Experiment 253: Radioactive Activation and Decay

Aims

1. To verify the exponential nature of radioactive decay.
2. To determine the half-life of Indium 116.
3. To determine the build-up rate of activity of an Indium sample in a slow-neutron flux.
4. To fit the data using PYTHON and the function `phylab.regress`.

Note: This experiment should not be attempted until Experiment 251 on Radioactivity has first been completed.

References

1. D. de Soete et al., "Neutron Activation Analysis", Wiley Interscience.
2. M.N. Thompson and J.M. Taylor, "Nuclear Instruments and Methods", 37 (1965) pp 305 – 308. This reference gives measured spectra of neutron sources.
3. E. Rutherford, J. Chadwick and C.D. Ellis, "Radiations from Radio-active Substances". Cambridge University Press 1930. This classic old text is worth browsing through, especially Chapter 1.

Introduction

In this experiment a sample of Indium is made radioactive by irradiation with slow neutrons. Natural Indium, which is a heavy and very soft metal, not unlike lead, consists of about 96% ^{115}In and 4% ^{113}In. When exposed to a neutron flux, one of the reactions which occurs is the following:

$$^{115}_{49}\mathrm{In} \; + \; \mathrm{n} \; \rightarrow \; ^{116}_{49}\mathrm{In}$$

followed by the decay

$$^{116}_{49}\mathrm{In} \; \rightarrow \; ^{116}_{50}\mathrm{Sn}^{*} \; + \beta^{-}$$

The isotope ^{116}In has a metastable state with a half-life of 54 minutes and a ground state with a half-life of 14 seconds (see Figure 1). In this experiment it is the gamma rays from the excited states of the ^{116}Sn nucleus which are detected.

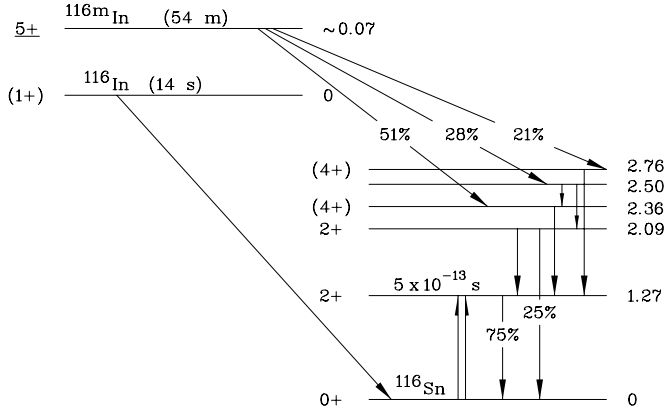

Figure 1: ^{116}In decay scheme.

The irradiation source is a mixture of Americium and Beryllium which produces a broad spectrum of neutrons of a few MeV energy accompanied by gamma rays. Its behaviour is discussed in the Appendix.

In this experiment the 54 minute half-life is measured over about three half-lives and the results analysed by least-squares fitting using the PYTHON function `regress` on one of the computers. The function file takes the form: `param,error = regress(x,y,err,n)`. In addition, by irradiating six samples for measured time intervals, the rate of build-up of activity can be measured and compared with theory.

Theory

Consider a system containing a large number of identical nuclei at some initial time. The nuclei decay at the decay rate λ. We shall calculate the number of undecayed nuclei present at some later time. If there are N undecayed nuclei at time t, then the number decaying in the following time interval dt can be written as dN. Since λ is the probability that a particular nucleus will decay in one second, λdt is the probability that it will decay during the time dt, and $N\lambda dt$ is the probability that any one of the N nuclei will decay in that interval. Thus the average number of decaying nuclei is:

$$dN = -N\lambda dt \tag{1}$$

where the minus sign accounts for the fact that N decreases with time. Rearranging the terms and integrating, we obtain:

$$\frac{dN}{N} = -\lambda dt$$
$$\int_{N(0)}^{N(t)} \frac{dN}{N} = -\lambda \int_0^t dt = -\lambda t$$

that is

$$\ln N(t) - \ln N(0) = \ln \frac{N(t)}{N(0)} = -\lambda t$$

or

$$\frac{N(t)}{N(0)} = \exp(-\lambda t)$$

so

$$N(t) = N(0) \exp(-\lambda t) \tag{2}$$

In expression (3), $N(0)$ is the number of undecayed nuclei at the initial time 0 and $N(t)$ the number of undecayed nuclei at the later time t.

