Neutron flux through γ spectroscopy

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1 Introduction

Radioactive decay is a phenomenon in which a nucleus emits an ionizing particle. The type of particle and its energy depend on the process causing the emission and the material. The latter cause implies that different sources of radioactive material have different energy spectra and thus the radioactive spectrum may be used as identification of substances.

Radioactive decay occurs naturally, but artificial means may be used to concentrate and attenuate radiation, such as in a nuclear reactor. In such cases is it of primary interest to know the radiation being emitted, may it be for reasons of security or simply controlling power generation.

This lab report presents the results of three experiments conducted around EPFL's nuclear reactor CROCUS. The experiments consist of techniques for measuring neutron flux and identification of unknown radioactive materials using spectroscopy. Experiment procedures are briefly reviewed, however detailed explanation is given in another notice [1].

2 Determining the diffusion length of neutrons

The goal of this experiment is to calculate the diffusion length of neutrons and compare the results with a simple theoretical model. To do so, we will measure radial distribution of the neutron flow in the nuclear installation CARROUSEL. Numerical analysis of the results will provide an experimental value of the diffusion length, which can then be compared to the value predicted by simple model.

2.1 Experimental setup

2.1.1 The CARROUSEL

CARROUSEL is a cylindrical container, filled with water, containing a cylindrical Pu-Be source in its center. Neutrons are produced by using the Be as a target for the α particles produced by the Pu, following the reaction:

$$\alpha + {}^{9}\text{Be} \longrightarrow {}^{1}\ 2\text{C} + n$$

The water slows down all the neutrons to a thermal energy level of about 0.025 eV.

2.1.2 Detectors

We first determined which of the four available detectors was the most suitable for measurement at hand. The four detectors are gaseous ionization detectors, two with ^{3}He gas and two with BF_3 gas. These kind of detectors detect particles by measuring the total charge of the ions produced by the ionization of the gas in the detector due to the passing particle. To prevent the recombination of the ions before they reach the anode respectively the cathode, high voltage is applied between the two electrodes which induces a migration of the ions, which in turn induces a current.

In the ^{3}He gas, the neutrons produce protons following the reaction:

$$^{3}_{2}\text{He} + n \longrightarrow ^{3}_{1}\ \text{He} + p$$

One can show that the proton and the tritium have an energy of respectively 0.573 MeV and 0.191 MeV.

In the BF_3 gas, the neutrons produce alpha particles following the reaction:

$$^{10}_{5}B + n \longrightarrow ^{7}_{3}\ \text{Li}^{(*)} + \alpha$$

The asterix stands for the possible excited state the Li can be in with a 6% probability for thermal neutrons. This can be verified by comparing the height of the respective peaks. Again, one can show that the α particle and the lithium have an energy of 1.47 MeV respectively 0.84 MeV.

To measure the neutron flux, we measure the number of hits and time it took to get this number of hits. The flux is then proportional to ration of these two quantities because the surface of the detector is constant during the measurements.

2.2 Theoretical model

In this simple theoretical model, we assume the source to be a point. We then can resolve analytically the diffusion equation, which results of the combination of the neutron balance and Fick's law, given by:

$$D\nabla^2\Phi - \Sigma_a\Phi + Q = 0$$

where D is the diffusion coefficient, Σ_a the macroscopic absorption cross section and Q is a Dirac peak, i.e. it is equal to zero everywhere except at the origin where it is equal to Q neutrons per second. If we introduce the diffusion area $L^2 = \frac{D}{\Sigma_a}$ and assume a spherical symmetry of the problem, by plugging $u = r\Phi$ into the equation, we obtain the following equation in every point except the origin:

$$\frac{d^2u}{dr^2} - L^2u = 0$$

The solution of this equation is given by:

$$u(r) = Ae^{-r/L} + Be^{r/L}$$

We can take $B = 0$ because the flux has to go to zero when the radius goes to infinity. Fick's law tells us that

$$\boldsymbol{J} = -D\boldsymbol{\nabla}\Phi$$
$$\Rightarrow J = -D\frac{d\Phi}{dr} = \frac{AD}{r}\left(1 + \frac{r}{L}\right)e^{-r/L}$$

