LABORATORY EXERCISE IN REACTOR PHYSICS (II)

DETERMINATION OF THERMAL FLUX FROM A ^{252}Cf NEUTRON SOURCE

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INTRODUCTION

The objective of this exercise is to determine the thermal neutron flux at a certain distance from an external neutron source (^{252}Cf). This is done by activating (through neutron capture) two gold foils, of which one is covered by cadmium. Taking advantage of the coincidence technique for measuring the radioactivity of the gold foils an absolute value of the activity can be obtained.

THEORY OF SPONTANEOUS FISSION

Spontaneous fission is a form of radioactive decay. If we divide a heavy nucleus into two fragments we can think of spontaneous fission as a Coulomb repulsion effect in which two fission fragments are preformed inside a parent nucleus. It is the result of a competition between the nuclear binding force and the Coulomb repulsion force. It becomes increasingly important for heavy nuclei because the repulsive Coulomb force increases with Z^2 while the nuclear binding force increases as A. In the process, a heavy nucleus splits into two lighter nuclei and a few neutrons are released. The reaction may be written:

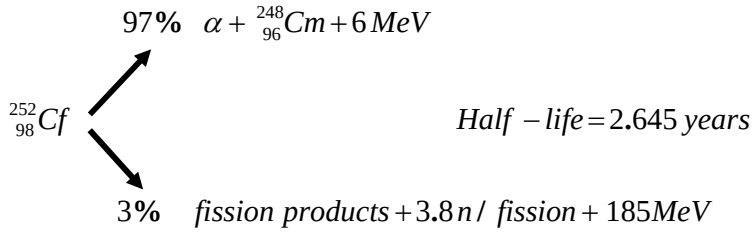
$$ {}^{A}_{Z}X \rightarrow {}^{A1}_{Z1}X_1 + {}^{A2}_{Z2}X_2 + \nu \cdot n $$

where A is the mass number, Z is the atomic number, and ν is the number of neutrons emitted. The fission fragments, X_1 and X_2 that are formed are extremely rich in neutrons. They compensate this neutron excess through emission of a few neutrons immediately after the fission process (prompt neutrons). The typical decay time may be 10^{-15} s or less. In general, spontaneous fission is a process with a long half-life, and since α-decay competes with it, spontaneous fission is often only a weak decay branch. For elements with atomic number lower than Z = 90, the fission potential barrier is so high that no spontaneous fission occur. The fission barrier prevents the nuclei from separating into two fragments, and the decay probability is infinitely small. With increasing Z^2/A, however, spontaneous fission becomes more common, i.e. the half-life for this decay process decreases. For ^{230}Th it is $1.5 \cdot 10^{17}$ years; for ^{244}Cm, $1.4 \cdot 10^{7}$ years; for ^{252}Cf, 85.5 years; and for ^{256}Fm it is 2.5 hours.

CALIFORNIUM-252 AS A NEUTRON SOURCE

A common source of neutrons is the spontaneous fission of ^{252}Cf. Neutrons are emitted directly in the fission process, at a rate of about 3.8 neutrons per fission. The effective half-life of 252Cf is 2.65 years. Spontaneous fission occurs in about 3% of the decays, α-decay accounts for the rest, and the neutron production rate is $2.3 \cdot 10^{12}$ neutrons/s per gram of ^{252}Cf. The neutron energies are characteristic of fission, a continuous Maxwellian distribution with an average energy of 2 MeV. A summary of radioactive decay properties is presented below:

Half-life (effective):	2.65 years
Half-life (spontaneous fission):	85.5 years
Half-life (α-decay):	2.73 years
Neutron emission:	$2.3 \cdot 10^{12}$ neutrons/s per gram
Average neutron energy:	2 MeV
Specific activity:	$2.0 \cdot 10^{13}$ Bq per gram



Spontaneous fission neutron sources have many useful applications due to their simplicity, reliability, and small size. ^{252}Cf is convenient since it may provide moderate neutron intensity over a sufficiently long half-life. Low rate of heat emission, γ-radiation, and helium production allow fabrication of simple and small ^{252}Cf neutron sources.

