NEUTRON FLUX MEASUREMENTS IN CROCUS

1. INTRODUCTION

An integral measurement of the neutron flux can be carried out either directly using a neutron detector such as a fission chamber or a BF_3 counter, or indirectly by applying a foil activation technique. In the latter case, neutron capture or another nuclear reaction leads to activation of a specific irradiated sample and, following the irradiation, the resulting radionuclide's activity can be measured using a radiation detector. The induced activity is an integral parameter and depends upon the energy dependence of the neutron flux (i.e., on the neutron spectrum), as also of the given activation cross-section. Effectively, the activity measurement corresponds to the determination of an integral of the given neutron reaction rate over energy and the irradiation time.

Since neutron absorption cross-sections are usually higher at low energies, activation detectors are commonly employed for the measurement of slow neutrons. The neutron mean free path (average neutron interaction distance in a given medium) for such reactions is very small at low energies, so that the material thickness for activation studies needs to be small for avoiding any significant perturbation of the neutron flux at the given location, as well as the self-shielding effect within the sample itself. The most commonly employed types of activation detectors are thin foils or small-diameter wires. If several detectors are irradiated simultaneously, then there must be sufficient spacing between them in order to avoid shadowing effects, i.e., a reduction of the neutron flux incident on a given sample due the presence of other detectors.

Activation measurements can also provide useful information on the neutron energy spectrum. Thus, for example, at a given location in a fast reactor, one could use an appropriate set of threshold reactions (i.e., activation reactions with finite cross-sections only above a certain characteristic, minimal threshold value). By irradiating the corresponding activation detectors at the given location and measuring their activities, integral data are generated which, in turn, can be analysed using a suitable spectrum unfolding algorithm.

1.1 Neutron Activation

When a thin foil is irradiated in a constant neutron flux, the rate of formation of the new radioactive species, in its general form, is given by:

Formation rate = Production rate – Decay rate – Destruction rate by absorption

Mathematically, one may write:

$$\frac{dN_a}{dt} = N_0 \int_0^{E_m} \sigma_a(E)\varphi(E)dE - \lambda N_a - N_a \int_0^{E_m} \sigma_c(E)\varphi(E)dE \qquad (1)$$

where N_0, N_a, and λ denote the number of target atoms in the foil, the number of atoms of the radionuclide and its decay constant, respectively, while σ_a, σ_c and φ stand for the microscopic cross-section for the activation reaction, the neutron cross-section for absorption (usually capture) in the radionuclide and the average neutron flux over the foil surface, respectively. N_0 can be calculated from $N_A m/M$, where N_A, m and M represent Avogadro's number, the mass of the foil and the molecular weight of the foil material. The upper energy integration limit represents the maximum energy E_m, beyond which the neutron flux is zero.

To a first approximation, under low flux conditions, the last term is very small, and hence Equation (1) gets simplified to:

$$\frac{dN_a}{dt} = N_0 \int_0^{E_m} \sigma_a(E)\phi(E)dE - \lambda.N_a = R - \lambda.N_a \tag{2}$$

where R denotes the integral reaction rate in the foil leading to the formation of atoms of the radionuclide. The solution of Equation (2), with the initial condition $N_a = 0$, at time $t = 0$, is given by:

$$N_a(t) = \frac{R}{\lambda}(1 - e^{-\lambda t}) \tag{3}$$

The foil activity, defined as the number of disintegrations per unit time, thus builds up with time and approaches its asymptotic value (the so called saturation value) for infinitely long irradiation duration. The induced activity for a time of irradiation t_0 can thus be written as:

$$A(t_0) = \lambda\, N_a(t_0) = R\left(1 - e^{-\lambda t_0}\right) = A_s(1 - e^{-\lambda t_0}) \tag{4}$$

where A_s (= R) stands for saturation activity, i.e., the maximum activity that can be produced in a given sample for a given flux and activation reaction.

The buildup of activity as a function of time is shown in Fig. 1.