We implement the boundary condition: $\lim_{r\to 0} 4\pi r^2 J = 4\pi AD = Q$

$$\Rightarrow \Phi(r) = \frac{Q}{4\pi D}\frac{e^{-r/L}}{r} \tag{1}$$

The theoretical value of the diffusion length may be calculated using the following formula:

$$L_{th}^2 = \frac{D}{\Sigma_a} = \frac{1}{3\Sigma_t\Sigma_a} = \frac{1}{3N_{H_2O}^2\left(\sigma_a^O + 2\sigma_a^H\right)\left(\sigma_t^O + 2\sigma_t^H\right)}$$

where N is the number of water molecules per volume (i.e. $N = \rho\frac{\mathcal{N}_A}{M}$), σ_a^X is the absorbtion cross-section and σ_t^X the total cross-section of an element X. Using the following values:

$$N = 3.3368 \cdot 10^{22} cm^{-3} \quad \sigma_a^H = 0.3322b \quad \sigma_a^O = 1.9 \cdot 10^{-4}b \quad \sigma_t^H = 20.76834b \quad \sigma_t^O = 3.85b$$

the diffusion length is given by

$$L_{th} = 3.1501cm$$

2.3 Results

2.3.1 Detectors

The measured spectra with different detectors are shown in figure 1. The criteria for the most adequate detector is a high responsiveness in a small spectrum, i.e. the measured spectrum should present a high peak whose area is maximum. It can clearly be observed that detector one best meets the criteria (this can be made more apparent by looking at the spectrum with logarithmic y-scale, as is shown on figure 2. Therefore, detector 1 is used for the diffusion experiment.

2.3.2 Diffusion

By measuring the neutron flux (Φ) as a function of distance (r) to the source, it is possible to determine the diffusion length experimentally. Concretely, this is done by transforming formula (1):

$$\ln(r\Phi(r)) = \ln\left(\frac{Q}{4\pi D}\right) - \frac{r}{L}$$

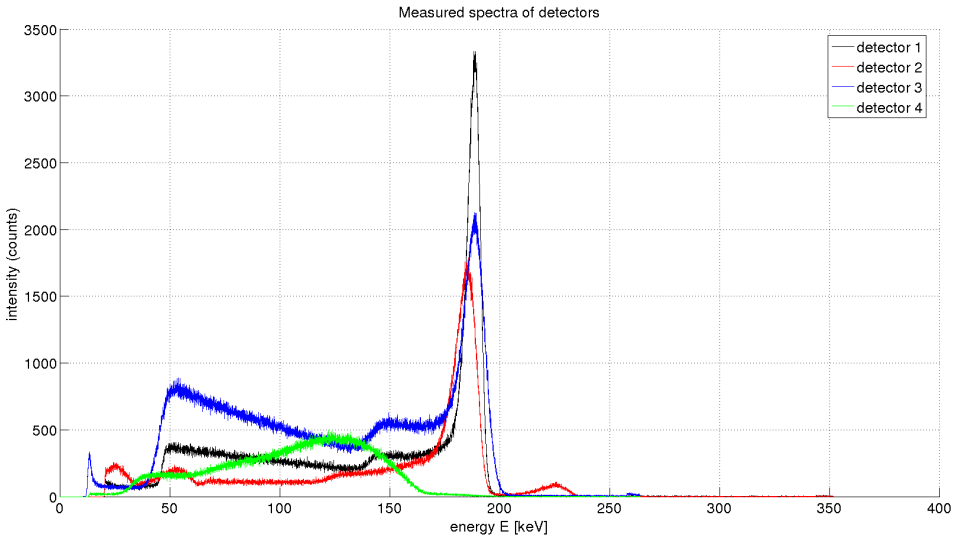

Figure 1: Spectra obtained with different detectors during the CARROUSEL experiment.

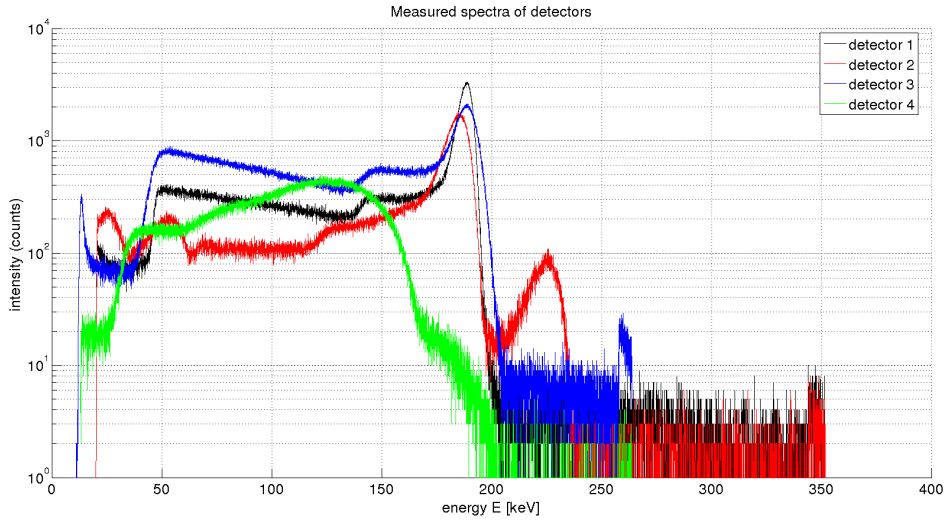

Figure 2: Spectra obtained with different detectors during the CARROUSEL experiment (log scale).

and plotting $\ln(r\Phi)$) as a function of r. The inverse of the plotted slope then gives an experimental value for the diffusion length L. The results of this procedure are given in figure 3. Furthermore, it can be seen that the constant term of the previous equation (1) contains the diffusion coefficient and therefore enables the calculation of an experimental value.