Since the calculation involves probabilities, its results are correct only on the average; but since the number of nuclei involved is usually very large, fluctuations from the average are small (see later). Figure 2 is a plot of this exponential decay law.

The lifetime (or mean life) T, characteristic of the decay, is the average time a nucleus survives before it decays. You should convince yourself that $T = 1/\lambda$ by integrating the decay law. Thus in one mean life the number of nuclei decreases by a factor e. The half-life, on the other hand, is the time required for the number of undecayed nuclei to decrease by a factor of 2. The relationship between the two times is then obtained from:

$$T_{1/2} = \ln 2\, T = 0.693\, T \tag{3}$$

The previous discussion applies to the situation after the sample has been removed from the slow-neutron source. While the sample is being activated it is exposed to a constant neutron flux. If the number of ^{115}In nuclei is not significantly depleted by this neutron flux, there will be a constant rate of production of radioactivity as well as an exponential decay. This may be described by the following differential equation:

$$\frac{dN}{dt} = R - \lambda N \tag{4}$$

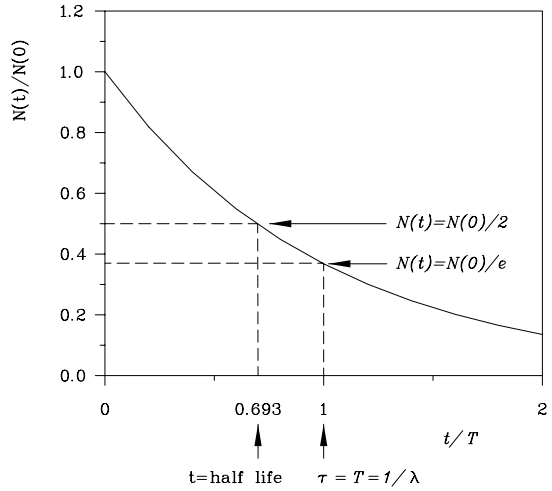

Figure 2: Plot of exponential decay.

R is the constant rate of production of radioactive nuclei and is proportional to the neutron flux. The solution of this equation is:

$$N = \frac{R}{\lambda}\left(1 - \exp\left(-\lambda t\right)\right) = A\left(t\right)$$

where we have introduced the conventional notation of $A\left(t\right)$ for the activity of the sample. Here, t is the time measured from the start of the irradiation, and the activity $A\left(t\right)$ is assumed to be zero at $t = 0$. Clearly this represents an exponential build-up to a maximum value R/λ when t is much greater than the mean life $1/\lambda$. If this maximum activation is labelled A_s then we may write

$$A\left(t\right) = A_s\left(1 - \exp\left(-\lambda t\right)\right) \tag{5}$$

In this experiment this build-up of activity is measured by placing six samples in the neutron flux, removing them one at a time at measured times and measuring the activity. Figure 3 then shows the form of the sample activity as a function of time.