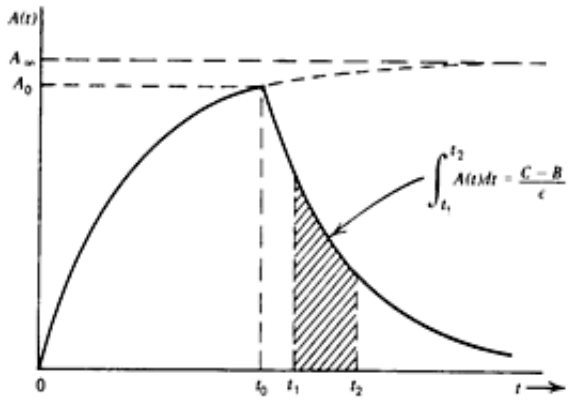

Fig. 1. The build-up of activity during an irradiation of duration t_0, followed by its decay; the shaded area indicates the total activity during a time of measurement between time t_1 and t_2.

In practice, one irradiates samples for a finite time to achieve a measurable activity, taking into consideration the decay time and the neutron flux [1]. The activity A_m, measured between time t_1 and t_2, of a sample irradiated for a duration t_0 (all times are considered from the start of the irradiation, $t=0$) can be written as follows:

$$A_m = A_s(1 - e^{-\lambda t_0}) \int_{t_1}^{t_2} e^{-\lambda(t - t_0)}\, dt = A_s(1 - e^{-\lambda t_0}) e^{\lambda t_0}(e^{-\lambda t_1} - e^{-\lambda t_2})/\lambda \tag{5}$$

or

$$A_m = A_s.(1 - e^{-\lambda t_0}).e^{-\lambda(t_1 - t_0)}.(1 - e^{-\lambda(t_2 - t_1)})/\lambda = A_s.(1 - e^{-\lambda t_0}).e^{-\lambda t_w}.(1 - e^{-\lambda t_c})/\lambda \tag{6}$$

where t_w and t_c represent the waiting time between the end of the irradiation and the start of counting, and the counting duration, respectively.

In case the activity is measured using a gamma detector, and if C and B denote the total counts under the photo peak and the corresponding background counts for the same time of measurement, then the saturation activity can be expressed as:

$$A_S = \frac{A_m \lambda}{(1-e^{-\lambda t_0}) e^{\lambda t_0} (e^{-\lambda t_1} - e^{-\lambda t_2})} = \frac{(C-B).\lambda}{\varepsilon_{de}.\varepsilon_{ep}.\varepsilon_{pm}(1-e^{-\lambda t_0}) e^{-\lambda t_w}(1-e^{-\lambda t_c})} \quad (7)$$

where ε_{ep}, ε_{de} and ε_{pm} denote the photon emission probability of the radionuclide, the photo-peak efficiency of the detector and the photon non-absorption probability, respectively.
With measured values for C and B, and assuming the parameters ε_{de}, ε_{pm} and ε_{ep} to be known, the saturation activity of the irradiated sample can be determined from Equation (7).

1.2 Effective cross-section and integral flux

The present experiment involves the use of thin gold foils (100% ^{197}Au), as activation detectors, for measuring the neutron flux in the CROCUS reactor. In this case, apart from the thermal neutron capture in ^{197}Au (σ_c = 98 barn, at 0.025 eV), a significant activity results from the first capture resonance (σ_c = 3.0E+5 barn, at 1.9 eV). Thus, self-shielding effects due to the finite thickness of the Au foils used can be significant, and an appropriate effective cross-section for the ^{197}Au (n,γ) reaction needs to be estimated, in order that the corresponding integral flux value can be determined from a given measurement of the saturation activity of the produced radionuclide (^{198}Au).

In terms of energy dependent neutron flux and microscopic cross-section, the induced saturation activity per atom of an irradiated foil can be written as:

$$A_s / N_0 = \int_{0.}^{E\max} \varphi(E).\sigma(E).dE = \frac{\int_0^{Emax} \varphi(E).\sigma(E)dE}{\int_0^{Emax} \varphi(E).dE} . \int_0^{Emax} \varphi(E).dE = \bar{\sigma}_{av} . \int_0^{Emax} \varphi(E).dE \quad (8)$$

In the above equation, the left-hand side is an experimentally determined quantity, and one can determine either the integral flux, knowing the average cross-section over the neutron spectrum, or the average cross-section if the integral flux is provided. In the present case, an effective average cross-section for the ^{197}Au (n,γ) reaction has been estimated for the neutron energy spectrum at the measurement positions in CROCUS. This has been done via the BOXER code, modelling of the experimental situation, i.e., for the case of Au foils of 17.8 mm diameter and 10 μm thickness, as to be used currently. The thus estimated average cross-section value is (40.600 ± 1.218) barn.