Using the fit applied to the obtained result, $f(x) = -0.1348x + 13.61$, experimental values of diffusion length and coefficient are given by:

$$L_{ex} = \frac{1}{0.1348} = 7.418cm$$

$$D_{ex} = \frac{Q}{4\pi} \exp(-13.61) = 0.84s^{-1}$$

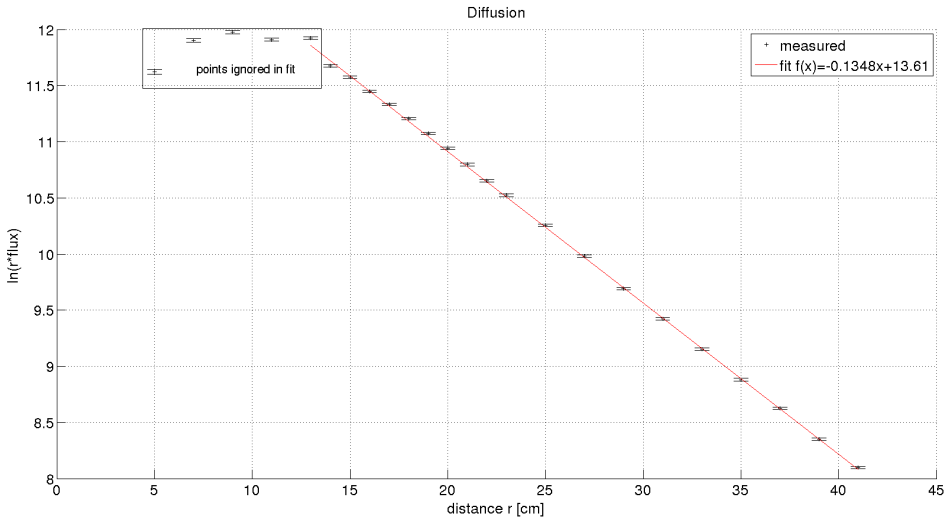

Figure 3: Measurement of diffusion length.

2.4 Discussion

The "winning" detector is a Helium 3 detector, this can be seen by the lack of a second peak, caused by the two elements in BF. However, the subsequent results obtained with it vary greatly with those predicted by the theoretical model. This may be due to a couple of reasons. First, in the theoretical model, the source is considered punctual, however in case of this experiment the source was of cylindrical form. Second, the neutrons must be thermal for the model to be applied, and this may not be the case.

Nevertheless, it should also be noted that the obtained results are linear, enabling a possible calibration of a detector. Thus, it may be concluded that this experiment showed the functioning and characteristics of a Helium 3 detector.

3 Identifying an unknown source using spectroscopy

The goal of this experiment is to identify an unknown radioactive source by identifying the emission peaks and their energy, as well as determining its half life. A secondary objective of the experiment is to measure the background noise, i.e. the ambient radioactivity and identify from which part of the natural chain the corresponding peaks come.

3.1 Calibrating the detector

To use the detector to identify the energy of given peaks, one needs to calibrate it first. This is done by measuring the spectrum of well known substance and adjusting the energy scale of the detector using the precisely known energy of the main peaks of the known substance. In this experiment, the calibration is done using an europium 152 source whose spectrum is given in figure 4.