For times greater than t_1 the activity is given by

$$A\left(t\right) = A_s\left(1 - \exp\left(\lambda t_1\right)\right)\exp\left(-\lambda\left(t - t_1\right)\right)$$

Clearly it is not possible to measure the sample activity at t_1, firstly since there will be a time delay between removing the source from the neutron flux and starting counting, and secondly because an activity at an

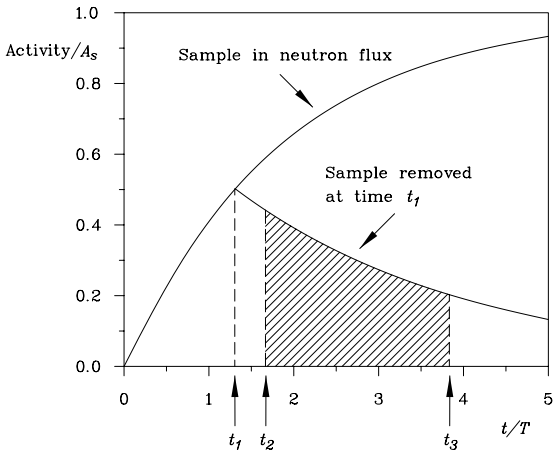

Figure 3: Build-up and decay of sample when placed in neutron flux at time $t = 0$ and removed at time t_1. The activity acquired by the sample may be determined by counting it from t_2 to t_3.

instant of time cannot be made with good statistics. Instead, one must make measurements from t_2 to t_3, during which time the sample may have decayed by an appreciable amount. The total number of counts C expected will therefore be given by:

$$C = \varepsilon \left[A\left(t_3\right) - A\left(t_2\right) \right] + B\left(t_3 - t_2\right) \tag{6}$$

where ε is the efficiency of the counting system and B is the background count rate. If $t_3 - t_2$ is short enough for $A\left(t_3\right) \approx A\left(t_2\right)$ then this is well approximated by:

$$C = \varepsilon \, \lambda \, A\left(t_2\right) \left(t_3 - t_2\right) + B\left(t_3 - t_2\right) \tag{7}$$

Procedure

The irradiation apparatus is located in the Radiation and Chemical Store (room G04D). Both the neutron source and sample 0 should be in place. The neutron source is on the end of the plastic rod sticking out of the top. Sample 0 should be in the central hole at the front. Samples 1 – 6, which contain less Indium than sample 0, should be with the detection apparatus. They should have been removed from the irradiation apparatus at least 24 hours previously so that they are no longer radioactive.

IMPORTANT!

At the end of EVERY lab session you MUST:

1. **Leave sample 0 in the central hole of the neutron source.**
2. **Leave samples 1 – 6 in the box by the detection apparatus.**

The detection apparatus should be set up according to the block diagram in Figure 4.

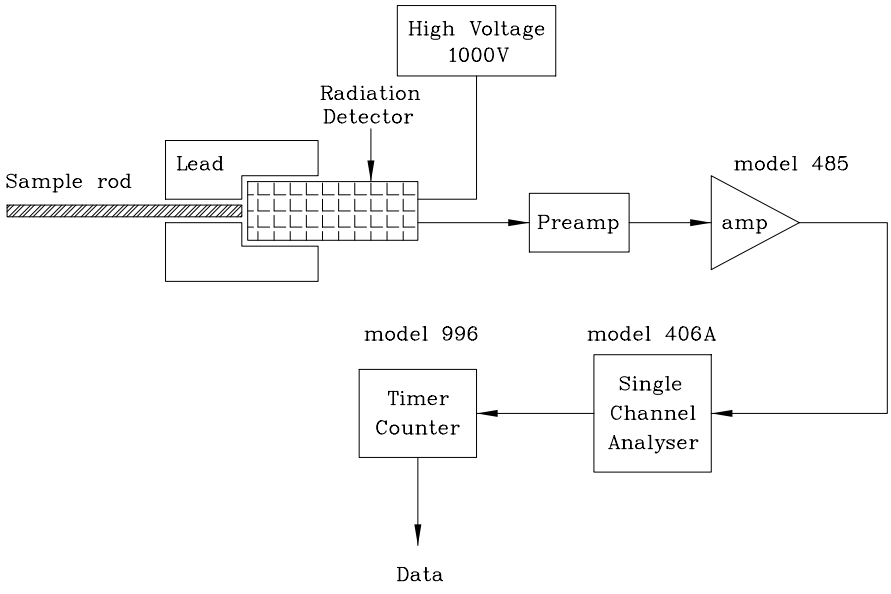

Figure 4: Block diagram of radiation detection apparatus.