The particular Californium-252 source used in this exercise is contained in a palladium alloy and has the shape of a pellet. The pellet is doubly encapsulated in welded stainless steel capsules. The capsule is mounted in a metal rod that is kept under water.

EXPERIMENT

The ^{252}Cf neutron source is submerged in a water tank. A gold foil is attached to the surface of two plastic rods. The plastic rods are mounted in the tank such that the gold foils are located at a certain distance away from the ^{252}Cf-source. One gold foil is shielded from thermal neutrons by covering it with cadmium plates. The gold foils are exposed for a few days, leading to capture of neutrons in the gold foils, which thereby become activated:

$$n + {}^{197}Au \rightarrow {}^{198}Au \qquad \text{“neutron capture in } {}^{197}Au\text{”}$$

The neutron capture cross-section of Au-197 is presented in Figure 1. Cadmium absorbs almost all neutrons with energy lower than 0.4 eV (see Figure 2), but is nearly transparent to higher energy neutrons. The difference of the flux in the foils with and without Cadmium cover gives the thermal flux.

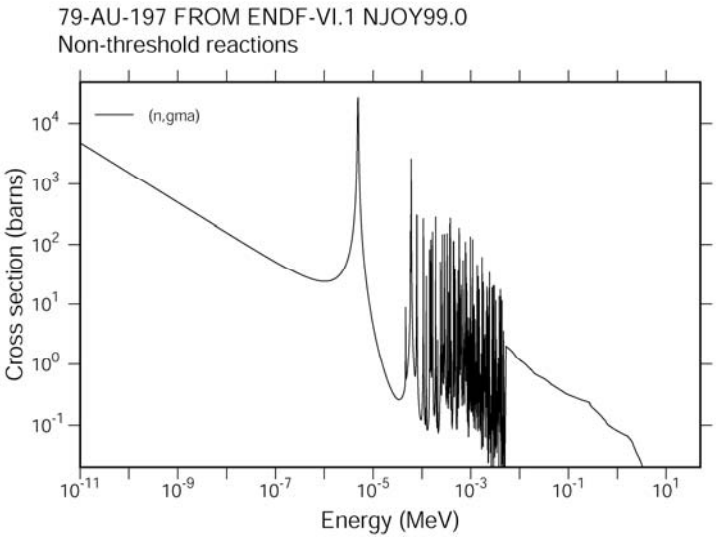


Figure 1. Neutron capture cross-section in Au-197

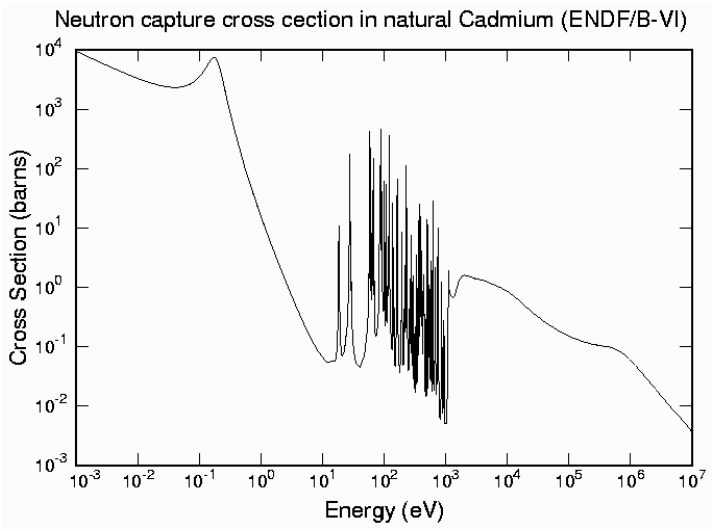


Figure 2. Neutron capture cross-section in natural Cadmium.