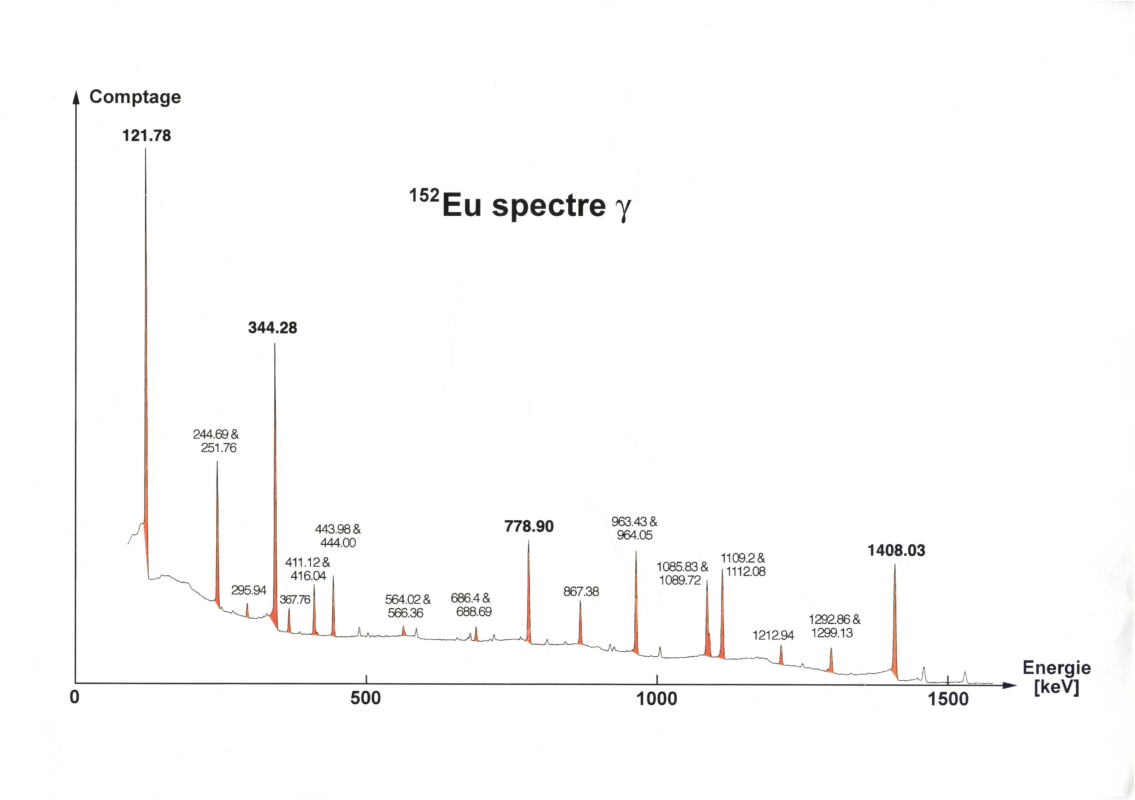

Figure 4: Spectrum of Europium 152.

3.2 Determining the efficiency of the detector

The efficiency of the detector for a given energy E is defined as the ratio of the number of detected photons and the number of emitted photons for that energy E. We can therefore write the following formula:

$$\eta(E) = \frac{\frac{N(E)}{t}}{A(t)p(E)}$$

where $N(E)$ is the number of detected photons and t the acquisition time, and $A(t)p(E)$ is the number of photons emitted at an energy E at the time t.

3.3 Measuring the background noise

We are constantly exposed to very small quantities of radioactivity in our everyday lives. In this experiment, by taking a measurement during a whole, we will see the different emission peaks and identify them. This "ambient" radioactivity is due to the disintegration chain of ^{238}U, ^{235}U and ^{232}Th, which are radioactive substances present in the Earth crust since its formation. These substances then disintegrate in other radioactive substances present on earth, which are then present in the soil, in rocks and the atmosphere. Another means of producing radioactive substances naturally is due to the interaction of the neutronic part of the cosmic radiation with the atmosphere. Examples are ^{12}C and ^{3}H.

3.4 Measuring the spectrum of the unknown source

By measuring the spectrum, we obtain two informations which permit us to identify the source. First, by determining the energies of the emission peaks and comparing those to tables, we obtain a first identification of the source. Second, by measuring the counts per second of the main emission a week, one can determine the half life, which is characteristic of a source, thereby determining the source.

Proposition 1. *If the activity of a source is known at a time t_1 and t_2, then the half life is given by:*

$$T_{1/2} = -\ln 2 \frac{t_2 - t_1}{\ln\left(\frac{A(t_2)}{A(t_1)}\right)}$$

Proof. We have

$$A(t_2) = A(t_1)e^{-\lambda(t_2-t_1)} \quad \Leftrightarrow \quad N(t_2) = N(t_1)e^{-\lambda(t_2-t_1)}$$

Also

$$\frac{N(t_2)}{N(t_1)} = \frac{1}{2} = e^{-\lambda T_{1/2}} \Rightarrow T_{1/2} = \frac{\ln 2}{\lambda}$$

Using this, one obtains:

$$\frac{A(t_2)}{A(t_1)} = e^{-\lambda(t_2-t_1)}$$

$$\Rightarrow \quad -\ln\left(\frac{A(t_2)}{A(t_1)}\right) = \frac{\ln 2}{T_{1/2}}(t_2 - t_1)$$

$$\Rightarrow \quad T_{1/2} = -\ln 2 \frac{t_2 - t_1}{\ln\left(\frac{A(t_2)}{A(t_1)}\right)}$$

□

3.5 Results

3.5.1 Calibration

Unfortunately, due to data corruption, our measured Europium spectrum was lost. However, before the writing of this report, we were able to calibrate our detector and thus all following measurements are accurate. As a side note, the measured spectrum closely resembled that of figure 4. Furthermore, it is possible to trace back the calibration by assuming knowledge of the unknown source and its spectrum and working back to find the calibration factors.

