicon surface barrier detectors comprise a wafer of n-type silicon which has a very thin gold contact evaporated on the front surface and an aluminum contact evaporated on the back surface as the ohmic contact. P-type region is formed when a thin oxide layer forms beneath the gold layer. When reverse bias voltage is applied to this p-n diode, a depletion region will be formed. Due to the reverse bias voltage, there is a very low density of charge carriers in the depletion region. When electron-hole pairs are created in the depletion region by the incident radiation, the electron-hole pairs are rapidly collected by the two electrodes [2]. Instead of evaporation of metal contacts, the contacts of the thermally diffused p-i-n detector used in this experiment are created by bombarding the surfaces of the detector with dopant ions. The drawback is that the contact is slightly thicker with a modified internal electric field distribution in the silicon wafer [5].

It is assumed that 100% intrinsic efficiency of the detector,

$I$is the absolute activity of the source, which can be calculated by

$$I = \frac{C_\alpha}{f} \times \frac{4\pi d^2}{A} \qquad (4)$$

$C_\alpha$count rate,
$f$branching ratio,
Aactive area of the detector and
$d$distance between the detector and the source.

The stopping power can be define as a rate of loss of energy (E) per unit path length x for a particle travelling through matter.

$$S = -\frac{dE}{dx}$$

To define the range R_α, Bethe-Bloch equation can be applied which describes S to carry out the following integration:

$$R_\alpha = \int_{Initial\ energy}^{0} -\frac{1}{S} dE \qquad (5)$$

It is very hard to evaluate the breakdown the Bethe-Bloch equation at velocity near zero. Therefore, instead of using equation (5), an empirical range-energy can be used to analyze the theoretical range of alpha particles in air:

$$R_\alpha = 0.318{E_\alpha}^{3/2}\ cm\ of\ air \qquad (6)$$

$E_\alpha$stand for peak energy of the alpha particles measures in MeV and air is at standard temperature and pressure (STP).

To validate the equation (6), the range of alpha particles of the isotopes can be determined experimentally by changing the air pressure of the chamber containing the source and detector, and then using an extrapolating plot of peak energy versus air pressure to evaluate the air pressure P_{stop}at which the alpha particles stop. To compare the experimentally determined with the range calculated by equation (6), the unit mg cm^{-2} is usually used. Equation (6) can be changed to:

$$R_\alpha = 0.318{E_\alpha}^{\frac{3}{2}} \times \rho_{STP}\ mg\ cm^{-2} \qquad (7)$$

$\rho_{STP} = 1.293\ mg\ cm^{-3}$ is the density of air at STP. In addition, from ideal gas law, air pressure of the chamber P is proportional to the air density ρ in the chamber. Therefore, one may calculate the air densityρ_{stop} at which the alpha particles stop by:

$$\rho_{stop} = P_{stop} \times \frac{\rho_{STP}}{P_{STP}} \qquad (8)$$

$P_{STP} = 1013$ mbaris the air pressure at room temperature.Finally, experimentally determined range is given by

$$R_\alpha = d \times \rho_{stop}\ mg\ cm^{-2} \qquad (9)$$

As alpha particles loss more energy with increasing air density in the chamber, it is also expected that the intensity of the alpha peaks drops with air pressure. A graph of count rate versus air pressure can be drowned to show this prediction.

$FWHM_{tot}$Peak width depends onstatistical noise and electronic noise according to [2]:

$$FWHM_{tot} = \sqrt{(FWHM_{elec})^2 + (FWHM_{stat})^2} \qquad (10)$$

$FWHM_{elec}$Stands for full-width-at-half-maximum of the pulser peak
$FWHM_{stat}$Stands for full width at half maximum due to counting statistics

Except the cunning part of $FWHM_{stat}$, the FWHM mentioned in other parts is talking about $FWHM_{tot}$. By using a pulser to inject a small charge into the preamplifier of known magnitude, one may measure the $FWHM_{elec}$ and analyze the contribution of electronic noise. Subsequently, $FWHM_{stat}$ can be determined through equation (10). Error in the FWHM σ_{FWHM}can be calculated by the following equation [2]:

$$\sigma_{FWHM} = \frac{FWHM}{2.35\sqrt{2A_n}} \qquad (11)$$

A_nStands for total number of counts of the peak.