1.3 Cadmium ratio for a given reaction rate

It is useful to have the neutron flux expressed in two energy groups under consideration of thermal reactor neutronics. From the experimental viewpoint, related information can be generated by using activation detectors in conjunction with a covering material that strongly absorbs thermal neutrons. Cadmium is an ideal material in this context, as indicated in Fig. 2, showing the total neutron cross-section of Au and Cd. (The scattering cross-section being fairly constant and small in each case, the curves essentially represent the absorption, i.e., (n,γ) cross-sections.)

It can be seen that the large thermal capture cross-section of Cd, though close to *1/v* at very low energies, peaks to about 7200 barns at 0.176 eV, before dropping sharply to very low values at higher energies. Clearly then, Cd could be employed for filtering the thermal neutrons from a given neutron spectrum, since these would be strongly absorbed by the Cd and the epithermal neutrons would pass through without significant interaction. Thus, the neutron-induced activity in a material such as Au, with significantly high cross-sections at both thermal and epithermal (i.e., resonance) energies, could be separated into the corresponding components by simultaneously irradiating two types of foils of the material, viz. bare and Cd-covered. The saturation activity of the former foil

type would correspond to the total absorption rate, while that of the latter would represent the epithermal component, i.e., the absorption rate of neutrons above the Cd cutoff.

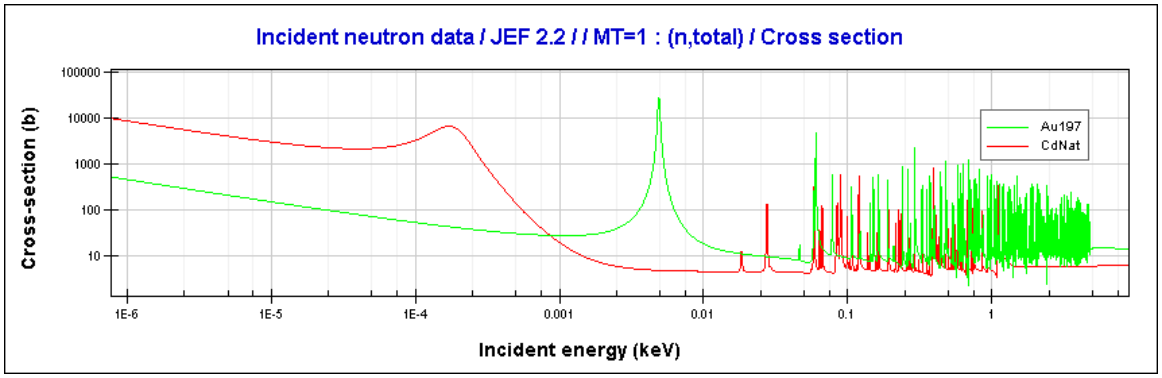

Fig. 2. Total neutron cross-sections of Au and Cd vs. energy.

In accordance with the above, the cadmium ratio of a certain reaction rate in a given neutron spectrum is defined as the saturation activity of a bare foil of the activation material to that of a cadmium-covered foil of the same material, i.e. [2]:

$$R_{Cd} = \frac{A_s^b}{A_s^{Cd}} \tag{9}$$

where the superscripts *b* and *Cd* denote the bare and *Cd*-covered foil, respectively.

The above equation can be written as:

$$R_{Cd} = \left(A_s^{sub-Cd} + A_s^{epi-Cd}\right) / A_s^{epi-Cd}$$

or

$$R_{Cd} - 1 = A_s^{sub-Cd} / A_s^{epi-Cd} \tag{10}$$

where the superscripts *sub-Cd* and *epi-Cd* denote the reaction rate components below and above the Cd cut-off energy, respectively. The Cd ratio minus one may thus be regarded as a measure of the importance of the thermal neutron activation, relative to the epi-thermal.