3.5.2 Efficiency of the detector

The initial activity A_1 of the Europium probe at a specific date is given by the manufacturer. Knowing its half-life ($T_{1/2} = 13.33 \pm 0.1$years) and the time passed since the initial activity measurement ($t_2 - t_1$), it is possible to calculate the probes current activity A_2 using proposition 1:

$$A_2 = A_1 \exp\left(-\ln 2 \frac{t_2 - t_1}{T_{1/2}}\right)$$

Therefore, by reading the number of neutrons detected (counts) for different energies of the spectrum, an efficiency curve may be established. This curve is shown on figure 5. It can be observed that the efficiency behaves exponentially, increasing with decreasing energies.

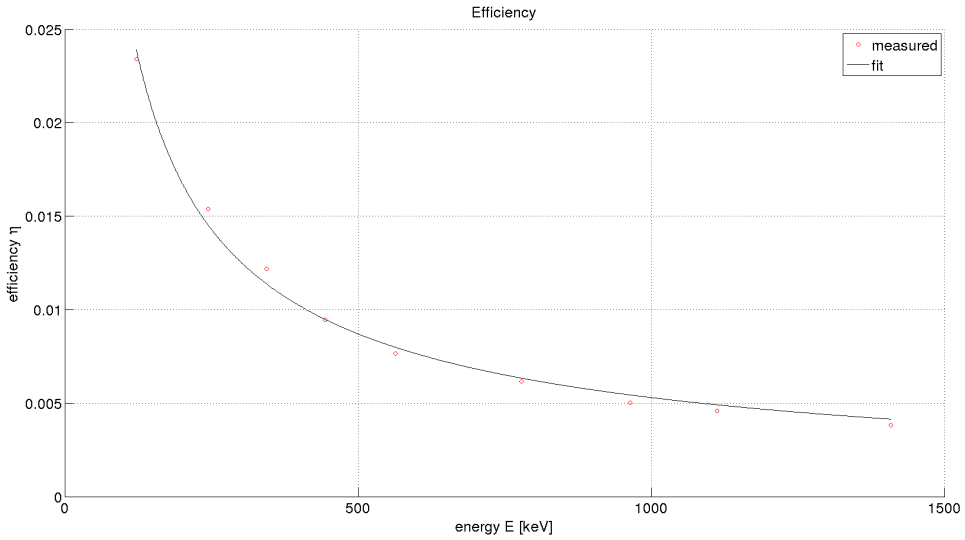

Figure 5: Obtained efficiency curve of detector.

3.5.3 Background noise

The spectrum of background noise (over 5 days) is shown on figure 6. The sources of the main spectral lines are given on the figure for a discussion on their meaning see section "Discussion".

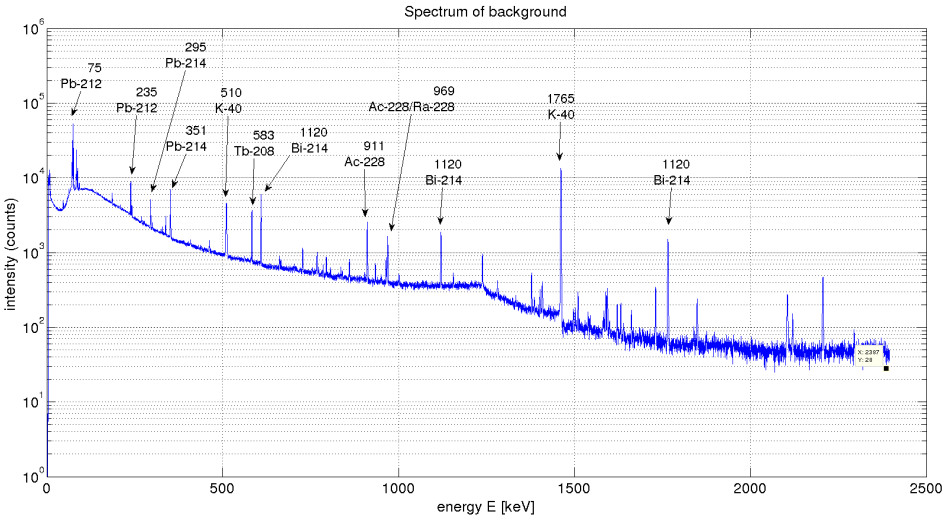

Figure 6: Spectrum of background. Most intense spectral lines highlighted: the peak's energy is given above, the source below.