ACTIVITY IN THE IRRADIATED MATERIAL

During irradiation, the number of activated nuclei N_V per unit volume of an isotope is given by the following differential equation:

(1) $$\frac{dN_V}{dt} = \Sigma_c \phi(r) - \lambda N_V(t)$$

$N_V(t)$	number of activated nuclei per unit volume at time t [nuclei/cm^3]
Σ_c	macroscopic capture cross-section of ^{197}Au [cm^2/cm^3]=[cm^{-1}]
λ	decay constant of ^{198}Au [s^{-1}]
$\phi(r)$	flux at position r [n/cm^{2}s]
$\Sigma_c\phi$	production rate of ^{198}Au through neutron capture in Au197 [n/cm^{3}s]
$\lambda N_V(t)$	decay rate of ^{198}Au [n/cm^{3}s]

Assuming that the gold foil is placed in the neutron flux during time t_1, the number of activated nuclei at the end of the irradiation is obtained by solving Eq. (1). The solution is:

(2) $$N_V(t_1) = \frac{\Sigma_c \phi}{\lambda}\left(1 - e^{-\lambda t_1}\right)$$

$N_V(t_1)$	number of activated nuclei (Au198) per unit volume at time t_1 [nuclei/cm^3]
t_1	activation time [s]

At the end of the irradiation the activated nuclei continues to decay. The number of activated nuclei that remain after cooling time t_2 is given by the general equation for simple radioactive decay:

(3) $$N_V(t_2) = N_V(t_1)e^{-\lambda t_2}$$

$N_V(t_2)$	number of activated nuclei per unit volume at time t_2 [nuclei/cm^3]
t_2	cooling time [s]

and during measuring time t_3, a fraction $\left(1 - e^{-\lambda t_3}\right)$ of the isotopes decays. Thus the number of decays per unit volume during t_3 is:

(4) $$N_V(t_3) = \frac{\Sigma_c \phi}{\lambda}\left(1 - e^{-\lambda t_1}\right)e^{-\lambda t_2}\left(1 - e^{-\lambda t_3}\right)$$

$N_V(t_3)$	number of nuclei per unit volume that decayed during time t_3 [nuclei/cm^3]
t_3	measuring time [s]

Multiplying equation Eq. (4) with the volume of the gold foil, V, one gets the number of nuclei that decayed in the gold foil during time t_3. This number is calculated by:

(5) $$N(t_3) = \frac{V\Sigma_c \phi}{\lambda}\left(1 - e^{-\lambda t_1}\right)e^{-\lambda t_2}\left(1 - e^{-\lambda t_3}\right)$$

$N(t_3)$	number of nuclei in the gold foil that decayed during time t_3 [nuclei]

COINCIDENCE COUNTING

The coincidence counting technique is a simple method for measurements of absolute disintegration rates (Walther Bothe was awarded the Noble Prize in Physics in 1954 for the discovery of the coincidence method). It can be applied to the decay of ^{198}Au. When ^{197}Au captures a neutron it forms unstable ^{198}Au.

As a reminder, the β-decay process, in which a neutron is converted to a proton, is repeated.

$$n \rightarrow p + e^{-} + \overline{\nu}_e \qquad \textit{"General } \beta\textit{-decay process"}$$

In the case of β-decay of ^{198}Au it means that ^{198}Au is converted into ^{198}Hg and an electron and an antineutrino is emitted from the nucleus:

$$^{198}Au \rightarrow {}^{198}Hg + e^{-} + \overline{\nu}_e \qquad \textit{"}\beta\textit{-decay of } {}^{198}Au\textit{"}$$

^{198}Au decays in a β - γ cascade, see Figure 3, with the half life $T_{1/2}$ = 2.6943 days.