It may be noted from Fig. 2 that, in the case of the ^{197}Au (n,γ) reaction, the main contribution to the epithermal activation will be that of the large low energy resonance i.e., will occur in the range $0.4 \leq E \leq 2$ *eV*. Clearly, measurements with Cd-covered foils provide information related to the resonance integral of the reaction in question.

1.4 Reactor power calibration

The reactor power can be determined on the basis of the neutron flux measurement at a certain location in the reactor, if an appropriate calibration factor is available for the necessary conversion. The latter can be deduced, for example, via a neutronics modelling of the reactor, in which the flux at the given location is related to the total rate of fissions occurring in the reactor.

In the present experiment, several Au foils are to be irradiated along the central axis of the CROCUS reactor, so that the saturation activity per atom can be determined from the counting of the 411.8 keV ^{198}Au gamma rays using a calibrated HP(Ge) system. By substituting the average cross-section value of (40.600±1.218) barn in Equation (8), one can then obtain values for the

integral flux at the corresponding locations, allowing one to determine the maximum flux in CROCUS. This, in turn, can be converted to the reactor power, using the calibration factor provided.

2. PURPOSE

The purposes of this experiment are:

- Determination of the integral neutron flux.
- Measurement of the Cd ratio from the Au foil saturation activities per atom, with and without Cd cover.
- Plot of the axial flux distribution and use of the maximum value (obtained from a cosine fit) to calculate the reactor power by using the reference calibration factor (3.95E-8 W/cm^2s), [3].

3. SAFETY MEASURES

In order to work under the standard regulations for control of radiation exposure the safety measures during the experiment are:

- Mass determination of the foils considering the activity after the irradiation.
- Personal dosimeter for each participant of the experiment (teachers, students and reactor operators) provided by the radiation safety group.
- Use tweezers for the preparation of the foils and manipulation in the laboratory.
- Use gloves while handling the Cd covers in the laboratory.
- For fixing the Au foils in position, do not apply scotch tape directly on them. Instead, the Au foils should first be covered with a thin plastic foil (use the provided machine) and then with scotch tape. An additional, long piece of scotch tape covering all the foils is used to block their positions. Verify that the Au foils cannot move.
- Use lab coat, gloves and tweezers for the manipulation of the irradiated foils in the control area.
- Each participant of the experiment should check for possible contamination using a hand monitor.
- The foils are only handled after a proper decay time and in the presence of a radiation safety inspector.
- The detector-to-foil distance, i.e. the counting geometry, should be held fixed throughout the measurements.
- While not measured, the foils are kept under biological shielding.
- At the end, place the used foils in a plastic box, indicating the date on the cover.
- Drinking and eating is not allowed in the control area.

4. INSTRUMENTS AND MATERIALS

This experiment measures the axial distribution of thermal neutron flux and the Cd-ratio, therefore is done in the core of a nuclear reactor. It needs the following:

- The CROCUS reactor, with an operating power of 10 or 20 W for this experiment.
- 5 thin Au activation foils (10 micron thickness and 17.8 mm diameter)
- Cylindrical Cd cover boxes.
- Plastic bar sample holder to attach the foils during the irradiation.
- HPGe detector and gamma spectrometer.
- Calibrated ^{152}Eu gamma source.
- Scotch tape scissors, tweezers and gloves.