Gain settings should be as follows:

Preamplifier gain:	maximum		
435A amplifier	polarity: NEG	coarse gain: 4	fine gain: 5.00
406A single channel analyser mode:	mode: NOR	lower level: 0.80	upper level: 10.0
Photo-multiplier high voltage:	1000V (3.33 on scale)		

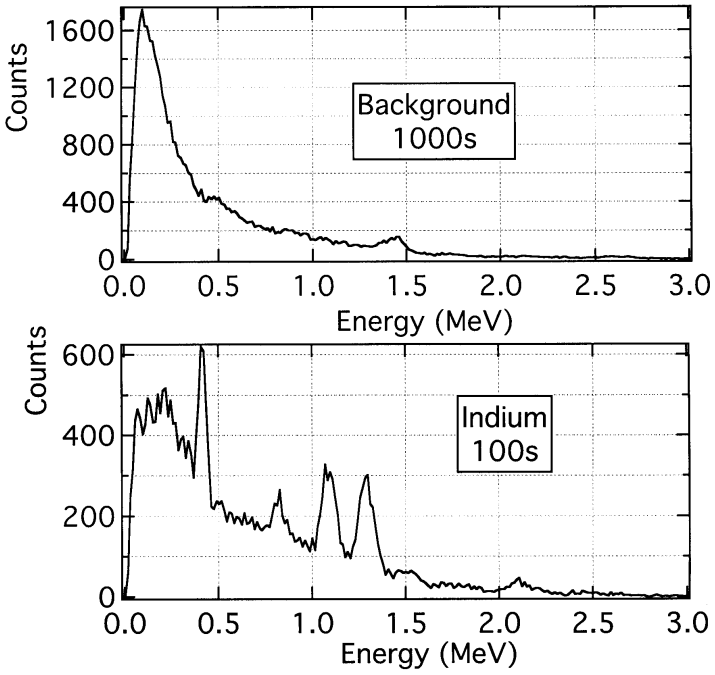

Figure 5: Gamma ray spectra

You do not need to know the details of how the detection apparatus works. If you are curious, here is a quick run-down. Gamma rays from the activated sample are detected by a sodium iodide crystal and photo-multiplier combination. Output pulse height depends upon gamma ray energy. Figure 5 shows gamma ray energy from the indium taken for 100 s, and from room background, taken for 1000 s. The single channel analyser is set to choose all gamma rays above about 0.4 MeV energy. If you suspect this setting has been changed, ask a demonstrator to set it up for you using a ^{22}Na source. Although gain settings are not critical, they should not be changed once the experiment is started. The detected gamma rays are counted by the timer counter (scaler) for a preset time.

Part 1: Half-life measurement

(1) Set the timer scaler to 100 seconds. Use a time base setting of 0.01 s. Toggle the display to preset and set up the preset display for 1×10^4. Measure the background count rate with no sources nearby. Repeat this several times and take the mean. Calculate the standard deviation in the mean (error) by taking the square-root of the sum, and then dividing by the number of measurements made. Note that the timer and counter will automatically recycle after a dwell time of 10 s during which time the data must be recorded unless "stop" is pressed.

(2) Remove sample 0 from the irradiation apparatus and put it in the lead shield so that it is just touching the detector face. The radioactive Indium actually has two modes of decay. One has a half-life of about an hour and the other about 14 seconds. If you wait a minute or two before taking measurements the 14-second component will not affect your results.

(3) Measurements with a counting time of about 100 seconds should be made at regular intervals of about 5 minutes over a period of at least two hours. The time at which each measurement is made should be determined with an accurate clock or stop-watch.