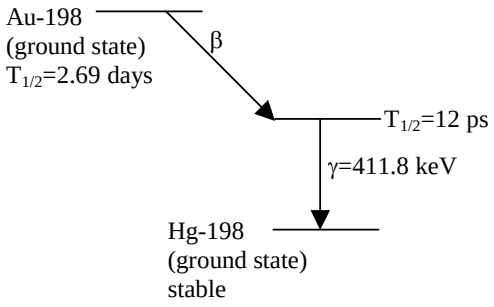


Figure 3. Decay scheme of Au-198

This decay scheme is measured with a coincidence detector in a set-up according to the following figure:

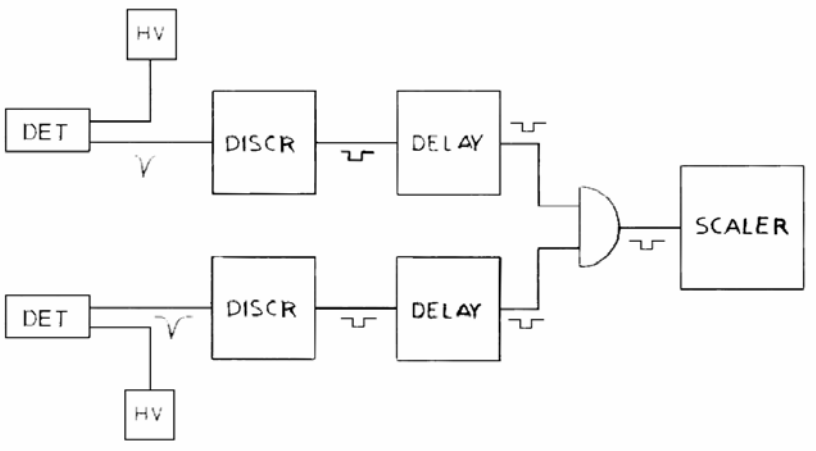


Figure 4. Schematic diagram of coincidence detector set up

The primary object of the coincidence detector is to determine whether the γ-radiation is in time coincidence with the β-radiation. That is, do the two radiations come close enough together in time to originate from the same nucleus in a sequential or cascade emission. A time coincidence of two radiations from the same nucleus is called a true or real coincidence. It is also possible for radiations from different nuclei to trigger the timing circuits; this produces an unwanted false or accidental coincidence. False coincidences must be subtracted from the total observed coincidence rate, and corrections must be applied for dead time losses in the equipment and for the background contribution to the counting rates.

Both the γ- and the β-detector in Figure 4 are scintillation counters. The absorption of nuclear radiation in a scintillator is converted to light energy through the luminescence process. In the γ-detector, crystalline NaI is used as phosphor. The large Z (53) of iodine gives a high probability for photon absorption. A plastic scintillator (anthracene) is used in the β-detector. Compared to a NaI phosphor, β-rays have a much smaller possibility to scatter out before being stopped in a plastic scintillator. γ-rays interact weakly with the plastic scintillator. In front of the γ-detector there is a thick aluminum window, so no β-particles are detected in that counter. In front of the β-detector there is a thin aluminum window to prevent light to enter the photo-multiplicator. If the scintillator crystal in the β-detector is chosen properly and the high voltage is adjusted, no γ-particles will be counted in the β channel. The mass of the gold foil must be measured before starting. A suitable measuring time is chosen in order to obtain sufficiently small statistical errors. Note the starting time. Also make a background measurement. Assume that the probability that the β-particle is registered in the counter is P_β and that the same is valid for the γ channel with the probability P_γ. Assume that during t_3 the total number of decays is N_{tot}. If P_β and P_γ are independent, the following relations for the number of detected decays are valid:

$$N_\beta = P_\beta N_{tot}$$
$$N_\gamma = P_\gamma N_{tot} \qquad (6)$$
$$N_{\beta\gamma} = P_\beta P_\gamma N_{tot}$$

N_β β counts during time t_3 [counts]

N_γ γ counts during time t_3 [counts]