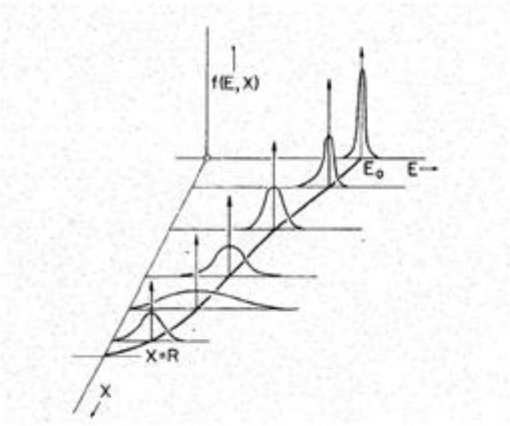

Fig. 1 Schematic plot of energy distribution of a beam of initially monoenergetic charged particles at various penetration distances. *E* is the particle energy and X is the distance along the track [2].

As air pressure increases, it is expected that the peaks get wider due to energy straggling. The density of the air in the chamber increases with the air pressure, and more collisions of the atomic particles between the alpha particles and the air particles produce. Consequently, more oscillations will happen in the energy of the alpha particlesas a result of different energy loss that the alpha particles experience along their paths.Fig.1 shows a schematic diagram to illustrate energy straggling [2].The peak narrows again near the end of the range because the mean particle energy has greatly been reduced.

To predict the energy straggling, a theory was developed by Bohr (3). The equation may also be written as (5):

$$FWHM_{stat} = 41.6(\Delta x)^{1/2}\ keV(12)$$

ΔxStands for air-path thickness measured in mg cm^{-2}. A graph of peak width versus air pressure can be drowned to show the idea of energy straggling and verify equation (12).

III. EXPERIMENTAL METHOD

The model of the detector used was Hamamatsu S1223-01 Si PIN photodiode while the source used were triple-alpha source that contains three isotopes Plutonium-239,Americium-241 and Currium-244 (239Pu,241Am and 244Cm (S216.PH disc)).The details of the peak energies and their corresponding branching ration are shown in Table 1.Ortec model 676 alpha spectrometer in NIM is linked to the following; MCA,PC and a vacuum pump.Fig.2 demonstrates the experimental setup.

The alpha source was placed on the source holder in the vacuum chamber which was built in the alpha spectrometer module.The alpha source was kept at a distance of about 1.5 ± 0.1 cm away from thep-i-n detector.The energy range was set to be about 3-8 MeV as the peak energies of the triple-alpha source was about 5 MeV.The pressure of the vacuum chamber was varied using the vacuum pump and pressure gauge.The time for acquiring a spectrum was set to 300 seconds for all the cases.

The first section of the experiment, the pressure of the chamber was initialized to start at the lowest value which was 20 ± 10 mbar. A spectrum of all the alpha sources was acquired and the calibration was carried out by using the three main peaks of the triple-alpha source. Afterwards,the absolute activity of the source was calculated using equation (4).

Second section of this research work, Americium-241 is formed very close to a pulser peak to know the width of the pulserpeak$FWHM_{elec}$. With the aids of this equation (10), $FWHM_{stat}$ was calculated. The air pressure of the chamber was change to different setting of intervals of 200 each to determine the range of alpha particles. The changes goes 200, 400, 600, 800, 1000 mbar with an error of ± 10 mbar. A sixspectra of alpha particles using the above values of air pressure were found and taken. As stated in the theoretical part, the procedure was adapted to determine the range of alpha particles. A plotted graph of FWHM that is peaks versus air pressure was plotted to understand the challenges of energy straggling and to verify equation (12). Lastly, a graph of count rate against air pressure was plotted to study the variation of intensity of travelling alpha particles in the air.