(4) Tabulate your results, subtract the background and calculate the standard deviation according to the method described in Experiment 251. Make use of the spare time you have between measurements to

plot the results as you go on semi-log paper. Remember to put error bars on your graph.

(5) Use equation (3) and the slope of the graph to determine the half-life of ^{116}In. This is possible without knowing the numerical value of ε. You should do this first by drawing the best straight line by eye through the points and also by using `regress`.

Part 2: Radioactive build-up

The theory makes it clear that since sample 0 had to be exposed to the neutron source for at least 24 hours, its activity must reach the saturation value in that time. For exposure times less than the half-life the activity will be correspondingly less.

(6) To measure the build-up curve, measure first the background with each of the samples 1 – 6 in position touching the face of the detector. They should give results which do not differ statistically from each other. All of these samples should have been away from the neutron source for at least 24 hours.

(7) Now put all the samples 1 – 6 into their corresponding holes in the irradiation apparatus. Note the time when this is done. The samples should be inserted into the ends of the brass tubes. As the tubes are closed off with thin brass shim, do not ram them in roughly or you may get a wet surprise!

(8) After about 20 minutes remove sample 1. Note the time of removal and also start a stop-watch. Put the sample in the detection apparatus and start counting precisely 1 minute after removal from the source. Count for 15 minutes and tabulate the result.

(9) After 40 minutes from the start of irradiation remove sample 2 and proceed as before. In this way you will have established six points on the buildup curve. Plot these results in a way that enables you to compare them with theory. Note that the samples are not all of the same mass. You will therefore need to correct for this before plotting your results.

Questions

These must all be answered as part of your write-up.

1. The radioactive portion of the source is ^{241}Am. This was manufactured on 26 February 1976 and has a half-life of 458 years. It decays via a long chain of alpha emitters to a stable nucleus; however the first daughter product ^{237}Np, has a half-life of 2.2×10^6 years. This means that nearly all the energy released over a short period of time is due only to the decay of the ^{241}Am. From a table of nuclear masses determine the mass difference between ^{241}Am and ^{237}Np plus an alpha particle. Assume that all this energy is absorbed by the water moderator and calculate the rate of temperature rise of the water assuming no heat loss. Take the activity of the ^{241}Am as 300 MBq.
2. From equation (8) determine the ratio of the neutron kinetic energy after collision with an oxygen nucleus to that before for the situation where the energy loss is a maximum. Calculate the minimum number of collisions with ^{16}O needed to reduce the neutron energy to less than 1% of its original value.

List of Equipment

1. Am–α-Be source and water moderator.
2. Seven Indium samples mounted in perspex rods.
3. NaI crystal and photo-multiplier tube detector assembly.
4. Lead shield for detector.

5. Scintillation counter pre-amplifier
6. ORTEC 485 main amplifier
7. ORTEC 406A single channel analyser
8. ORTEC 996 timer and counter
9. High voltage power supply

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February 2003.

This version: October 1, 2014.

APPENDIX

Figure 6 shows the measured neutron spectrum (Ref. 2). The Indium activation has a higher probability for slow and thermal neutrons than for those of a few MeV energy so they are slowed down by means of a moderator consisting of a 40 cm cube of ordinary water. The action of a moderator can be understood in terms of the classical theory of collisions. If a billiard ball makes a head-on elastic collision with another ball of equal mass it loses all its energy in that one collision. Similarly, the maximum energy loss for a neutron encountering a proton (which has approximately the same mass) is enough to reduce it to thermal velocities.