$N_{\beta\gamma}$ coincidence counts during time t_3 [counts]

By coupling the formulas above one may get rid of the detection probabilities. Hence, the coincidence technique, in principle, affords a method of determining the rate of emission without knowledge of the individual detector efficiencies. The total number of nuclei that decayed during time t_3 can be expressed as:

$$N_{tot} = \frac{N_\beta N_\gamma}{N_{\beta\gamma}} \qquad (7)$$

N_{tot} total number of nuclei that decayed during time t_3 [nuclei]

and the total activity is given by:

(8) $$n_{tot} = \frac{N_{tot}}{t_3}$$

n_{tot} activity in the gold foil [Bq]

A similar expression is used when calculating the activity of the gold foil that was covered with cadmium.

IMPORTANT TO REMEMBER:

Corrections

When measuring the activity of the gold foil the following corrections must be made:

- Subtract the background pulses from n_β, n_γ and $n_{\beta\gamma}$
- False coincidences, due to finite resolving time of the coincidence counter, must be subtracted from the total observed coincidences according to:

(9) $$N_{\beta\gamma(true)} = N_{\beta\gamma} - \frac{2\tau_k N_\beta N_\gamma}{t_3}$$

$$\tau_k = 0.2\ \mu s$$

- Make corrections for the dead time in the detector. Associated with each counting channel is a "dead time" (τ_D), the period after each detection of a pulse during which the channel is inoperative. The loss of counts is expressed by the relation:

(10) $$N_{true} = \frac{t_3 N_{observed}}{t_3 - \tau_D N_{observed}}$$

N_{true} true detected decays [nuclei]
$N_{observed}$ observed decays [nuclei]
τ_D dead time, τ_D=0.2μs (for both detectors)

Simplifications

The following sources of error are likely to be small and are thus neglected:

- The β-counter also counts conversions electrons from the inner photo-effect if the upper voltage is too high.
- About 1 % of the decay is not according to the presumed γ - β cascade.
- P_γ and P_β are not completely independent, there is a small angular correlation between the γ- and the β-particle

DETERMINATION OF THE THERMAL NEUTRON FLUX

The number of nuclei that decayed during time t_3 was calculated using Eq. (5). Hence the rate of decay, or activity, in the gold foil is obtained by dividing Eq. (5) with t_3. The activity in the uncovered gold foil, n_{tot}, and the cadmium-covered gold foil, n_{Cd}, may now be written as:

(11) $$n_{tot}(t_3) = \frac{V_{Au}\Sigma_{ctot}\phi_{tot}}{t_{3Au}\lambda}\left(1-e^{-\lambda t_1}\right)e^{-\lambda t_{2\,Au}}\left(1-e^{-\lambda t_{3\,Au}}\right)$$ *"Uncovered gold foil"*

(12) $$n_{Cd}(t_3) = \frac{V_{Cd}\Sigma_{cCd}\phi_{Cd}}{t_{3Cd}\lambda}\left(1-e^{-\lambda t_1}\right)e^{-\lambda t_{2Cd}}\left(1-e^{-\lambda t_{3Cd}}\right)$$ *"Cadmium covered gold foil"*

n_{tot}	activity in the uncovered gold foil [Bq]
n_{Cd}	acitivty in the cadmium covered gold foil [Bq]
V_{Au}	volume of the uncovered gold foil [cm^3]
V_{Cd}	volume of the cadmium covered gold foil [cm^3]
Σ_{ctot}	effective one-group macroscopic capture cross-section of the uncovered gold foil [cm^{-1}]
Σ_{cCd}	effective one-group macroscopic capture cross-section of the cadmium gold foil [cm^{-1}]
ϕ_{tot}	total neutron flux at the position of irradiation [neutrons/cm^2s]
ϕ_{Cd}	neutron flux in the cadmium gold foil at the position of irradiation [neutrons/cm^2s]
ρ	density of gold = 19.32 [g/cm^3]
λ	$2.978\cdot10^{-6}$ =decay constant of Au198 [s^{-1}]
t_1	irradiation time (same for both Au and Cd) [s]
t_{2Au}	cooling time for the uncovered gold foil [s]
t_{2Cd}	cooling time for the cadmium covered gold foil [s]
t_{3Au}	measuring time for the uncovered gold foil [s]
t_{3Cd}	measuring time for the covered gold foil [s]