3.5.4 Unknown source

The measured spectrum of the unknown source is given in figure 7. Three intense peaks can be observed at: 511, 1274 and 1787 keV. The first peak is due to electron–positron annihilation and

will be discussed later. The second and third peaks can be looked up in a table of nuclides and correspond to the emission rays of Na-22.

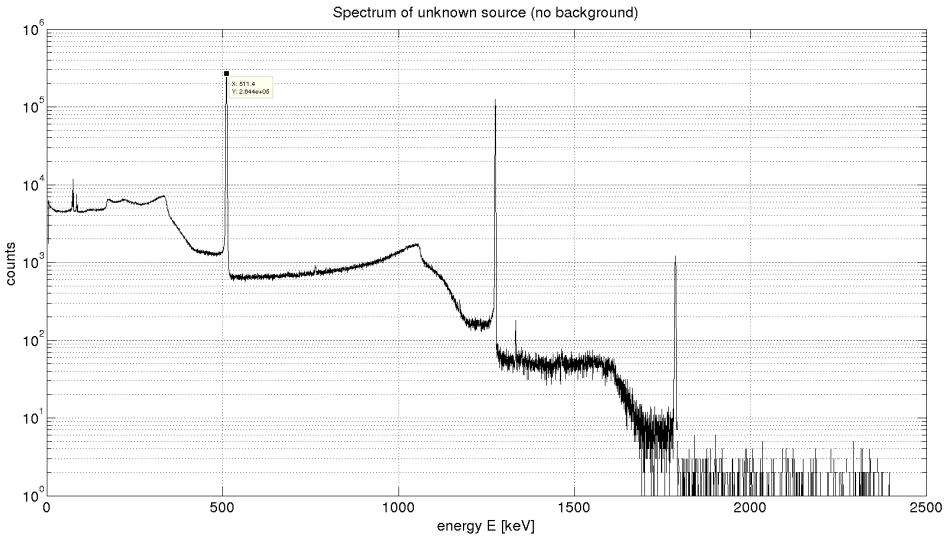

Figure 7: Spectrum of unknown source.

Furthermore, by examining a spectrum measured a week after the first, loss in activity may be observed and a half-life calculated. Take as an example the highest peak (excluding the 511 annihilation peak), situated at 1274 keV. The initial intensity of this peak is given by $N_1 = 7.87E5$, and the final intensity is given by $N_2 = 7.83E5$. Applying proposition 1 (assuming both measurements were of equal duration, activity may be substituted by counts), the half-life of the unknown probe can be calculated: $T_{1/2} \approx 2.6088$years. This value closely resembles the half-life of Na-22 given in tables [2].

It may therefore be concluded that the unknown source is in fact a Sodium isotope, namely Na-22.

3.6 Discussion

Regarding the efficiency of the detector, it should be noted that the efficiency is only valid for a certain distance to the detector. This is the reason all subsequent experiments were carried out at the same distance to the detector.

Considering the background spectrum, many interesting sources of natural radioactivity can be observed. Some, such as Actinium are themselves the product of another radioactive element (Radium in this case). One very interesting source is Potassium, an element that is also found in human bodies. Another, very distinctive peak is given at the energy 511keV, in fact this peak occurs in all measured spectra. The peak is due to electron-positron annihilation: at high energies (that the used detector cannot measure), positron electron pairs are created. However, as soon as it comes in contact with matter, antimatter annihilates. The energy of such an annihilation is released in the form of two photons each of the rest energy of an electron, namely 511keV.

The characteristic peaks of a spectrum allowed the successful identification of an unknown source. This immediately leads to possible applications. We were even presented with one such application in the lab, a device called “The Identifier”, capable of identifying the compositions of an unknown radioactive material.

4 Determining the angular distribution of the neutron flux in the CROCUS reactor

The goal of this experiment is to determine the absolute value of the axial neutron flux distribution in the CROCUS nuclear reactor, using the hypothesis that the geometry is cylindric and comparing that result to the prediction of a basic theoretical model.

4.1 Experimental procedure

Eight gold sheets are placed on a plastic bar, which is then inserted axially into the CROCUS. Depending on the position of the gold sheet inside the reactor, it will be more or less activated, thereby becoming radioactive. Due to this radioactivity, the sheets will emit γ rays. It is then possible to relate the number of photons to the neutron flux that irradiated the sheets, thereby measuring the axial neutron flux distribution of the reactor.

4.2 Calculating the necessary power

In this section, we calculate the power of the reactor necessary to have at least 10^5 counts of photons. Figure 8 shows the radioactivity of the gold sheets.