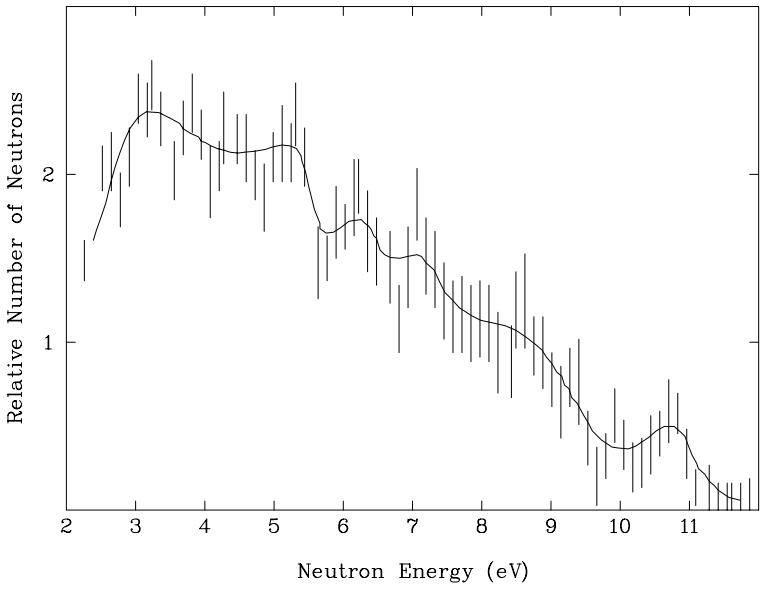

Figure 6: Spectrum of neutrons from Am-α-Be source.

The ratio of the kinetic energy T' of the neutron after a collision with a nucleus with mass M_t to that of its original kinetic energy T is given by:

$$\frac{T'}{T} = 1 - \frac{4M_n M_t \cos^2\phi}{(M_n + M_t)^2} \tag{8}$$

where M_n is the neutron mass and ϕ is the angle of the recoiling particle M_t. A head-on collision corresponds to $\phi = 0$. From this it is easy to see that water should be an efficient moderator since it contains a lot of hydrogen nuclei. Similarly, paraffin wax would also be a suitable material. You may therefore wonder why these simple substances were not used as moderators in the earlier nuclear reactors. This is because elastic

scattering is not the only nuclear event that can occur between a neutron and a proton. There is a non-zero probability that the neutron and the proton will coalesce to form deuterium or heavy hydrogen. This removes a neutron from the system thereby reducing the flux of thermal neutrons produced.

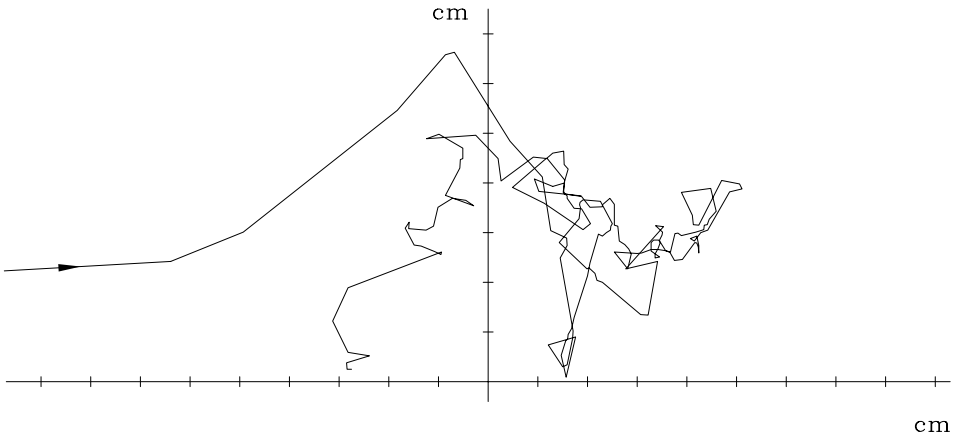

Figure 7: Typical motion of a 4 MeV neutron during thermalisation.

To get an idea of the motion of a neutron during thermalisation see Figure 7. The neutron is incident from the left with about 4 MeV energy. Its path has been calculated by a Monte Carlo method and plotted by computer. The medium in which the neutron is moving is wax which has the average composition of human tissue and therefore consists mainly of H, C, N and O.