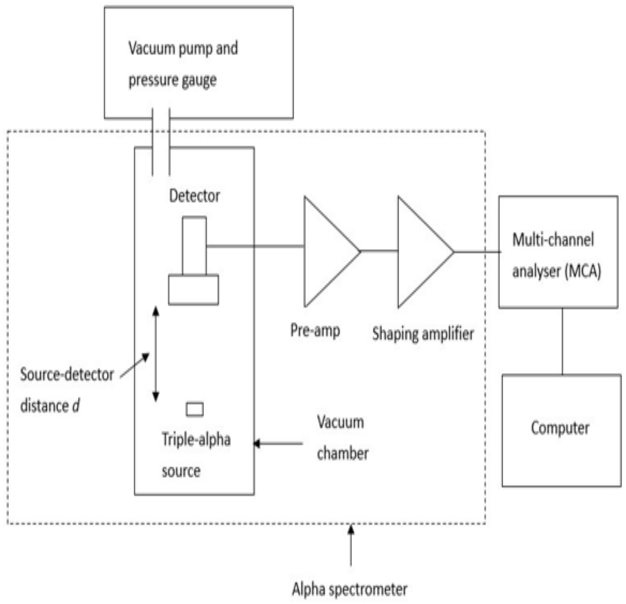

Fig. 2 Alpha Spectroscopy experimental set-up

IV. RESULTS AND DISCUSSION

Alpha Spectrometry

The spectrum at pressure 20 ± 10 mbar learnt that Figure 3 showing three different peaks as expected of triple alpha source according to table 1, In the appendix 1, a thinner spectrum energy is observed for the three radioisotopes peak.It was known that the peak energy that is very close to the first highest branching order/ratio could be found for Americium-241 and curium-244. According to Ortec Maestro, Plutonium-239 corresponded to the energy 5105 (KeV) for the second peak. Moreover, corresponded peak of 5143 KeV actually overlaps with main peaks of Pltonium-239.

Conferred with Theoretical part, weak “fine structures” of few tens of electron volts (eV) might be noticed. Though, there was nowhere to be found in the Figure 3. Rather, peak was seen around 1800 KeV. Being the energy range of the alpha spectrometer was actually 3 – 8 MeV, the lowest limit for the detection of this experimental set up is 1800 KeV. In order to detect the lower-energy region, there might need for another spectrometer for weak “fine structures” peaks.

Considering the main energy peak, the full width at half maximum (FWHM) was manually determined by calculating the width at half maximum than directly getting it from the Ortec Maestro.

The FWHM for the main energy peaks were determined by manually calculating the width at half maximum rather than obtaining directly from OrtecMaestro. This happens as a result of when the air pressure increases, the peak expands, Maestro provided an arbitrary full width at half maximum for the main peak. The manually calculated full width at half maximum (FWHM) were considered to be more reasonable. For comparison, the data was tabled in Appendix 2.Therefore, to keep consistency, the main peak’s full width at half maximum (FWHM) were manually determined while conducting this experiment except for the pulser peak. In the Fig. 4 is a graph of full width at half maximum (FWHM) for the three (3) peaks against peak energy.

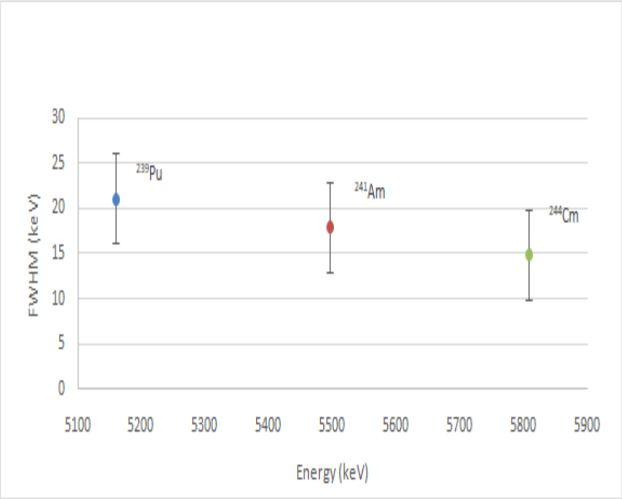

Fig. 4 Full Width at Half Maximum versus Energy of the triple-alpha source

This particular procedure induced huge error as the locations of half maximum were hard to find, more especially the peak were wider at the range of 600, 800 to 1000 mbar. The FWHM's error was projected to be ±5 keV because one phase size in Maestro was about 2.5 KeV. In the cases of 600, 800 and 1000 mbar, it was likely that the true error in full width at half maximum was higher than 5 KeV because of the difficulties in finding the half maximum.