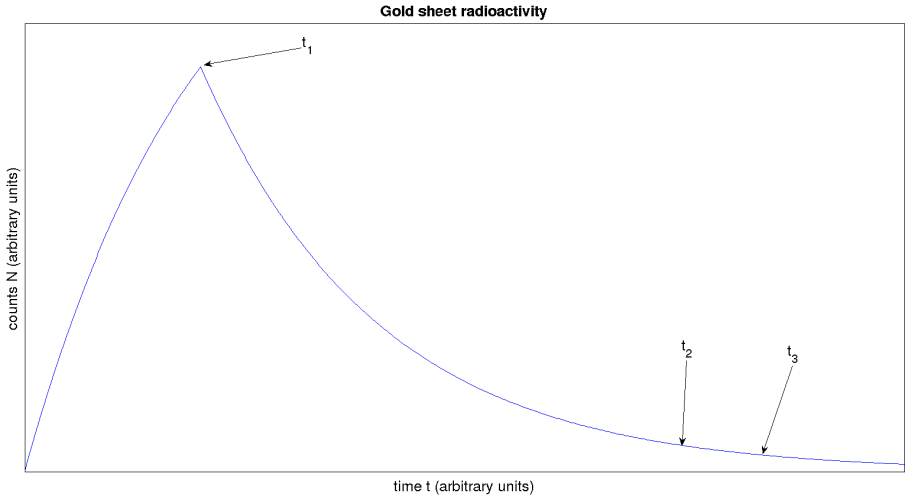

Figure 8: Radioactivity of the gold sheets.

First, the sheets are irradiated:

$$\frac{dN_1}{dt} = -\lambda N_1 + V\Sigma\phi$$

where V is the volume, Σ the macroscopic cross section and ϕ the flux. The solution of this equation is:

$$N_1(t) = \frac{V\Sigma\phi}{\lambda}\left(1 - e^{-\lambda t}\right)$$

Once the sheets are out the reactor, it is a simple disintegration process:

$$\frac{dN_2}{dt} = -\lambda N_2$$

The solution is:

$$N_2(t) = N_1(t_1)e^{-\lambda(t-t_1)} = \frac{V\Sigma\phi}{\lambda}\left(1 - e^{-\lambda t_1}\right)e^{-\lambda(t-t_1)}$$

The counts of photons is therefore given by:

$$N_\gamma = [N_2(t_2) - N_2(t_3)]p\eta \tag{2}$$
$$= \frac{V\Sigma\phi}{\lambda}\left(1 - e^{-\lambda t_1}\right)(e^{-\lambda(t_2-t_1)} - e^{-\lambda(t_3-t_1)})p\eta \tag{3}$$
$$= \frac{1}{\lambda}V\frac{N}{V}\sigma\phi\left(1 - e^{-\lambda t_1}\right)(e^{-\lambda(t_2-t_1)} - e^{-\lambda(t_3-t_1)})p\eta \tag{4}$$
$$= \frac{N\sigma\phi}{\lambda}\left(1 - e^{-\lambda t_1}\right)(e^{-\lambda(t_2-t_1)} - e^{-\lambda(t_3-t_1)})p\eta \tag{5}$$
$$\tag{6}$$

where $t_1 = 1$ hour is the irradiation time, $t_2 - t_1 = 6$ hours, $t_3 - t_2 = 20$ minutes is the time we measure the photon counts, $\sigma = 40.6$ barns is the microscopic cross section, p is emission probability of the the photon at 412 keV and η is the efficiency of the detector at this energy (determined earlier).

A constant that is give to us is the flux-power ratio in the reactor core $\alpha = 2.53 \cdot 10^7 \frac{n}{cm^2 s}\frac{1}{W}$. We therefore obtain the formula for the power:

$$P = \frac{\phi}{\alpha}$$

In our case, for a minimal photon count of 10^5, this yields a power of:

$$P \approx 5.97 \pm 0.31\text{W}$$

This power is acceptable since the reactor well capable of supplying more than 10W power.

4.3 Theoretical model

In this section, we solve the diffusion equation for a cylindrical geometry. The diffusion equation then is:

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\phi}{\partial r}\right) + \frac{\partial^2\phi}{\partial z^2} + B^2\phi = 0$$

We do a separation of variables $\phi = Z(z)R(r)$, thereby obtaining the following equation for the z-variable:

$$\frac{d^2Z}{dz^2} = -\lambda Z$$

The boundary conditions we impose are that $Z(H/2 \pm d) = 0$ where H is the height of the reactor. d is a supplementary distance to the reactor height where the flux is effectively zero. The solution is therefore given by:

$$Z = A\cos(\sqrt{\lambda}z) + B\sin(\sqrt{\lambda}z)$$

Implementing the boundary conditions, then constrains λ:

$$\sqrt{\lambda} = \frac{\pi(2k+1)}{2(\frac{H}{2}+d)}$$

$$\Rightarrow Z(z) = A\cos\left(\frac{\pi}{H+2d}z\right) \tag{7}$$

Similarly, the flux dependence in radius can be calculated (the solution are Bessel functions). However, the radius dependency is not of interest in this report.