5. EXPERIMENTAL PROCEDURES

- Clean the plastic bar from glue and adhesive tape residuals using alcohol and paper.
- Check the five gold sheets. If there are too damaged, use new sheets.
- Calculate the position of the gold sheets along the bar.
- Weight the sheets with precision scale.
- Mark each sheet with a number.
- Fix the gold sheets to the bar in the correct position by covering them with a thin transparent plastic sheet and by fixing it to the bar using adhesive tape. The plastic sheet avoids the contact between the gold and the tape. The gold sheets are very thin and may break up if somebody tries to detach them from the tape.
- Ensure that the gold sheets are well fixed and do not move along the bar.
- Add a long adhesive tape all over the plastic bar to ensure that the gold sheets will stay fixed.
- Irradiation of the Au foils, with and without the Cd cover, is carried out at the pre-selected positions along the central axis (operator), see Fig. 3.
- Operating power of the reactor is 10 or 20 W, and the irradiation time of the activation foils is 1 hour (operator).
- Perform energy and efficiency calibration of the HP(Ge) system with a reference ^{152}Eu gamma source during the irradiation time.
- Switch on all the units of the HP(Ge) system DSA-1000, including PC, apply the specified high voltage and set the appropriate amplifier gains, etc.
- Place the ^{152}Eu source at a fixed distance from the top surface of the detector containment and perform a spectrum acquisition with the multi-channel analyser (DSA-1000) for energy and efficiency calibration. Verify that the acquired ^{152}Eu spectrum is correct.
- The reactor is shutdown after the one hour of the gold foils irradiation (operator).
- The plastic bar is removed from the reactor core and transferred to the HP(Ge) laboratory in the control area.
- Before detaching the sheets, measure their position again (they could have moved during the insertion into the reactor core).
- Measure the activity of each gold sheets (duration: 20 minutes each sample) and calculate the flux at each point.
- Fit the flux points with a cosine curve (cosine function is the axial solution of the diffusion equation in the cylindrical coordinates).
- Find the extrapolated distances and compare them with the exact dimensions of the reactor core.

- Use the maximum value of the neutron flux (obtained from a cosine fit) to calculate the reactor power by using the reference calibration factor, [3].

Fig.4. Insertion of the gold foils' support in the centre of the CROCUS reactor core.

6. MAIN PARAMETERS MEASURED

In order to obtain the gamma spectrometer absolute efficiency curve, it is necessary to collect the following data:

- Calibrated source activity at the moment of measurement, obtained from the initial activity value and the period of time between the radioactive source calibration and measurement.
- Live measured time.
- Full energy peak counts at the energies of interest.

Related to the reaction rate calculations, it is required to collect the following data:

- Activation detector masses.
- Irradiation times: start and shutdown. If it is possible, the power history during the irradiation.
- Live measured times.
- Decay time, between the end of the irradiation and the beginning of measurements.
- Full energy peak counts at every energy of interest. In this case: 411.8 keV.

7. TYPICAL REAL DATA COLLECTED

Examples of the typical real data collected:

Table 1 4π rates of emission of the ^{152}Eu source, as given by the manufacturer, for the different energies considered for efficiency calibration. The source activity was 45.4 kBq on 13.01.1999.

Gamma Energy [keV]	4π Emission/s (1 σ % error)
121.78	1.289E+4±1.5
344.28	1.204E+4±1.7
778.90	5.875E+3±1.5
963.44	6.626E+3±1.5
1408.23	9.443E+3±1.5

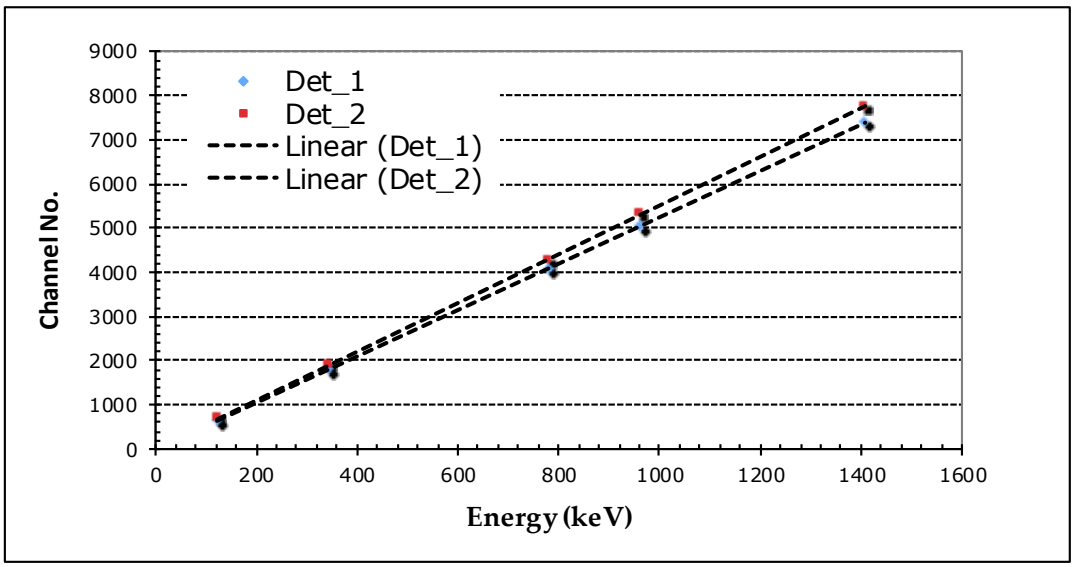

Fig. 5. Energy vs. channel number for the two HP(Ge) detectors.

