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Measurement of thermal neutron flux and cadmium ratios using neutron activation analysis

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ABSTRACT

Irradiation of known foil samples in a reactor and subsequent measurement of induced activity allows determination of neutron flux. This is an important information for many purposes. It can be used reciprocally to determine contents of unknown samples irradiated at the same position in the reactor. On the other hand, neutron flux information is used in the assessment of a safe operation of nuclear power reactors. In the frame of this work, bare and cadmium covered foils of gold and dysprosium have been irradiated at different positions of the reactor. Induced activity measurements were made using a standard procedure. Thermal neutron flux and cadmium ratio values were determined for the irradiation positions.

1. Introduction

A description of the principles of Neutron Activation Analysis (NAA) along with a review of advantages, limitations and applications is given by (Win, 2004). Use of NAA for many purposes have been reported. Including identification chemical warfare agents (Heller, *et. al.* 2001), trace elements in 52 Chinese medicines (Lin *et. al.*, 2003) and characterization of Micronesian ceramics (Descantes, 2001).

NAA is a powerful tool for quantitative analysis of trace elements in a sample up to parts per billion (Radiation Center, 2003). When NAA is used to determine the content of unknown sample the method requires irradiating the sample in a reactor and subsequent measurement of induced activity. Measured activities and neutron flux values permit determination of an unknown sample content.

In measuring thermal neutron flux (TNF) we follow reverse procedures. Irradiating a known sample and measuring the induced activity, which allows calculation of the flux. Obtaining information on the neutron flux by NAA is routinely used in nuclear power reactors vessel surveillance programs. This information is very important in the analysis needed to ensure safe operation of reactors.

Cadmium covered foils irradiated by neutrons will not respond to thermal neutrons since they are absorbed by the cadmium. Bare and cadmium covered identical foils irradiated at the same position can be used to measure the contribution of thermal neutron flux only.

2. Principle of measurement

The measured activity of foil detector irradiated for time τ and measured at time t after the end of irradiation is given by (Nuclear Science Division, 2000).

$$A(t) = \eta\gamma_a\phi_{th}\sigma_{act}N_T\left(1 - e^{-\lambda\tau}\right)e^{-\lambda t} \quad (1)$$

Here:
η- Efficient of the measuring system.
ɣa - ɣ ray abundance.
ϕ_{th}-Thermal neutron flux (neutrons. cm^{-2}. s^{-1}).
σ_{act}-Microscopic activation cross section (cm^2).
N_T-Number of target atoms in the sample.
λ-Decay constant (s^{-1}).

These relationships will, however, be accurate only if each atom of the sample is irradiated by the same neutron flux. In case of a foil of non-negligible thickness, the average neutron flux will be lower inside it (ϕ_{th}^{x}) than on the surface (ϕ_{th}^{0}) due to neutron absorption. Their ratio is the self-shielding factor:

$$G_{th} = \frac{G_{th}^{x}}{G_{th}^{0}} \quad (2)$$

We should account for self-shielding when this effect exists. The self-shielding problem can be solved experimentally by using a diluted alloy of the detector material. For the case of no self-shielding (surface) and the case when self-shielding exists (inside) we have at a certain time from Eq. 1:

$$A_{th}^{0} \propto \phi_{th}^{0} \;\; and \;\; A_{th}^{x} \propto \phi_{th}^{x}$$

Using Eq. 2

$$A_{th}^{x} \propto \phi_{th}^{0} \cdot G_{th}$$

Then we have

$$G_{th} = \frac{A_{th}^{x}}{A_{th}^{0}} \quad (3)$$

3. Determination of TNF from the activity of an irradiated foil detector.

The activation cross section values (6o) available in the literature (Herman, 2011) are for temperature T_0=293 K. In the present case, we are going to determine the thermal neutron flux belonging to the average velocity of the Maxwellian distribution. Therefore, the cross section value has to be converted taken into account the real neutron Temperature (Tn) we get:

$$\sigma_{act}(T_n) = \sigma_0\sqrt{(\pi/2)(T_0/T_n)} \quad (4)$$

To determine thermal flux the activity induced by thermal neutrons only should be taken into consideration. However, when irradiating a detector at a position where a significant epithermal flux is present, part of the induced activity is due to the epithermal neutrons. Therefore, a correction has to be applied to determine the contribution of epithermal neutron flux. This is possible because the neutron absorption cross-section of cadmium is very large at thermal energies (Fig. 1), while negligible in the epithermal range.