Both actvitities, n_{tot} and n_{Cd}, are calculated using Eq. (8). Note that the cooling time and measuring time for the uncovered gold foil and the cadmium-covered gold foil may be different.

The activity, n_{tot}, is the total activity induced by all neutrons. The total neutron flux, ϕ_{tot}, include neutrons of a broad range of energies (2 MeV to 0.025 eV) and the macroscopic capture cross-section of gold is strongly dependent on energy. However we may collapse all neutrons into one energy group and then use an "effective" macroscopic capture cross-section, Σ_{ctot}, for gold. In the one-group approximation, all processes are considered as occurring at one neutron energy, with neutrons of all energies counted in the flux. Unfortunately, to be able to calculate Σ_{ctot} we must know the energy distribution of the total flux, $\phi_{tot}(E)$, which we do not. Similar problem occurs when finding the effective macroscopic capture cross-section of the cadmium covered gold foil, Σ_{cCd}.

Taking the difference between the neutron flux in the uncovered gold foil, and the cadmium-covered gold foil gives the thermal flux, ϕ_{th}:

(13) $$\phi_{th} = \phi_{tot} - \phi_{Cd}$$

It is also true that the capture rate in Au197 in the gold foil, caused by thermal neutrons, is given by:

(14) $$\phi_{th}\Sigma_{cth} = \phi_{tot}\Sigma_{ctot} - \phi_{Cd}\Sigma_{cCd}$$

Σ_{cth} effective thermal macroscopic capture cross-section [cm^{-1}]

ϕ_{th} thermal neutron flux at the position of irradiation [neutrons/cm^2s]

where Σ_{cth} can be calculated using the thermal microscopic capture cross of ^{197}Au, which is found in Figure 1 (a good estimation gives 80 barns). Since we know the capture rates in both gold foils, given in Eq. (11) and (12), we are able to restrict the problem to thermal interactions only and we may calculate the thermal neutron flux, ϕ_{th}, from Eq. (14). This flux corresponds to the thermal flux at the position of the gold foil.

ERROR ESTIMATION

Statistical errors should be calculated. The decay process is strictly random; the probability of a number of disintegrations - and hence the number of counts recorded - in a given time is described by Poisson's law which, for large numbers, approximates to the Gaussian distribution. Thus, the error associated with one single observation of a large number of events is expressed by:

(15) $$\sigma_N = \sqrt{N}$$

where σ is the standard deviation and N is the absolute number of events. The relative error in a measurement becomes:

(16) $$\varepsilon = \frac{\sqrt{N}}{N} = \frac{1}{\sqrt{N}}$$

The standard deviation after subtraction of the background count rate (b) is given by:

(17) $$\sigma = \sqrt{\sigma_n^2 + \sigma_b^2}$$

In order to determine the final statistical error of the thermal flux ϕ_{th}, apply the formula for "propagation of error":

(18) $$\Delta f(x_1, x_2, ...) = \sqrt{\left(\frac{\partial f}{\partial x_1}\Delta x_1\right)^2 + \left(\frac{\partial f}{\partial x_2}\Delta x_2\right)^2 + ...}$$

The relationship between the often used phrase ”confidence limit” and standard deviation is simply the method of quoting the error of a measurement. If the error is quoted as ±σ, the probability that the true value lies within these limits is 0.68; called the 68% confidence limit. For example, limit of error: ±0.675σ, σ, 2σ, 3σ; corresponds to confidence limits of 50%, 68%, 95%, and 99.7% respectively.