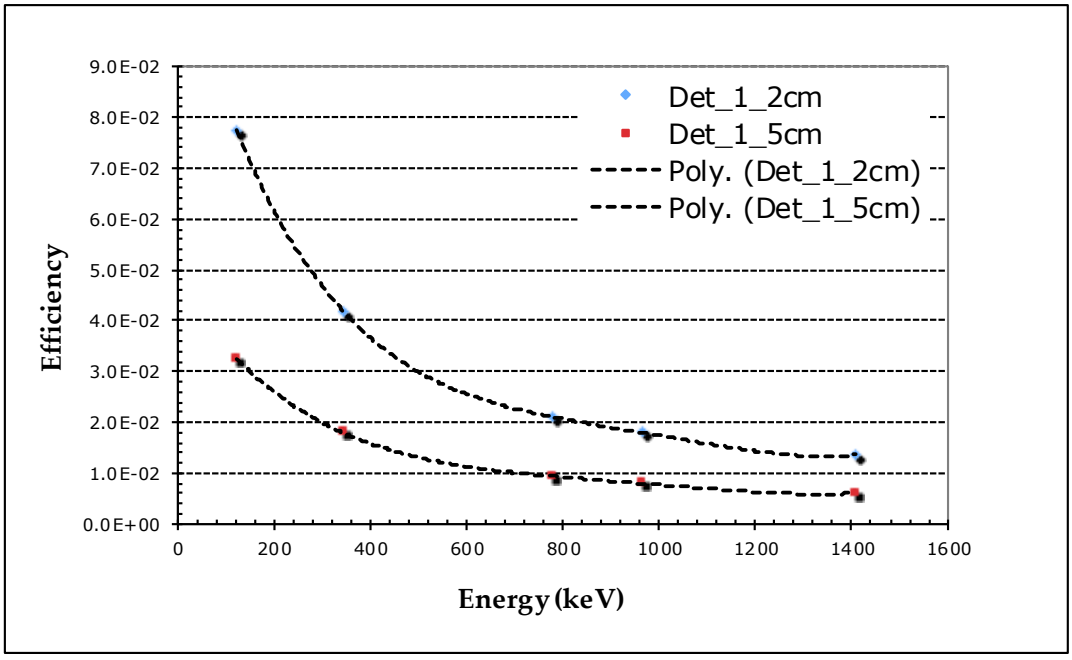

Fig. 6. Efficiency vs. energy for DET#01, at 2 and 5 cm distance from the detector surface.

Table 2. Weight and position of each Au foil (the measurement accuracy of the distances is 0.5 cm).

Foil	Foil Material	Weight [g]	Distance from central foil [cm]
1	^{197}Au	0.0344	30.00
2	^{197}Au	0.0525	20.00
3	^{197}Au	0.0475	10.00
4	^{197}Au	0.0343	0.00
5	^{197}Au	0.0360	-10.00
6	^{197}Au covered with Cd	0.0326	-20.00

Table 3. Saturation activity

Foil	Area [counts]	Area Error [counts]	Waiting Time t_w [s]	Counting Duration t_c [s]	Saturation Activity A_s [MBq]
1	102000	321.78	1930	895.40	1.406±0.004
2	195000	443.20	2991	892.00	2.707±0.006
3	204000	454.51	5004	900.00	2.823±0.006
4	161000	403.23	6001	900.00	2.235±0.006
5	160000	402.53	3993	900.00	2.208±0.005
6	60000	246.28	718	896.94	0.823±0.003