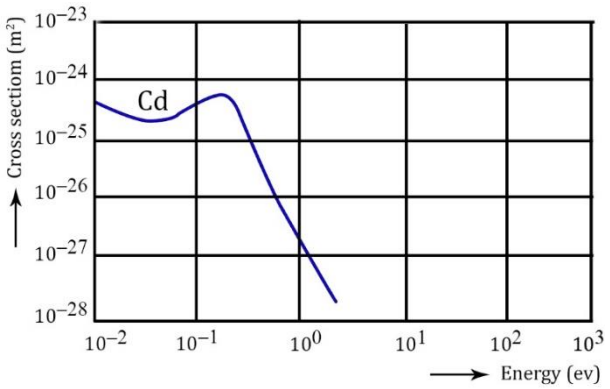


Fig. 1. Thermal neutrons activation cross-section of cadmium

Hence, if the detector is covered with cadmium of appropriate thickness, thermal neutrons will be practically fully absorbed by the cadmium as a result activity of the foil detector will be induced only by epithermal neutrons of energies above about 0.4 to 0.7 eV, depending on the thickness of the cadmium cover. Activity-induced by thermal neutrons is simply determined by irradiating two identical detectors, one bare and the other covered with cadmium:

$$A_{th} - A_b - A_{cd} \tag{5}$$

A_{th}–Activity-induced by thermal neutrons.
A_b-Activity of bare foil detector (A_{th}+A_{epi}).
A_{cd}-Activity of cadmium-covered foil detector (A_{epi}).

Using a bare and a cadmium covered identical foil detectors, the thermal neutron flux is calculated from Eq. 1 as follows (here we include Gth to account for self-shielding and Tn correction):

$$\phi_{th} = C\frac{e^{\lambda t}}{1-e^{-\lambda\tau}}\left(A_b(t) - A_{cd}(t)\right) \tag{6}$$

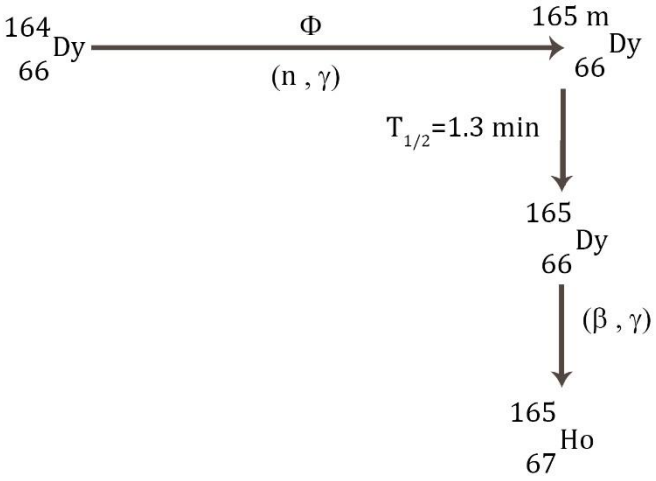
where

$$C = \sqrt{\frac{T_n}{\pi t_0}}\frac{2}{\gamma_a \eta N_T \sigma_0 G_{th}}$$

In case of detector material with activation cross section following the (1/v) law throughout the neutron energy range, the value of A_{th} is very high and thus, the contribution of epithermal neutrons to the induced activity may be neglected. Dy foils detectors were used, the following series of reactions will take place in Dy when irradiated with neutrons (Scheme 2).

Fig. 2 shows thermal neutron activation cross section curve for Dy, which is (1/v) type. No need for cadmium cover when measuring TNF using Dy foils. A gold foil detector was used in the determination of the absolute value of thermal neutron flux. The following series of reactions will take place in gold irradiated by neutrons (Scheme 1).

Of this multistep reaction series, actually, it is sufficient to take the first one only into consideration. The activation of Au-198 by neutron capture can be neglected due to experimental conditions (ϕ_{th}, σ_{act}, irradiation time, etc.). In the range of thermal neutron energies, the activation cross-section of Au-197 follows the (1/v) law but it has a number of resonance peaks in the resonance region (Fig. 3), stressing the role of cadmium cover.



Scheme. 1. Dy reaction with neutron

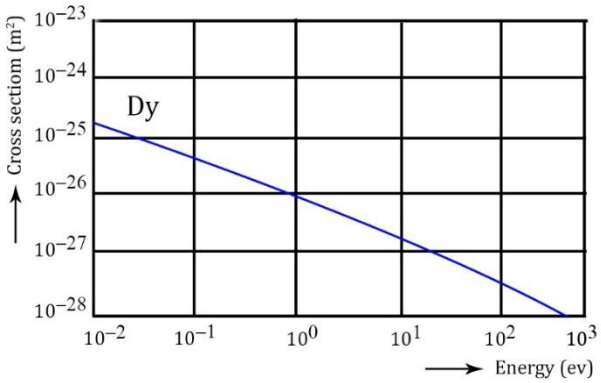
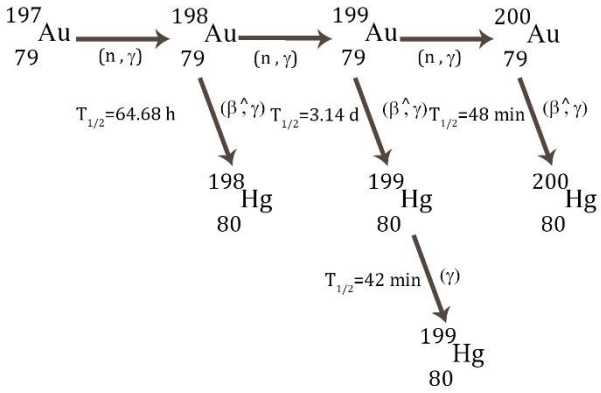