DETERMINATION OF THE SOURCE INTENSITY

Since fission neutrons are born at high energy and then moderated to thermal energy it means that the neutrons undergo a very rapid change in energy as they move away from the source. The one-group neutron diffusion equation is not valid under such conditions. Hence, in order to obtain an absolute value of the ^{252}Cf source intensity it is necessary to solve an energy-dependent diffusion equation including energy-dependent nuclear data. Under such circumstances it is difficult to perform any calculations by hand. Instead, a transport calculation using a Monte Carlo method is suitable. This is accomplished in a separate exercise (Lab III).

Monte Carlo Method

The Monte Carlo method is, in contrast to deterministic codes, a stochastic method to simulate particle transports in an arbitrary geometry. While deterministic codes solve the Boltzmann transport equation for an *average* particle, Monte Carlo obtains a solution by simulating *individual* particles and then referring their *average* behaviour. It is particularly useful for complex problems that cannot be modelled by deterministic codes. The problem description is often relatively short an easily constructed. The method consists of following each particle created in a source, throughout its life, from birth to death (absorption, escape etc.), and its interactions with other particles (fission, capture, scattering). The process is run for a large number of source particles to obtain a statistically reliable result and. The program records the average behaviour of the simulated particles. To simulate the particle interaction with the matter data libraries (i.e. JEF2.2, GENDL-VI, ENDF) are used with cross section information for all relevant isotopes.

The basic advantage of Monte Carlo codes over deterministic codes is that they require fewer modelling approximations. In Monte Carlo there are no averaging approximations required in space, energy, and time. A disadvantage of Monte Carlo is that the solution contains statistical errors. All results in Monte Carlo represent estimates with associated uncertainties and the calculations can be rather time consuming as the precision of the results increases.

PREPARATORY EXERCISES

- The gold foils are held in place in the coincidence apparatus by tape. Towards which detector should the tape be facing, γ or β? Why?
- Why are we using the coincidence technique to measure the activity in the gold foils?
- What error will be most significant in the gold foil measurements?

DOCUMENTATION

The laboratory exercise should be documented in a complete report in English. The required contents are the following:

- Abstract – The abstract should summarize your results and methods. It should be short but precise.
- Introduction – Give an introduction to the experiment making it possible for a fellow student who has not performed this laboratory exercise to understand the rest of the report and to relate the result to what she already knew. The last part of the introduction should be written like as a brief introduction to the following parts of the report.
- Theoretical background – Describe the theory behind the experiment. Present the mathematical framework.
- Description of the experimental set-up – Explain how the experiment is set up. Go into details where it is necessary; avoid it where it is not.
- Description of the experimental procedure and performed calculations – First of all measured data from the experiment should be presented. Give the raw data as it is before you perform any calculations. As an example the time for a messurement should be given as 2007-12-04 16.34.50 and 2007-12-04 16.35.10, not as 20 s. Then perform the calculations presenting intermediate results where necessary. Here the 20 s would be presented. Use the layout to present your data and calculations nice and clear. Use tables.
- Results – Present your results.
- Error estimation – Perform an error estimation as described in these instructions.
- Conclusions and discussion – Conclude and discuss your results, why are they important? Are they correct?
- Preparatory exercises – Present the solutions of your preparatory exercises, preferably as an appendix to your report.

REFERENCES

- W. Gudowski, Chapter "*Radioactive decay*" and "*Thermalization and Diffusion*", Lectures in Reactor Physics, CD-ROM, 1998.
- D. J. Bennet, J. R. Thomson, "*The Elements of Nuclear Power*", Longman Scientific & Technical, 3rd edition, 1989.
- G. Choppin, J.O. Liljenzin, J. Rydberg, "*Radiochemistry and Nuclear Chemistry*", 2nd edition, 1995.
- Weston M. Stacey, "*Nuclear Reactor Physics*", Wiley-Interscience, 2001