Absolute activity of each of the radioisotopes

The absolute activity of each of the radioisotopes was calculated and summarized in Table 2, with an assumption of 100% intrinsic efficiency as stated in the equation (4). The branching ratio f was the sum of 0.151 and 0.733 instead of 0.733 for Plutonium-239, as the peaks (5143 and 5155 KeV) met. So the true value of the activity should be bigger than the calculated absolute activity because it was perfectly assumed that 100% intrinsic efficiency in the activity. The record for the true activity is not available, therefore no comparison. The noise for the background was not recorded separately for a spectrum but Maestro was used for background noise.

Radioactive isotope	Energy(keV)	Activity (Bq)
^{239}Pu	5155	555± 57
^{241}Am	5486	375 ± 40
^{244}Cm	5805	101 ± 14

Table 2 Calculated values for absolute activity of the three radio isotopes

$FWHM_{elec}$ and $FWHM_{stat}$

By changing gain of spectrometer produced the Americium-241 (^{241}Am) very close to the pulser peak.

Full width at half maximum(FWHM) of the pulser peak was determined to be 4.89 ± 0.01 KeV. This particular value's error was believed to be based on approximation than equation (11). Because total count A_n relies totally on time and if 300 seconds set for the used to obtain the pulser peak, the total count would surely become bigger to full width at half maximum (FWHM). Using equation (11) will produce us with an error closer to zero. It was not telling us anything reasonable, thus high estimated number 0.01 KeV was used.

$FWHM_{stat}$ for the americium-241 (^{241}Am) peak was determined with the aid of equation (10) to be 17.29 ± 5.17 keV, this value is almost the same with the valueof $FWHM_{tot} = 17.67 \pm 5.00$ KeV.

As can be seen here, the peak's error largely rest on the noise and the electrical noise is insignificant for this spectrometer. As a result, equation (12) was applied. The linear responses for Multi-channel Analyser were noticed as the shaping gain was changed.

The alpha range

About six different alpha spectra were observed and recorded at air pressure set at 20, 200, 400, 600, 800, 1000 ±10 mbar. Below in Figure5, is a plotted graph of Energy against pressure.

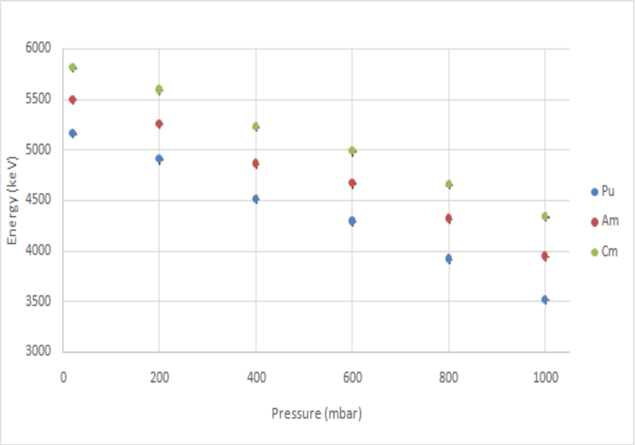

Fig. 5. Peaks' Energy versus pressure

The error bars were nowhere to be seen on the figure 5 shown above, though the peak energy have the errors of order in between 0.1 to 1. The relationship between the energy and pressure were linearly seen. By the means of linear regression in Microsoft word rather Excel, the root square value (R^2) were solved to be 0.9942, 0.9946 and 0.9946 for ^{241}Am, ^{239}Pu and ^{244}Cm respectively. As can be seen, the R^{2}values were all found to be very close to 1, and it shows that all the points are on the same line (6).This result indicates that the evidence is indirectly related to good performance of detectors in while detecting the peak energy of the source.

By inferring the linewhich is suitable to zero energy and pressures corresponding to zero energy of the alpha particles were determined and summarized in Table 3 below;

Radioisotope	Pressure (mbar)
^{239}Pu	3164 ± 118
^{241}Am	3562 ± 138
^{244}Cm	3896 ± 95

Table 3 Pressure corresponding to zero energy of the alpha particles releasedby the source

The range for alpha sources for each radioisotope was calculated as seen in a tabular form below;

Radioisotope	Range calculated by equation (9) (mg cm^{-2})	Theoretical Range given by equation (6) (mg cm^{-2})
^{239}Pu	6.06 ± 0.46	4.81
^{241}Am	6.82 ± 0.53	5.28
^{244}Cm	7.46 ± 0.53	5.75

Table 4 Experimentally determined range and calculated range by using equation (6)

The empirical range energy written in equation (6) and the determined range are in the same order of range. With the aid of T-test, the latter on is expressively smaller than the former one. This difference comes from some assumptions that were not truth base on kinetic theory of gasses that is ideal gas equation as can be seen in equation (8). Also the distance between the source and detector was not measured as it has built-in detector in the chamber, so the detector's position was not known. It could be the spectrum might wrongly be calibrated. The three energies of the main peaks of radioisotopes were fully calibrated. As such, the calibration of the energy region confined by all the three peaks might not be right in the region.