Table 4. Neutron integral flux

Foil	Saturation Activity A_s [MBq]	N_0	Integral Flux ϕ [$cm^{-2}s^{-1}$]
1	1.406±0.004	1.02E+20	$(3.392\pm0.102)\times10^{8}$
2	2.707±0.006	1.60E+20	$(4.154\pm0.125)\times10^{8}$
3	2.823±0.006	1.45E+20	$(4.789\pm0.144)\times10^{8}$
4	2.235±0.006	1.05E+20	$(5.250\pm0.158)\times10^{8}$
5	2.208±0.005	1.10E+20	$(4.941\pm0.149)\times10^{8}$
6	0.823±0.003	9.97E+19	$(2.033\pm0.062)\times10^{8}$

8. DATA ANALYSIS, ASSUMPTIONS AND EQUATIONS

- The reference calibration factor, relating the maximum value of the integral flux in CROCUS to the reactor power, is 3.95E-8 W/cm^{2}s^1 [3].

- As mentioned, the cadmium ratio is to be determined via measurements at the two positions +20 cm (bare Au foil) and -20 cm (Cd-covered foil). It is known, from separate measurements, that there is a certain axial flux asymmetry in CROCUS, the ratio of the flux at -20 cm to that at +20 cm being 1.048. This needs to be applied as a correction factor.

- The values for gamma emission probability and photon non-absorption probability in the foil are as follows [3]:

  $\varepsilon_{ep} = 0.954$
  $\varepsilon_{np} = 0.988$

- Nuclear data of ^{197}Au are presented in Table 5.

Table 2. Nuclear data of ^{197}Au.

Isotope of Interest	^{197}Au	Reference
M_{mol} of element	196.967	[4]
Isotopic abundance	1.000	[4]
Reaction of interest	^{197}Au(n,γ)^{198}Au	-
σ_a^{197} [barn]	40.600±1.218	[3]
Gamma emitted [KeV]	411.804	[5]
λ [1/s]	2.976E-6	[4]
Yield	0.959	[5]

- It is also important to consider other parameters contributing to error apart from the counting statistics. More often, the data obtained need to be processed by additional steps such as addition, subtraction, multiplication and other functional manipulations, in order to arrive at the specific parameter of interest. The numerical values arising from these steps will be distributed in a way that is dependent on both the original distribution and the types of operations carried out.

- The standard deviation on a quantity u(x, y, z, …) can be derived from the standard deviation of each independent variable denoted by $\sigma_x, \sigma_y, \sigma_z ....$, using the standard error propagation formula given by:

$$\sigma_u^2 = \left(\frac{\partial u}{\partial x}\right)^2 \sigma_x^2 + \left(\frac{\partial u}{\partial y}\right)^2 \sigma_y^2 + \left(\frac{\partial u}{\partial z}\right)^2 \sigma_z^2 + ...$$

9. PRE-KNOWLEDGE REQUIRED FROM STUDENTS

The students should be familiar with following contents:

- Nuclear measurements: different types of photon detectors, basic experimental setup, activated foils handling.
- Nuclear physics: radioactive decay, neutron induced reactions (particularly neutron activation).
- Reactor physics: neutron flux in a reactor, spatial and energetic distribution, control rod perturbations.
- Radiation protection: dose rates, limits and all aspects regarding radiological protection, necessary to perform the experience under high security standards.

10.RESULTS

Saturation activity and integral neutron flux were calculated according to Equation 7 and 8, respectively. Results are summarized in Tables 6.

Table 6. Saturation activity and neutron integral flux

Foil	Distance from Central Foil [cm]	Saturation Activity A_s/N_0 [Bq/atoms]	Integral Flux ϕ [$cm^{-2}s^{-1}$]
1 ^{197}Au	+20.00	$(1.860\pm0.004)\times10^{-14}$	$(4.59\pm0.138)\times10^{8}$
2 ^{197}Au	+10.00	$(2.110\pm0.005)\times10^{-14}$	$(5.18\pm0.155)\times10^{8}$
3 ^{197}Au	0.00	$(2.270\pm0.004)\times10^{-14}$	$(5.59\pm0.168)\times10^{8}$
4 ^{197}Au	-10.00	$(2.130\pm0.005)\times10^{-14}$	$(5.25\pm0.158)\times10^{8}$
5* $^{197}Au^{Cd}$	-20.00	$(0.862\pm0.003)\times10^{-14}$	$(2.12\pm0.064)\times10^{8}$

* the gold foil No. 5 was placed in the Cd cover.