Fig. 2: Thermal neutrons activation cross-section of Dy



Scheme 2. Au reaction with neutron

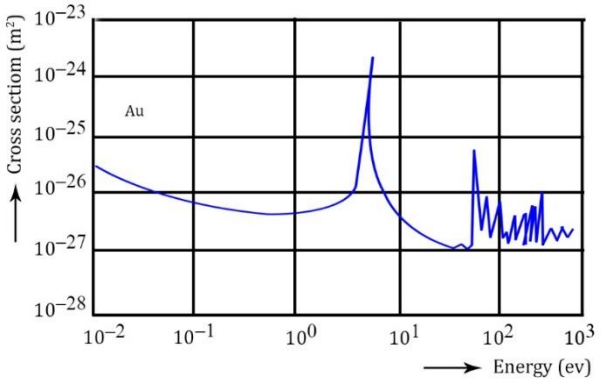


Fig. 3. Thermal neutrons activation cross-section of Au

According to (Marshall, 2014) the cadmium ratio (CR) is defined as:

$$CR = \frac{A_{bare}}{A_{Cd}} = \frac{A_{th} + A_{epi}}{A_{epi}} = 1 + \frac{A_{th}}{A_{epi}} \quad (7)$$

Here:
A_{epi}–Activity-induced by epithermal neutrons

4. Irradiation and induced activity measurement

Foil irradiation was conducted at the Budapest Technical University (BTU) research reactor. Table 1 gives a complete description of the reactor. The pneumatic transfer system allows insertion and subsequent withdrawal of samples automatically a schematic diagram of the reactor is shown in Fig. 4.

Table 1

Characteristics of BTU research reactor.

PARAMETER	DESCRIPTION
Type of fuel element	EK-10 type of fuel bundles each contains 16 rod-shaped fuel elements. The active diameter of each fuel element is: about 7 mm inactive length: About 50 cm, the cladding is aluminum of a wall thickness of 1.5 mm and 10 mm outer diameter.
Type of fuel	A 10% enriched in U-235 (UO_2) content being 8 g per fuel element and 128 g per bundle
Number of fuel element bundles and fuel rods	21 complete bundles, 3 partially loaded bundles, and 369 fuel elements.
Critical mass of active core	Cold: 2740 g or ^{235}U total: 2952 g of ^{235}U (excess for poisoning)
Moderator	Desalted water (H2O)
Coolant	Desalted water (H2O)
Reflector	Side wards: graphite + desalted water (H2O). Lower & upper: desalted water (H_2O)
Number of vertical irradiation channels	One position in the active core (F3), and 18 positions in the reflector with no connection to the pneumatic transfer system. One position in the active core (D5) and other two positions in the reflector (G5) all are connected to the Pneumatic transfer system.
Max. Power	100 kW

The foils were prepared in the form of thin circular shape. Foils irradiated at the same position situated together in a polythene holder suitable for the pneumatic system. Ten activation detectors in the form of thin foils have been activated in different positions as follows:

Two pairs of cadmium covered (Cd) and bare (b) Dy-Al alloy foils were situated at positions D5 (core) and G5 (reflector). The same has been done with Au-Al alloy foils. In addition, a bare and a cadmium covered pure Au foils have been irradiated at the core position D5.

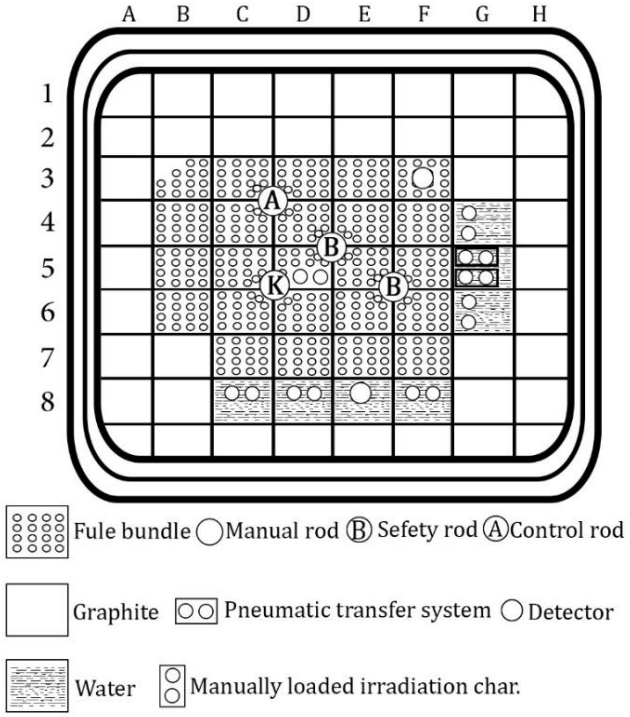


Fig. 4: BTU research reactor core (top view)