Energy Straggling

A plotted graph of FWHM of peaks versus pressure for each radioisotope in Fig. 6, it shows that the peaks increases with air pressure as a result of energy straggling as discussed in the theoretical aspect. Graph of full width at half maximum (FWHM) of Americium-241 against the square root of the air thickness was plotted to verify the theoretical prediction in describing the energy stragglingin equation (13), as can be seen in Fig. 7. $FWHM = a(\Delta x)^{1/2}$in Microsoft excel, the coefficient a is 41.6 keV mg$^{-1/2}$ cm. Using T test, the coefficient's value is higher than the theoretical value. Their differences happen as a result of determining the full width at half maximum of the peak. Subsequently, there were additional peaks near the main peak of the entire three radioisotopes. Because of the energy straggling, the peaks partly covered when air pressure was added as can be seen in the in figure 8. As a result, the main peak was affected by the neighboring peaks. The full width at half maximum recorded was probably bigger than the real value as the neighboring peak invested their peak energy. As can be seen in the figure 7, the slope is bigger than it was expected. This proves that the derived coefficient$a$, in other word the calculated value was bigger than the theoretical value.

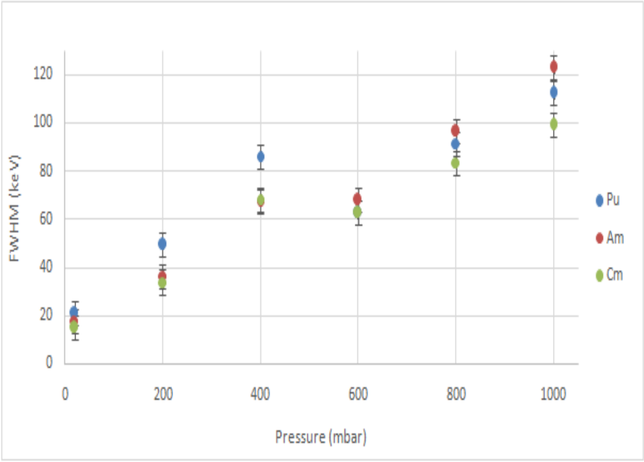

Fig. 6 Full Width at Half Maximum versusPressure

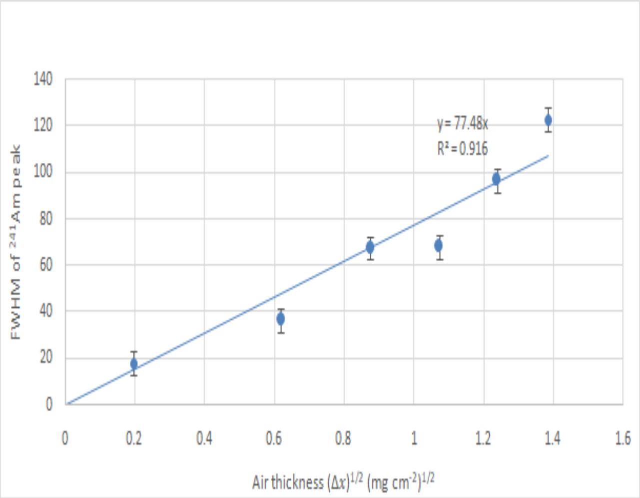

Fig. 7. Full Width at Half Maximum of Americium-241 peak against the air thickness

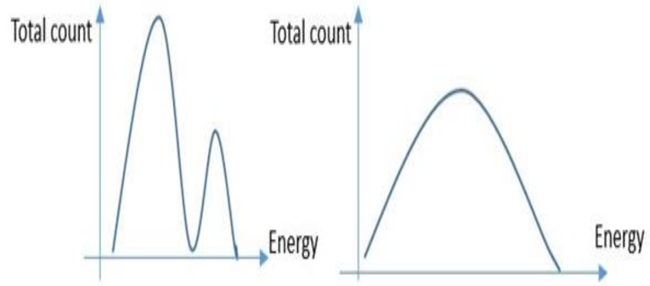

Fig.8 The two diagrams shown in the above have different peaks as can be seen, the left onedisplays two separate peaks as observed at a lower air pressure and the right onedisplays the met peak as can be seen when air pressure amplified.