The normalized axial flux distribution is plotted in the Fig. 7. The value of the Cd-covered foil at the axial position of -20.00 cm was replaced by theoretically calculated value using the correction factor of 1.048 as mentioned in section 8 of this protocol.

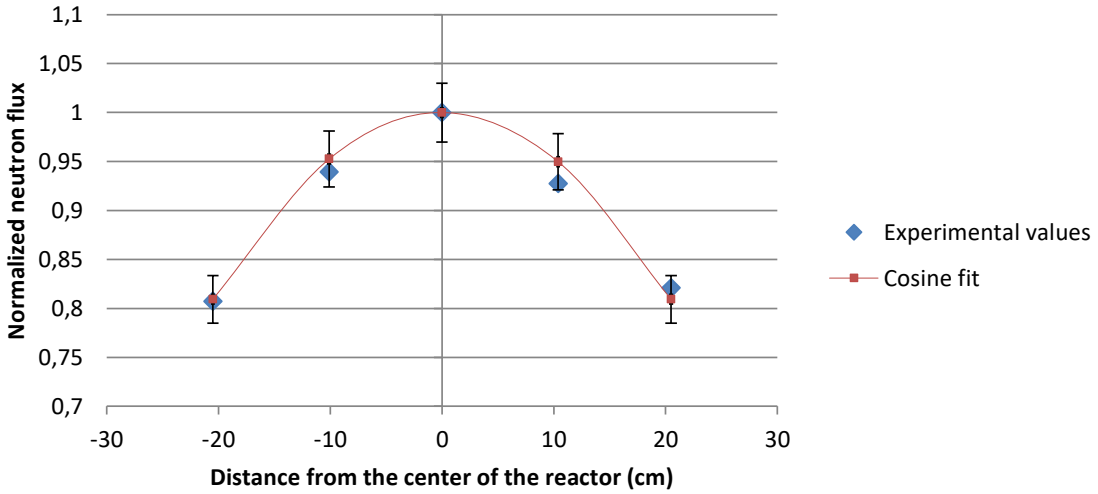

Fig. 7. Axial flux: experimental and cosine fit values

The cosine fit was done by the method of the least squares and yields excellent result: the value of the coefficient of determination R^2 is 0.9992. The maximum value of the flux was determined to be $(5.590\pm0.168)\times10^{8}$ $cm^{-2}s^{-1}$, which corresponds to a power of (22.10±0.66) W using the reference calibration factor. The operating power of the reactor was (20.50±1.00) W. Thus, obtained results are in the span of uncertainty.

Now we compare the foils placed at positions -20 cm and +20 cm. The axial distribution of the flux is almost symmetric (cf. above), thus the foils should be activated by the same flux. In fact, as it was mentioned previously, there is a slight asymmetry due to the tank of water below CROCUS. Indeed, we have to apply a correction factor of 1.048 before comparing the activity of the two foils so as to remove this effect.

From Equation (7), the cadmium ratio is R_{Cd}=(2.260±0.068) and R_{Cd}−1=(1.260±0.068), which means that thermal neutrons and epithermal–fast neutrons have approximately the same contribution to the activation of the gold foils. The term R_{Cd}−1 as described in Equation (8), represents the importance of the thermal neutron activation relative to the epithermal−fast. Therefore, the thermal neutrons contribute to 56%, against 44% of the epithermal−fast ones.

11.CONCLUSIONS

The basic techniques of the neutron activation method were tested experimentally and a rudimentary description of the neutron spectrum in the core using gold foils with and without a Cd encasing. Five different axial positions where used in the central axis of the reactor core. The saturation activities of the gold foils were measured with a HP(Ge) detector and the integral neutron flux and core power were derived. The uncertainties in these measurements are around 3%. When using the peak axial neutron flux to calculate the core power, a value of (22.10±0.66) W was obtained. In addition, the Cd ratio was calculated and found to be R_{Cd}=(2.260±0.068) when using a Cd-covered gold foil.

12.REFERENCES

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