The foils in polythene sample holder are forwarded to the operating reactor by means of the pneumatic system joining the irradiation channel. These ten foils were then used for absolute measurement of the TNF and the investigation of the cadmium ratio. Irradiation data are placed in Table. 2. Irradiated foils intensity measurement and subsequent analysis were made using instrumental setup and computer programs available for routine NAA.

Table 2.

Irradiation data.

Sr no	Detector material	Det. type b: bare Cd: cadmium covered	Position within reactor	Reactor power (kW)	Irradiation time (sec)
1	Dysprosium (Dy-Al alloy)	b.foil	D5	1.0	30
2	Dysprosium (Dy-Al alloy)	Cd.foil	D5	1.0	30
3	Dysprosium (Dy-Al alloy)	b.foil	G5	10.0	10
4	Dysprosium (Dy-Al alloy)	Cd.foil	G5	10.0	10
5	Gold (pure)	b.foil	D5	1.0	60
6	Gold (pure)	Cd.foil	D5	1.0	60
7	Gold (Au-Al alloy)	b.foil	D5	10.0	1000
8	Gold (Au-Al alloy)	Cd.foil	D5	10.0	1000
9	Gold (Au-Al alloy)	b.foil	G5	10.0	1000
10	Gold (Au-Al alloy)	Cd.foil	G5	10.0	1000

Gamma spectrometry is a standard procedure that needs no details since it is not the subject of this work. Further-more uncertainties (mainly due to nuclear data) have been taken into account to estimate the accuracy of results.

5. Results and discussion

The induced activity values of pure and Al alloyed gold foils are used to evaluate the thermal self-shielding factor using Eq. 3, we can write:

$$G_{th} = \frac{A_b^x - A_{cd}^x}{A_b^0 - A_{cd}^0} \quad (8)$$

In this equation. measured activity values for bare and cadmium covered pure gold foils represent A_b^x and A_{cd}^x, on the other hand corresponding values for gold-AL alloy represents A_b^0 and A_{cd}^0 . A value of 0.93∓0.02 for thermal neutron self-shielding factor was obtained. Value of G_{th} is used in the calculation of the TNF. The results of pure gold foils are used to determine the absolute TNF at core position D5 using Eq. 6. A value of 2.73×10^{14} neutrons m^{-2} s^{-1} was obtained from this experiment at 1.0 kW reactor power. This value about 5% lower than the value obtained by calculation (Tashani, 2010). This deviation is found due to the (TNF) depression caused by control rods and it is changing with different rods positions. In the present case (irradiation position D5) the manual rod (K) has the dominating effect.

The results of bare and cadmium covered AL alloyed foils were used in CR calculations using Eq. 7. The results at different core positions for Dy and Au are shown in Table 3. The dependence of the CR on the detector cross-section can be seen when considering the corresponding values obtained for Dy and Au foils irradiated at the same position.

Table 3

Cadmium ratio results

Material	Core position	Value of cadmium ratio
Dy (alloy)	D5	75.0 ∓ 0.83
Dy (alloy)	G5	150.0 ∓ 1.41
Gold (alloy)	D5	1.65 ∓ 0.02
Gold (alloy)	G5	2.67 ∓ 0.03

For Dy a large value of CR was obtained due to its (1/v) cross section. As a result, one may use Dy foils for thermal neutron flux measurement without Cadmium cover. On the other hand, the epithermal contribution to the activity of the irradiated Au foil is high (60–40 %) due to high resonance cross section of gold at 4.9 eV.

The spectrum effect on the (CR) values for the same foil material is also seen on Table 4. For Au and Dy the (CR) values obtained at position G5 are higher than those obtained at position D5. Position D5 is within the active core where the spectrum is harder (high epithermal flux) than in position G5 which is in the reflector.

6. Conclusion

The main goals of this work have been achieved. The absolute thermal neutron flux value have been determined at D5 position of the core. This result can be used for future tasks (i.e. NAA). Moreover, the (CR) values at positions D5 and G5 for gold and dysprosium are now available. These values can be used in any subsequent calculations involving thermal neutron flux.

Finally, I like to emphasize that results verified different assumptions, approximations and precautions made. Moreover, this work was fruitful in experience gained.

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