Intensity of the alpha peaks

Fig. 9, shows a graph of count rate of the alpha particle at different values of air pressure. Ameicium-241 and Plutonium-239 counts rate decreases as the air pressure decreases. As the air increases, the alpha particles and air particles collide, this causes the alpha particles to loss energy and small creation of electrons hole happens in the detector. But Curium-244, the source activity is low and that makes the count rate to be low. There was a random fluctuation that makes the observation of Curium-244 to be hard in the count rate due to it reduction.

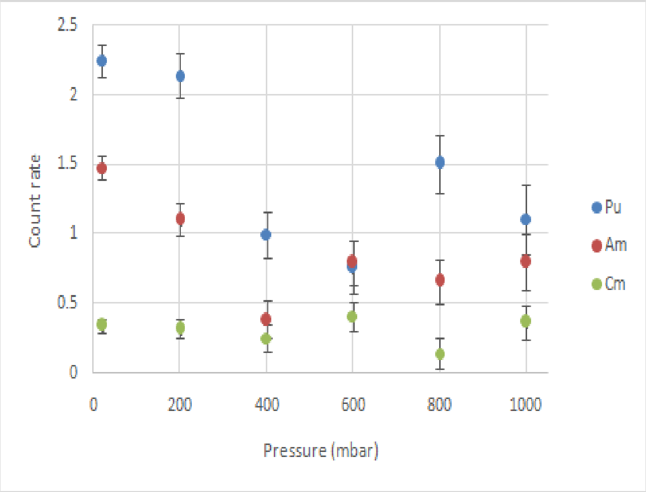

Fig. 9 Intensity of the alpha versus pressure

V. CONCLUSION

By the use of all the three sources (Plutonium-239, Americium-241 and Curium-244) and surface barrier detector, that makes it easier to measure the number of released alpha particles. These kinds of properties are the energy of the alpha particles and absolute activity of radioisotopes, which are 555 ± 57 Bq, 375 ± 40 Bq, 101 ± 14 Bq for Plutonium-239, Americium-241 and Curium-244respectively. The range of alpha produced from the radioisotopes which were found to be 6.06 ± 0.46 mg cm^{-2}, 6.82 ± 0.53 mg cm^{-2}and 7.46 ± 0.53 mg cm^{-2}for Plutonium-239, Americium-241 and Curium-244 respectively at standard temperature and pressure. In addition, the intensity found in the alpha particles decreased with the pressure, the straggling energy were found as a result of the increased in the pressure. Theoretically, the energy straggling (referring to equation (12)), was not verified because of the increased full width at half maximum (FWHM) overlapping two closer peaks of the radioisotopes.

In other words, this triple-alpha source was actually not suitable to verify equation (12). To improve this part of experiment, a source emitting alpha particles with only one peak energy can be used to replace the triple-alpha source.

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Appendix 2:

Results of FWHM foundfromOrtec Maestro and measured FWHM

Error is not shown as it is not meaningful if unreasonable FWHM is obtained.

Plutonium-239

Pressure (mbar)	FWHM got from Ortec Maestro	Measured FWHM
20	21.19	21.14
200	36.58	49.68
400	23.2	86.33
600	4.99	62.75
800	5.56	91.52
1000	37.39	112.43

Americium-241

Pressure (mbar)	FWHM gotfromOrtec Maestro	Measured FWHM
20	15.44	17.97
200	23.95	36.6
400	5.44	67.61
600	9.31	67.98
800	4.23	96.74
1000	6.84	122.9

Curium-244

Pressure (mbar)	FWHM got fromOrtec Maestro	Measured FWHM
20	8.44	14.96
200	9.77	33.99
400	3.76	68.27
600	13.47	62.75
800	10.89	83.67
1000	15.23	99.36