4.4 Results

After being irradiated in the CROCUS reactor, the characteristics and activity of the gold probes are given in table 1. Reversing formula (6), it is then possible to determine the neutron flux in the reactor. The resulting flux distribution is shown in figure 9. The shown fit is given by the equation:

$$Z(z) = 2.807 \cdot 10^8 \cos\left(0.02977 z\right) \tag{8}$$

and has an r^2-value of 0.9985. Given the fit, according to equation (7), the supplementary distance d is given by:

$$0.02977 = \frac{\pi}{H + 2d}$$

$$d = \frac{1}{2}\left[\frac{\pi}{0.02977} - H\right] = \frac{1}{2}\left[\frac{\pi}{0.02977} - 1\right] \approx 52\text{cm}$$

This means that about 2cm above and below the fuel bars the neutron flux should be negligible.

Probe	**Height z [cm]**	$\frac{\Delta z}{z}$	**Mass m [g]**	$\frac{\Delta m}{m}$	**Counts N_γ (411.77keV)**	$\frac{\Delta N_\gamma}{N_\gamma}$
1	-25.4	0.0079	0.0303	0.00330	9.08E+004	0.0033
2	-15.4	0.0130	0.0324	0.00309	1.14E+005	0.0030
3	-5.4	0.0370	0.0331	0.00302	1.26E+005	0.0028
4	4.6	0.0435	0.0312	0.00321	1.17E+005	0.0029
5	14.6	0.0137	0.0301	0.00332	1.03E+005	0.0031
6	24.6	0.0081	0.0304	0.00329	8.06E+004	0.0035
7	34.6	0.0058	0.0478	0.00209	8.37E+004	0.0035
8	-35.4	0.0056	0.0484	0.00207	1.02E+005	0.0031

Table 1: Results obtained

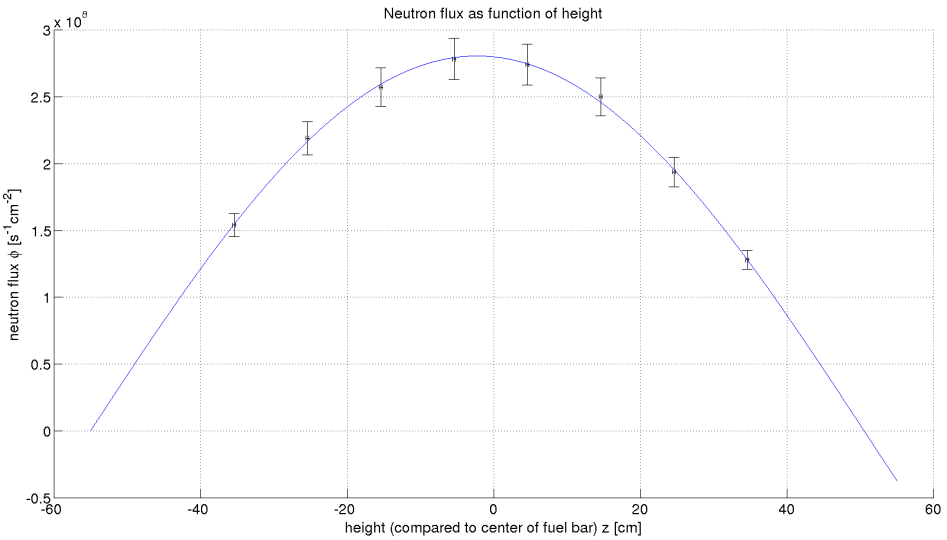

Figure 9: Height-distribution of neutron flux in reactor.

4.5 Discussion

This experiment showed a way to measure neutron flux in a nuclear reactor. The results seem satisfying as indicates the high goodness of fit. However, it can be observed that the flux distribution is not perfectly centered but slightly shifted to the left. This is because the reactor was

not completely filled (to 1m) with water, but a couple of centimeters below. This asymmetry in the centering however is barely noticeable and it can be concluded that the method of using gold plates works.[1]

[1] It may be a bit cumbersome and expensive to use in real nuclear power plants however!

5 Conclusion

This report gave three examples of the application of gamma spectroscopy and neutron flux. Methods were described on how to identify an unknown radioactive source and how to measure neutron flux. According to the obtained results, both methods were verified.

However, this report only presented a fraction of the technologies related to nuclear physics, and much more could be explored. As an example, the observations made on neutron flux could further be used in the planning of a nuclear reactor.

References

[1] D. Bertollo, P. Frajtag, and G. Girardin, *Resonance Magnetique Nucleaire*. EPFL, September 2011.

[2] "Table of Sodium Isotopes." http://en.wikipedia.org/wiki/Isotopes_of_sodium, May 2013.

List of Figures